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Review

The Role of Biocomposites and Nanocomposites in Eliminating Organic Contaminants from Effluents

Abdulilah Mohammad Mayet [1], Mohammad Hijji [2,*], Ebraheem Abdu Musad Saleh [3], Arif Reza [4,5,6], Sokaina Issa Kadhim [7], Sherzod Shukhratovich Abdullaev [8,9,10], Ali Alsalamy [11], Zahraa F. Hassan [12], Cristian Vacacela Gomez [13] and Talia Tene [14]

[1] Electrical Engineering Department, College of Engineering, King Khalid University, Abha 61411, Saudi Arabia; amayet@kku.edu.sa
[2] Faculty of Computers and Information Technology, University of Tabuk, Tabuk 71491, Saudi Arabia
[3] Department of Chemistry, College of Arts and Science, Prince Sattam Bin Abdulaziz University, Wadi Al-Dawasir 11991, Saudi Arabia; e.saleh@psau.edu.sa
[4] Department of Environmental Science, College of Agricultural Sciences, IUBAT—International University of Business Agriculture and Technology, Dhaka 1230, Bangladesh; arif.reza@stonybrook.edu
[5] New York State Center for Clean Water Technology, Stony Brook University, Stony Brook, NY 11794-5000, USA
[6] School of Marine and Atmospheric Sciences, Stony Brook University, Stony Brook, NY 11794-5000, USA
[7] Buliding and Construction Technical Engineering Department, College of Technical Engineering, The Islamic University, Najaf 54001, Iraq; sokaina.issa@iunajaf.edu.iq
[8] Faculty of Chemical Engineering, New Uzbekistan University, Tashkent 100007, Uzbekistan; sherzodbek.abdullaev.1001@gmail.com
[9] Scientific and Innovation Department, Tashkent State Pedagogical University Named after Nizami, Tashkent 100007, Uzbekistan
[10] Editory LLC, Tashkent 100007, Uzbekistan
[11] College of Technical Engineering, Imam Ja'afar Al-Sadiq University, Al-Muthanna 66002, Iraq; alihashimalsalamy78@gmail.com
[12] College of Dentistry, Al-Ayen University, Nasiriyah 64001, Iraq; zahraa.hassan@alayen.edu.iq
[13] Department of Physics, Escuela Superior Politécnica de Chimborazo (ESPOCH), Riobamba 060155, Ecuador; cristianisaac.vacacelagomez@fis.unical.it
[14] Department of Chemistry, Universidad Técnica Particular de Loja, Loja 110160, Ecuador; tbtene@utpl.edu.ec
* Correspondence: m.hijji@ut.edu.sa

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Abstract: Many contaminants such as heavy metals, dyes and hydrocarbon compounds are annually generated by industrial activities and discharged into water sources. They have destructive impacts on the environment. To eliminate these toxic contaminants from aqueous media, sorption can be considered a proper and efficient process, as it has indicated high efficiency for removing organic contaminants. This paper entails a comprehensive evaluation of different organic pollutants, pros and cons of the sorption process in removing these pollutants, comparing different types of adsorbents and investigating the recyclability of different types of composites. Moreover, the sorption mechanism and kinetic behavior of organic contaminants are investigated using biocomposites and nanocomposites. Moreover, the future perspectives of biocomposites and nanocomposites in eliminating organic compounds are discussed. Based on investigations, a large number of nanocomposites have been utilized for removing organic contaminants with high performance so far, including SiO_2/$MnFe_2O_4$/ZIF-8 MOF, magnetite/MWCNTs, nano zerovalent iron, barium/Cobalt/polyethylene glycol (PEG), graphene oxide/Fe_3O_4, kaolin/$CuFe_2O_4$ and Fe_3O_4@UiO-66. Moreover, polyaniline/sodium alginate/*Oscillatoria* filamentous cyanobacterium biomass, cow bones/zeolite/coconut biocomposite and AC derived from melamine and sucrose are excellent biocomposites in the removal of organic pollutants. Moreover, the reusability study shows that poly(N-isopropylacrylamide)/Fe_3O_4 and corn stalk biomass/chitosan/Fe_3O_4 biocomposites have significant stability compared to other composites in the elimination of organic pollutants.

Keywords: biocomposites; nanocomposites; effluent; mechanism; organic contaminants; reusability

1. Introduction

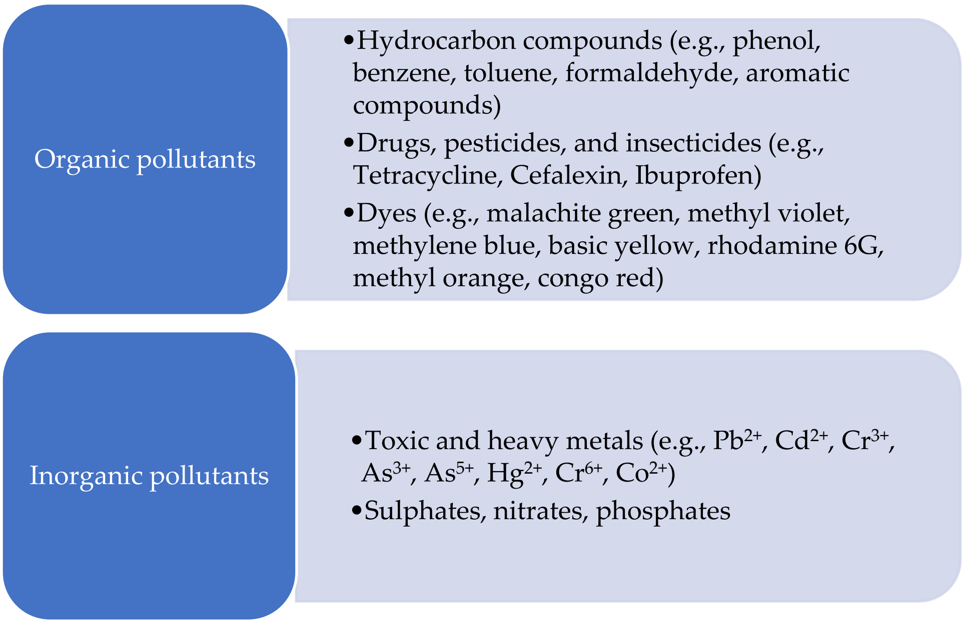
Water is a crucial natural source for human survival. With the increase in the world's population and the increase in the volume of contaminated water, access to a clean water source is of great importance [1]. There are various kinds of contaminants in effluents, including toxic ions, heavy metals, pharmaceuticals, organic dyes, pesticides and hydrocarbon compounds [2–4]. The presence of these pollutants, especially organic materials, has a detrimental influence on water quality. Eliminating these toxic ions from sewage, hence, is one of the main goals of various refineries. So far, many processes have been utilized for effluent treatment, including electrolysis, distillation, electrodialysis, gravity, solvent extraction, flocculation, ion exchange, microfiltration, reverse osmosis, sedimentation, flotation, oxidation, catalytic procedure and sorption. Among these processes, the sorption process is an efficient technique for wastewater treatment, owing to many benefits such as an abundance of adsorbents, high performance, simple operation and a low cost. With this process, the concentration of different organic pollutants can be cost-effectively diminished while improving the quality of the water resource [5–7].

One of the most significant features of adsorbents that must be considered is their reusability, which has a great influence on the economic viability of this process. The absorbed contaminant in this process must be retrieved from the sorbent surface [8]. Other benefits of the sorption approach that should be considered include the abundance of sorbents, a high sorption capacity, high recyclability, cost effectiveness and easy desorption of contaminants [9,10].

The main goal of this review paper is to compare the performance of nanocomposites and biocomposites in removing organic pollutants from water sources. To do this, various kinds of pollutants, different kinds of adsorbents and the influence of impressive variables in removing contaminants were studied. Sorption kinetics is another factor that was thoroughly studied. The sorption mechanism of organic contaminants was also studied. A comprehensive survey was performed to compare the removal efficiency of organic pollutants using nanocomposites and biocomposites.

2. Different Pollutants in Water

There are limited sources of freshwater available all over the world, which are not enough for human life. The industrialization of societies and the rapid growth of technology have resulted in declining water sources and increasing the volume of polluted water [11,12]. The presence of contaminants in water results in various diseases annually in humans and animals. Not only does polluted water pose a threat to human health, but it also poses a threat to the whole ecosystem. There are a myriad inorganic and organic contaminant types typically encountered in the aqueous environment (Figure 1). Heavy metal ions are in the group of inorganics, whereas dyes, hydrocarbons, pesticides and drugs are organic components. Organic compounds are utilized in various industries and can be generated by many industrial activities. These contaminants can enter sewage systems and pollute them. Other kinds of organics such as pesticides have remarkable toxic influences on living creatures. Owing to the detrimental influences of these materials, they must be eliminated from water resources [13].



Organic pollutants

- Hydrocarbon compounds (e.g., phenol, benzene, toluene, formaldehyde, aromatic compounds)
- Drugs, pesticides, and insecticides (e.g., Tetracycline, Cefalexin, Ibuprofen)
- Dyes (e.g., malachite green, methyl violet, methylene blue, basic yellow, rhodamine 6G, methyl orange, congo red)

Inorganic pollutants

- Toxic and heavy metals (e.g., Pb^{2+}, Cd^{2+}, Cr^{3+}, As^{3+}, As^{5+}, Hg^{2+}, Cr^{6+}, Co^{2+})
- Sulphates, nitrates, phosphates

Figure 1. Different pollutants in sewage.

2.1. Dyes

Dyes are a large and diverse group of organic contaminants existing in wastewaters that can enter effluent through various industries, such as printing, paper, painting and leather. They exist naturally and can be prepared synthetically. Every year, almost 100 tons of various dyes are discharged directly and indirectly into effluent. They are classified into three groups, including cationic, anionic and non-ionic. Depending on their concentration in aqueous media and contact duration, dyes can have detrimental influences on organisms. For instance, some of their impacts include mutation, dermatitis, skin irritation, allergies, cancer, etc. [14]. There are positive charges on the structure of cationic dyes, and they are known as basic dyes. These dyes are mutagenic, toxic, carcinogenic and water-soluble and are widely employed in many products, such as silk dyeing, acrylic and nylon. Cationic dyes have high color intensity and brightness. The most critical dyes of this group include basic yellow (BY), rhodamine 6G (RG), methylene blue (MB), methyl violet (MV) and malachite green (MG) [15]. Another destructive impact of cationic dyes in water at high concentrations is that the presence of these dyes prohibits light penetration in water and influences the photosynthesis process [16].

Unlike cationic dyes, there are negative charges on the anionic dye structure. Anionic dyes, known as direct dyes, comprise a wide range of reactive dyes. Some critical anionic dyes include methyl orange (MO), direct green (DG), direct red (DR), congo red (CR) and reactive black (RB). Dispersing dyes or non-ionic dyes are the last class of dyes, which are widely utilized for synthesizing polyamide, colored plastics, polyester, etc. Another negative effect of these dyes is causing allergies in the human body [14,17].

The presence of these dyes poses a destructive influence to water quality as well as human health. Consequently, many industries try to eliminate these toxic dyes from sewage before discharging these dyes into water sources. In this regard, the role of scientific communities in developing efficient methods is very crucial [17].

2.2. Drugs, Pesticides and Insecticides

Drugs have many applications and can be utilized to treat diseases. However, discharging drugs in high concentrations into wastewater has a detrimental impact on the environment and can be a serious threat to the lives of all living organisms. Moreover, some antibiotics such as chloramphenicol and sulphonamides are very toxic when present in sewage [18,19].

Pesticides are another organic compound that is extensively utilized for protecting plants against pests, fungi and weeds. Residues of these toxic materials have critical environmental influences on aquatic ecosystems. They can be spread in groundwaters in various ways, such as spraying and dusting [20]. Atrazine is a type of pesticide that disrupts the hormonal activity of living organisms such as humans. The presence of some pesticides in water, even at low concentrations, results in death. The elimination of pesticides, therefore, is of great importance [21,22].

2.3. Other Organic Pollutants

Volatile organic hydrocarbons, such as phenol, benzene, toluene and aromatic compounds, are another kind of organic contaminant that is widely present in industrial wastewaters, such as refinery wastewater systems. The presence of toxic organic compounds such as aromatic hydrocarbon compounds in effluent has detrimental impacts on vomiting, the liver, the kidneys, the nervous system, nausea, cataracts, eye irritation and the lungs [23]. Another type of organic contaminant in wastewater is hydrocarbon compounds such as oil and gas condensate. This type of wastewater is called oil-in-water emulsion. Figure 2 shows an image of an oil/water emulsion. As shown, oil droplets with various sizes are dispersed in the continuous water phase [24].

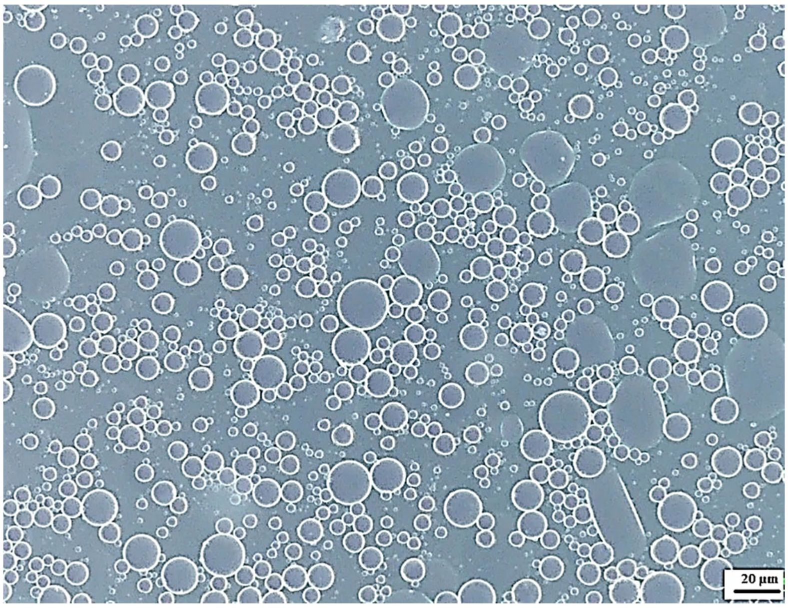
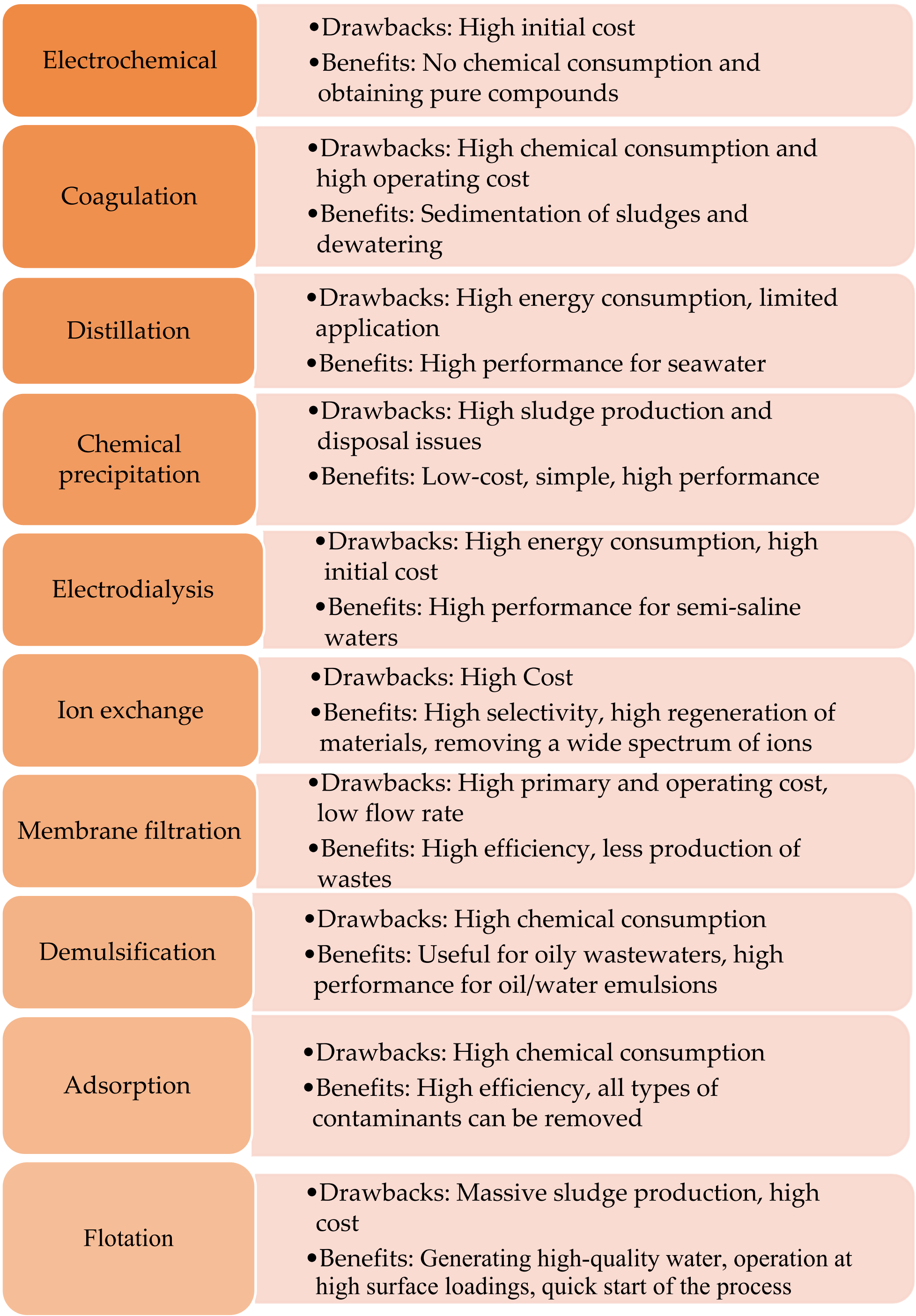


Figure 2. Distribution of oil droplets in the continuous water phase [24].

3. Wastewater Treatment Approaches

There are various procedures to eliminate organic contaminants from aqueous media, including membrane processes, demulsification, electrochemical procedures, ion exchange, electrodialysis, distillation, chemical precipitation, coagulation, gravity, filtration and sorption [6,25]. Different processes for organic contaminant removal, as well as their pros and cons, are described in Figure 3. Knowing these features is very critical for the efficient use of suitable approaches in pollutant removal [6,25].

Electrical treatment processes encompass a variety of technologies, such as electrochemical processes, electro-flotation, electro-coagulation, electro-precipitation, electrophoresis, etc. [26]. The electrochemical process is an efficient method to delete different kinds of pollutants and includes several procedures such as electro-coagulation, electrodeposition and electro-flotation. In this process, electrons move between organic contaminants and electrodes. The equipment in this technology is relatively expensive, and this process requires a high investment. Another drawback is high sludge disposal. However, this process consumes no chemicals and has high selectivity for metallic contaminants [27,28].



Electrochemical	• Drawbacks: High initial cost • Benefits: No chemical consumption and obtaining pure compounds
Coagulation	• Drawbacks: High chemical consumption and high operating cost • Benefits: Sedimentation of sludges and dewatering
Distillation	• Drawbacks: High energy consumption, limited application • Benefits: High performance for seawater
Chemical precipitation	• Drawbacks: High sludge production and disposal issues • Benefits: Low-cost, simple, high performance
Electrodialysis	• Drawbacks: High energy consumption, high initial cost • Benefits: High performance for semi-saline waters
Ion exchange	• Drawbacks: High Cost • Benefits: High selectivity, high regeneration of materials, removing a wide spectrum of ions
Membrane filtration	• Drawbacks: High primary and operating cost, low flow rate • Benefits: High efficiency, less production of wastes
Demulsification	• Drawbacks: High chemical consumption • Benefits: Useful for oily wastewaters, high performance for oil/water emulsions
Adsorption	• Drawbacks: High chemical consumption • Benefits: High efficiency, all types of contaminants can be removed
Flotation	• Drawbacks: Massive sludge production, high cost • Benefits: Generating high-quality water, operation at high surface loadings, quick start of the process

Figure 3. Various processes and their pros and cons for water decontamination [26–33].

Flocculation is also a common process in wastewater treatment systems that uses chemicals to coagulate contaminants and separate them from polluted water [29]. Flocculation destabilizes colloids via the neutralization of the forces that separate them. Aluminum, $FeSO_4$ and $FeCl_3$ are some critical and widely utilized flocculants that have been utilized extensively for water decontamination. The most significant drawbacks of this process is that this method has a high cost and requires a large quantity of chemicals. Coagulants are usually utilized in the primary treatment of wastewater [30,31].

Moreover, ion-exchange is an effective process for eliminating cationic and anionic pollutants, so this technology is very vital to diminishing heavy metals, owing to its high removal performance, significant regeneration and fast kinetics. There are many kinds of synthetic and natural resins that can be utilized in this technology, among which synthetic resins can be preferred. In ion-exchange technology, undesirable ions are exchanged with desirable ones. However, this process has some critical drawbacks, such as a high cost and a low removal efficiency for metal ions [29].

In many wastewater treatment systems, utilizing filtration such membrane technology is very common, especially in the treatment of seawater. The utilization of reverse osmosis and nanofiltration techniques has received much attention in recent years. These processes work at a high pressure to eliminate contaminants from sewage. Moreover, they are utilized to eliminate turbidity and microbial pollutants. The disadvantages of membrane technology are a high operation cost and a low flow rate, which limit their utilization. However, this technology is highly efficient for removing different kinds of ions with excellent efficiency, which makes this process unique for water purification, especially seawater [32].

Moreover, chemical precipitation is a simple and inexpensive approach that can eliminate most pollutants. However, this technology has some drawbacks, such as a large amount of sludge and disposal issues, which limit its application in some areas. The sediments in this procedure can be filtered to separate from wastewater [33].

4. Sorption Technique

Among all kinds of processes, the sorption process has received significant attention, owing to many benefits such as its cost effectiveness and high removal efficiency of organic contaminants. In this method, adsorbents play a critical role in economical discussion. Its low cost, environmental friendliness and high recyclability are crucial features that should be considered [6]. In fact, the sorption mechanism occurs between the sorbent surface and the sorbed contaminant. Moreover, the sorption of contaminants on the sorbent surface takes place physically or chemically, among which physical sorption is preferred, as in this case, the sorbent can be recovered easily and then reutilized. In physical sorption, a weak van der Waals force is formed between the contaminant and the sorbent surface. Therefore, this force is broken easily, and the contaminant is eliminated from the sorbent surface, whereas the chemisorption process is irreversible and needs significant energy. This technology can be utilized to remove contaminants even at low concentrations (1 ppm) [34–36].

5. Different Kinds of Sorbents

Finding a proper sorbent is a major challenge in industrial applications. To this end, generating an efficient sorbent for removing organic contaminants plays a vital role in this process. There are various kinds of adsorbents in the literature. In general, they are classified into two general types, including biosorbents and conventional sorbents [37].

5.1. Biosorbents

Biosorbents are materials that are extracted from nature. Figure 4 illustrates various materials that can be utilized in synthesizing biosorbents. As shown, agricultural wastes, marine algae, soil, microbial biomasses and industrial wastes are different sources for biosorbent production [38].

Agricultural wastes are abundantly found in nature. They are inexpensive and environmentally friendly. Some examples of agricultural wastes include peat bagasse, rice hull ash, pistachio peels, orange peels, pomelo peels, garlic peels, tree leaves, etc., which are extensively employed in removing contaminants from polluted water [34]. The main ingredients in these materials include lignin, cellulose, proteins and hemicelluloses. These materials have shown to be very efficient in wastewater treatment. Moreover, marine materials have many advantages, such as a low cost and abundance. Chitin and chitosan are two valuable materials that can be found in their structure, which have been extensively utilized as efficient sorbents for water decontamination in previous studies. Chitin in high amounts can be extracted from the bodies of crabs, shrimp, etc. Moreover, it can be extracted from the cell walls of fungi [7]. Moreover, industrial activities can generate significant solid wastes, which include industrial residues such as aluminum, steel, leather, fertilizer and paper waste [6,14]. These wastes are free of cost, and discarding them into the environment has detrimental influences. Therefore, the efficient utilization of these wastes in water decontamination is of great importance. For instance, red mud can be generated from aluminum industry wastes, which have been utilized in wastewater treatment. Zeolites (e.g., clinoptilolite), soil, clays (e.g., montmorillonite) and ore are another type of sorbent that has been employed widely in removing organic contaminants. There are negative charges on the clay structure that can be effective in the sorption of cationic dyes [39].

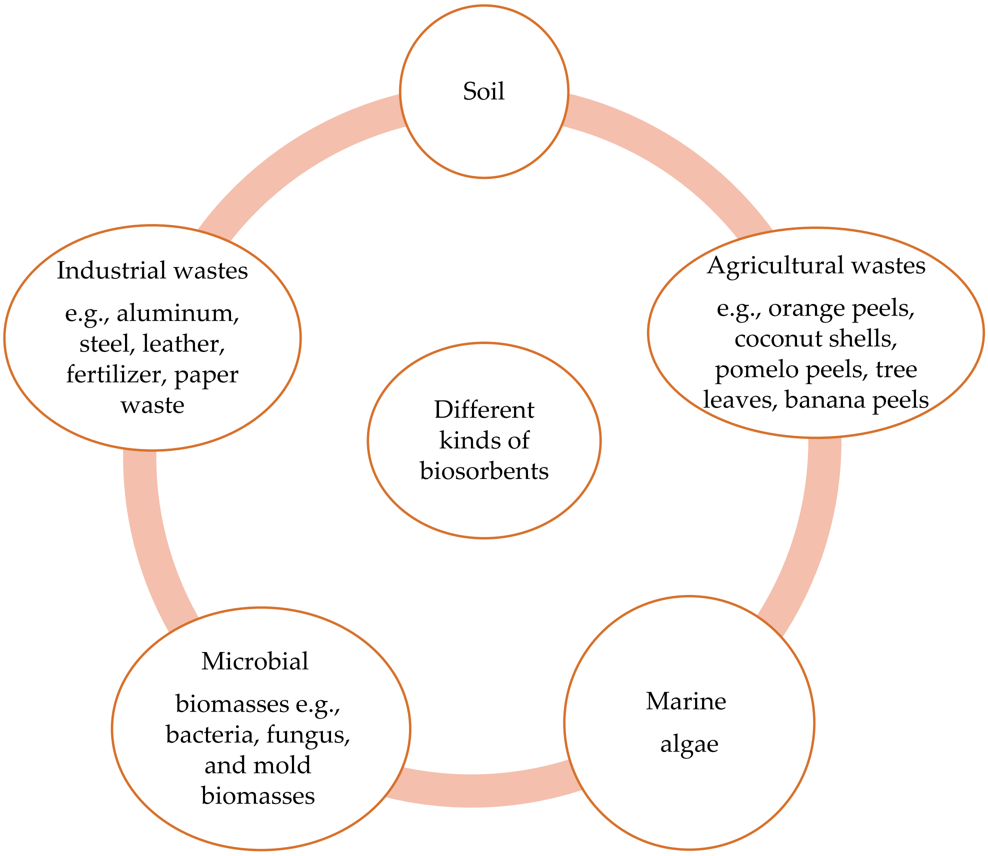


Figure 4. Various types of materials for biosorbent synthesis [38–42].

Microbial biomasses are another source of biosorbents that is extensively employed in treating water [40]. They are classified into bacteria (e.g., *Desulfovibrio desulfuricans*), algae (e.g., *Sargassum dentifolium*) and fungi (e.g., *Aspergillus* sp.). Microorganisms used in this case can be dead and/or alive, among which dead microbial biomasses are preferred due to the lack of nutrients in sewage and economical reasons. The presence of specific functional groups in microorganisms has made them unique in wastewater treatment [38]. There are enormous membranes in bacterial structures that can be utilized in eliminating pollutants [40]. Moreover, there are 40,000 different species of algae in nature, which produce about 52 million tons of organic carbon per year. Their structure is highly porous. Moreover, many kinds of functional groups are available in the algae structure, such as carboxyl, amino, sulfate, COO^-, OH^- and hydroxyl, indicating the high potential of algae in the uptake process [41]. Furthermore, the cell walls of fungi contain chitosan and chitin, which have many applications in the uptake process. The presence of hydroxyl, carboxyl, phosphate and amine groups in their structure is very useful in the adsorption of contaminants [42].

Table 1 reports the sorption efficiency and capacity of various composites as well as operating conditions in removing various kinds of organic pollutants. As shown, polyaniline/sodium alginate/*Oscillatoria* filamentous cyanobacterium biomass has a sorption capacity of 786.6 mg/g for Basic Blue 41 removal, which is the highest sorption capacity of this dye among all previous studies [43]. Moreover, palm oil was separated from an oily emulsion with a yield of 99.47% with the help of cow bones/zeolite/coconut biocomposite, indicating the high potential of this biocomposite in demulsification. Another efficient biocomposite in Table 1 is related to AC derived from melamine and sucrose in the elimination of MB dye, which, after 13 min, demonstrated a significant sorption capacity of 454.57 mg/g. Generally, it can be concluded that there are several biocomposites that illustrate remarkable potential in removing pollutants in addition to having a low cost and being compatible with the environment.

Table 1. Sorption capacities and sorption yields of various biocomposites in removing organic compounds.

Adsorbent	Contaminants	Conditions	RE (%)	q_{max} (m/g)	Ref.
Polyaniline/sodium alginate/ *Oscillatoria* biomass	Basic Blue 41	pH = 9, AD = 0.1 g/L, t = 2 h, T = 30 °C	-	786.6	[43]
Polyaniline/sodium alginate/ *Oscillatoria* biomass	Basic Blue 41	pH = 9, AD = 0.1 g/L, t = 2 h, T = 30 °C	-	700.1	[43]
Polyaniline/sodium alginate/ *Oscillatoria* biomass	Basic Blue 41	pH = 9, AD = 0.1 g/L, t = 2 h, T = 30 °C	-	634.4	[43]
Chitosan–alkali lignin composite	Remazol Brilliant Blue R	pH = 2, PC = 10 ppm, AD = 2 g/L, t = 1 h	-	111.1	[44]
Non-immobilized magnetic biocomposite	MB	pH = 7, PC = 10 ppm, AD = 5 g/L, t = 140 min	99.12	232.6	[45]
Cow bones/zeolite/coconut biocomposite	Palm oil	pH = 5.28	99.47	-	[46]
Chitosan/sepiolite biocomposite	Remazol Blue	pH = 5, PC = 3 ppm, AD = 0.8 g/L, t = 24 h, T = 45 °C	99	256	[47]
Alginate/magnetic/graphene oxide biocomposite	Aniline	-	98.82	18.39	[48]
Alginate/magnetic/graphene oxide biocomposite	*p*-chloroaniline	-	96.65	17.13	[48]
Alginate/magnetic/graphene oxide biocomposite	*p*-nitroaniline	-	93.55	15.24	[48]
Chitosan/diatomite biocomposite	Reactive Blue	pH = 4, AD = 0.75 g/L, T = 30 °C, t = 50 min	96.36	-	[49]
Chitosan/zeolite biocomposite	Acid Black 194	pH = 3, AD = 5 g/L, T = 22 °C	-	2140	[50]
Chitosan	MB	pH = 6, PC = 105 ppm, AD = 1.6 g/L, t = 6 h, T = 25 °C	85.85	11.04	[51]
10% chitosan/90% activated carbon	MB	pH = 6, PC = 105 ppm, AD = 1.6 g/L, t = 6 h, T = 25 °C	91.7	11.92	[51]
20% chitosan/80% activated carbon	MB	pH = 6, PC = 105 ppm, AD = 1.6 g/L, t = 6 h, T = 25 °C	95.81	11.99	[51]
Olive waste/KOH	MB	pH = 11, t = 0.5 h, T = 40 °C		504.9	[52]
Olive waste/KOH/PEG silica gel	MB	pH = 12, t = 0.5 h, T = 50 °C		161.44	[52]
Modified sepiolite	Everzol Yellow	pH = 2, PC = 25 ppm, AD = 50 g/L, t = 2 h	-	169.1	[53]
Modified zeolite	Everzol red	pH = 2, PC = 25 ppm, AD = 50 g/L, t = 2 h	-	111.1	[53]
Metal hydroxide sludge	Direct Blue 85	pH = 7, T = 25 °C	-	339	[54]
Aminated polyacrylonitrile	MB	pH = 8, t = 20 min	99.7	227.2	[55]
Magnetic/biochar nanocomposite	MB	-	-	62.1	[56]
Chitosan/laterite/Fe_3O_4	MB	-	-	16	[57]
Sucrose and melamine-derived AC	MB	t = 13 min	-	454.57	[58]

Note(s): RE: Removal efficiency; t: time; T: temperature; AD: adsorbent dosage; PC: pollutant concentration.

5.2. Nanocomposites

Another type of sorbent that has recently received much attention is nanocomposites. They consist of two phases, including the reinforcement phase and the matrix phase, whose dimensions are on the nanoscale. The most critical features of nanosorbents are their reversibility, high specific surface area, excellent sorption capacity, biocompatibility and high active sites [4,6]. Moreover, their pore size is tunable. Various analyses can be employed to assess nanocomposites, including DLS, TEM, SEM and XRD. The particle sizes of the nanocomposites can be determined via TEM and DLS analyses. Moreover, SEM and XRD analyses are utilized to determine the sizes of pores and crystals, respectively [6].

There are various kinds of nanoadsorbents, which include metallic nanomaterials, silicon nanomaterials, carbonaceous nanomaterials, metal chalcogenide nanomaterials, metal oxide nanomaterials and magnetic nanomaterials. Figure 5 shows different groups of nanoadsorbents. As shown, FeS, CuSe, MoS_2, WSe_2, MnS and NiS are some important metal chalcogenide nanoadsorbents. Moreover, aluminum oxide, nickel oxide, magnesium oxide, iron oxide, zirconium oxide and titanium oxide are several types of metal-based nanosorbents. Moreover, silicon nanosheets and silicon nanotubes belong to the group of silicon nanomaterials. Furthermore, chitosan, chitin, polyacrylonitrile, polysiloxane/graphene oxide, alginate, dendrimers, electrospun fabric and cellulose are some critical polymeric nanosorbents. Moreover, carbon nanotubes (CNTs), multi-walled carbon nanotubes (MWCNTs), graphene, graphene oxide and their composites are in the group of carbon-based nanosorbents. The presence of iron oxide in the composite structure gives it a magnetic feature that can be easily separated utilizing a magnet. Polydispersity and size control are factors that are critical for developing an efficient sorbent. Additionally, $CoFe_2O_4$, Fe_2O_3 and Fe_3O_4 are some important magnetic nanoadsorbents [59,60]. Polymeric nanocomposites, owing to their low cost and simple utilization, have recently been widely utilized and can be synthesized with various processes such as the sol–gel process, intercalation polymerization, etc. Moreover, supported metal-based composites have many advantages, including significant active areas, high stability (chemical and thermal) and remarkable catalytic activity, which make them unique materials. Moreover, carbon-based materials have enormous features, such as excellent specific surface areas, structured pores and a high potential for the elimination of organic contaminants [61]. The advantages of magnetic nanoparticles include high stability, easy separation from aqueous solutions, easy synthesis, a considerable specific surface area and high removal efficiency of contaminants [35].

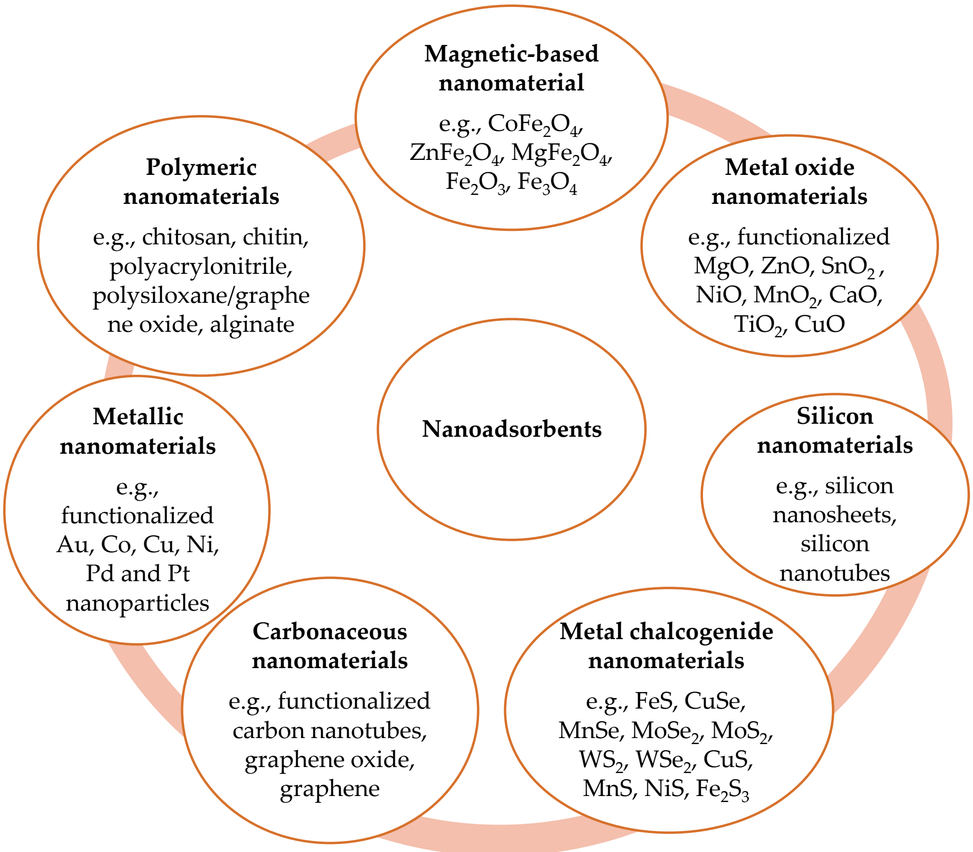


Figure 5. Different classifications of nanoadsorbents.

Several nanosorbents have been utilized in the sorption process so far, including silica/Fe_3O_4, clay/starch, bentonite/$CuFe_2O_4$ and activated carbon/Fe_3O_4 [62]. Table 2 reports the uptake capacity and uptake efficiency of various nanosorbents in organic contaminant removal from aqueous media. As given, the SiO_2/$MnFe_2O_4$/ZIF-8 MOF nanocomposite showed high performance in eliminating malachite green and methyl red, with a significant sorption capacity of 1000.03 and 1111.12 mg/g, respectively. This nanocomposite showed the highest uptake capacity among the conducted studies. Another significant nanocomposite was magnetite/MWCNTs, which was able to eliminate toluene with an uptake capacity of 1113 mg/g. Other nanocomposites, such as nano zerovalent iron, barium/Cobalt@ polyethylene glycol (PEG), graphene oxide/Fe_3O_4, kaolin/$CuFe_2O_4$ and Fe_3O_4@UiO-66, showed sorption capacities higher than 100 mg/g, indicating their high performance.

Table 2. Sorption capacities and sorption yields of various biosorbents in removing organic compounds.

Nanocomposite	Contaminant	Conditions	RE (%)	q_{max} (mg/g)	Ref.
Zinc (II) oxide nanorods loaded on AC	Crystal violet	PC = 14 ppm, AD = 0.025 g/L, t = 0.085 h	98.66	81.6	[63]
Zinc (II) oxide nanorods loaded on AC	MB	PC = 14 ppm, AD = 0.025 g/L, t = 0.085 h	99.46	83.9	[63]
Barium/Cobalt@PEG	MB	pH = 2, PC = 20 ppm, t = 1 h	78.6	215.08	[64]
Zeolite/Fe_3O_4	MB	pH = 7, PC = 2 ppm, AD = 2.8 g/L, t = 3 h	97.5	2.57	[65]
Kaolin/$CuFe_2O_4$	MB	pH = 8, PC = 10 ppm, AD = 1.5 g/L, t = 0.5 h, T = 25 °C	98.64	120.48	[66]
Kaolin/$CuFe_2O_4$	MV	pH = 8, PC = 10 ppm, AD = 1.5 g/L, t = 0.5 h, T = 25 °C	99.25	117.65	[66]
Clay/starch/ Fe_3O_4	MV	pH = 9, PC = 10 ppm, AD = 1.5 g/L, t = 2.5 h	99.73	29.67	[67]
Nano zerovalent iron	Acidic dye	pH = 5.5, PC = 30 ppm, AD = 0.5 g/L, t = 0.025 h	94.29	125.8	[68]
Graphene oxide/Fe_3O_4	Erythrosine	pH = 7, PC = 50 ppm, AD = 4 g/L, t = 2 min, T = 60 °C	97	149.25	[69]
Date seed AC/CaO/Fe_3O_4	Formaldehyde	pH = 7, PC = 5 ppm, AD = 2.5 g/L, t = 80 min, T = 25 °C	98.22	24.01	[70]
AC/Fe_3O_4	Formaldehyde	pH = 7, PC = 5 ppm, AD = 3 g/L, t = 100 min, T = 25 °C	95.67	24.21	[70]
CaO/Fe_3O_4	Formaldehyde	pH = 7, PC = 5 ppm, AD = 3 g/L, t = 100 min, T = 25 °C	95.14	21.28	[70]
Alginate@Fe_3O_4@bentonite	Formaldehyde	pH = 9.96, PC = 10.69 ppm, AD = 1.28 g/L, t = 16.53 min, T = 25 °C	94.56	50.25	[71]
Fe_3O_4@UiO-66	MO	pH = 4, PC = 20 ppm, AD = 0.2 g/L, t = 3 h, T = 25 °C	-	243.9	[72]
Fe_3O_4@UiO-66	MB	pH = 8, PC = 20 ppm, AD = 0.2 g/L, t = 3 h, T = 25 °C	-	769.23	[72]
SiO_2/$MnFe_2O_4$/ZIF-8 MOF	Malachite green	pH = 3, PC = 100 ppm, AD = 0.2 g/L, t = 1 h, T = 25 °C	-	1000.03	[73]
SiO_2/$MnFe_2O_4$/ZIF-8 MOF	Methyl red	pH = 3, PC = 100 ppm, AD = 0.2 g/L, t = 0.5 h, T = 25 °C	-	1111.12	[73]
Magnetite/MWCNTs	Toluene	pH = 5, PC = 50 ppm, AD = 0.02 g/L, t = 1 h, T = 35 °C	85	1113	[74]
GO/$CuFe_2O_4$/CdS	Imidacloprid and Dinoseb	pH = 7, AD = 0.15 g/L, t = 140 min	91–94	-	[75]
ZnO@$CoFe_2O_4$	Imidacloprid	pH = 10, AD = 0.1 g/L, t = 45 min	79.3	-	[76]

6. Sorption Mechanism

Many industrial effluents exist in the form of oil-in-water (O/W) emulsions, in which oil droplets are dispersed in the continuous water phase. There are many functional groups on the sorbent surface that play a key role in destabilizing the emulsion and separating oil molecules from water. In the sorption process, the oil droplets move toward the hydrophobic surface of the sorbent. The hydrophobic properties of the sorbent play an important role in the movement of non-polar oil molecules to each other and in the production of large droplets. Because composites (i.e., nanocomposites and biocomposites) have many functional groups on their surfaces, they are preferred for adsorbing pollutants over common sorbents such as activated carbon [24].

The sorption mechanism is carried out through the interaction and electrostatic forces between the adsorbent and the adsorbate [77]. The adsorption of oil droplets occurs through the interaction of the oil and the adsorbent surface and then through their physical sorption into the irregular pores on the adsorbent surface. In fact, the oil droplets are trapped on the adsorbent surface by adding the adsorbent to the emulsion [78]. After adding the adsorbent to the emulsion, various mechanisms may occur during the demulsification process, including coalescence, flocculation, creaming and aggregation. These phenomena often lead to the separation of the oil and water phases [79]. Figure 6 indicates different phenomena in an emulsion. If there is a density difference between the oil and water phases, the creaming phenomenon occurs. In this process, the oil droplets stick together and reach the surface of the water. In the flocculation process, oil droplets accumulate and form flocs. Various factors affect the flocculation performance, such as the density difference between two phases, temperature, oil concentration, water viscosity and electrostatic fields. Moreover, coalescence is an important stage in the demulsification process. In the coalescence step, the oil droplets join together and form larger droplets [24].

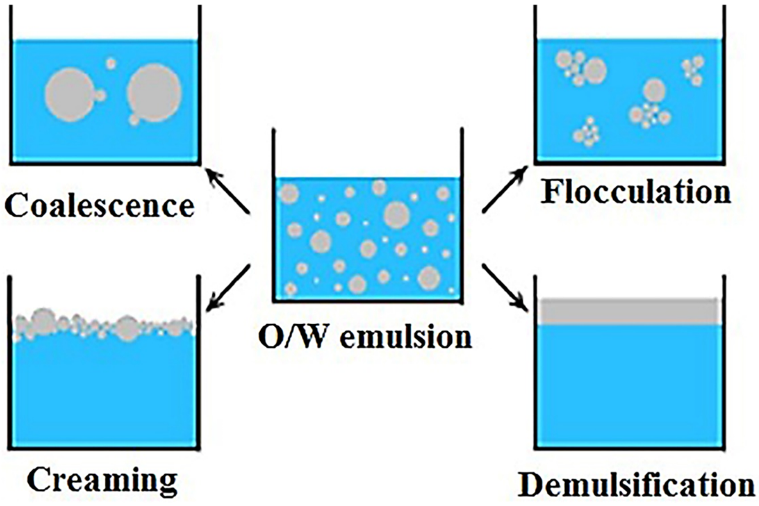


Figure 6. Various mechanisms of oil/water emulsion, including coalescence, flocculation, creaming and demulsification processes.

Figure 7 also indicates how pollutant molecules bond with adsorbent particles. As shown, there are two types of bonds, which include chemical and physical bonds. In physical sorption, a weak van der Waals force is formed between the adsorbent and the adsorbate, whereas in chemical sorption, a chemical bond is created between them [80]. The uptake process generally includes three steps, including (1) the transfer of the pollutant from the solution to the outer side of the sorbent surface, (2) the diffusion of the pollutant from the outer side of the sorbent surface to the sorbent pores and (3) the sorption of the pollutant on the sorbent surface [35]. Figure 8 also illustrates the different sorption mechanisms of organic pollutants on the sorbent surface, including electrostatic interactions, hydrophobic partitioning interactions, hydrogen bonding, pore filling, π–π interactions, and van der Waals forces. The interaction mechanism of hydrophobic partitioning is attributed to the partitioning of organic contaminants between water molecules and the nanocomposite surface. Hydrogen bonds consist of H^+ ions, which are considered a type of weak electrostatic interaction. Functional groups on the surface of the composite can influence the hydrogen bonding interactions. Moreover, π–π interactions can be considered a driving force between aromatic compounds. When organic pollutants and composite particles have electric charges, an electrostatic interaction is created between them. Moreover, the van der Waals force is a weak force that is created between the pollutant and the composite surface during the sorption process [81]. All these mechanisms have been repeatedly observed in previous studies. For example, Xu et al. [82] reported that the sorption of tetracycline from wastewater follows the π–π interaction mechanism. In another study, Wu et al. [83] reported that the sorption of carbamazepine and triclosan follows the hydrophobic interaction. Moreover, Razanajatovo et al. [84] showed that there are hydrophobic interaction and electrostatic interaction mechanisms between the adsorbate (e.g., sertraline and propranolol) and the sorbent surface.

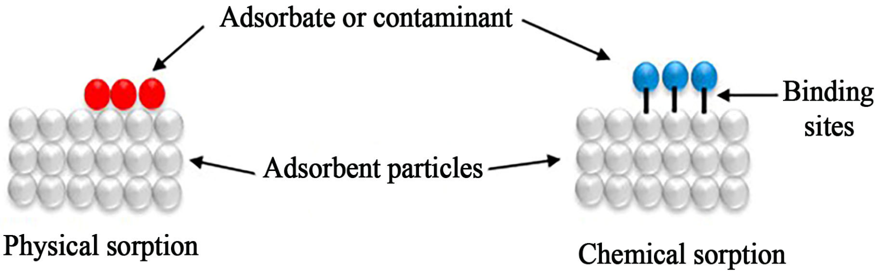


Figure 7. Physical and chemical bonds between adsorbate and adsorbent molecules.

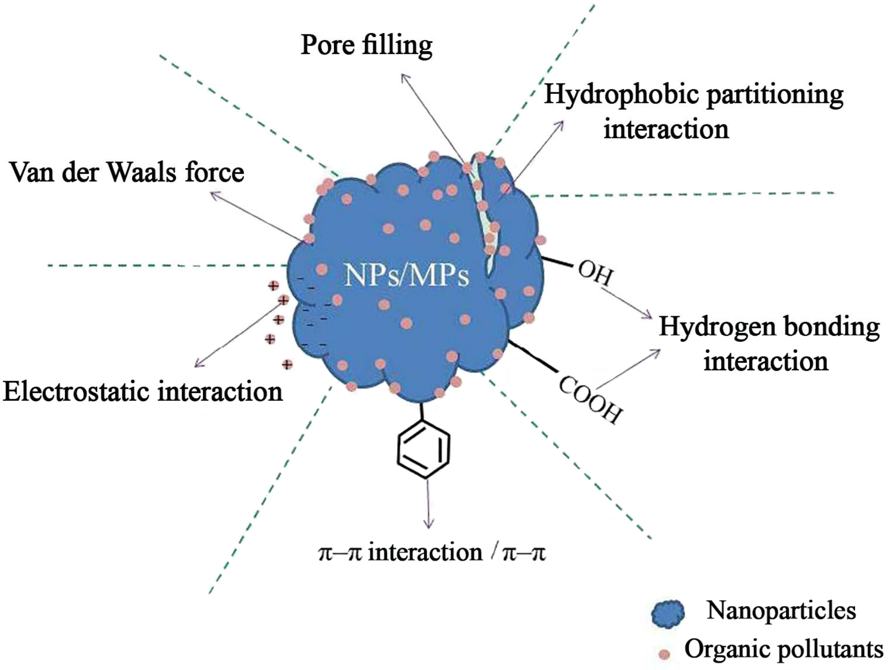


Figure 8. Different sorption mechanisms of organic pollutants on the sorbent surface [81].

7. Effective Variables on Sorption

Several variables affect the performance of a sorbent in the elimination of contaminants, including pH, stirring time, stirring rate, temperature, pollutant concentration and sorbent concentration. Among all variables, pH plays a critical role in separating contaminants from aqueous media. By changing the solution pH, the concentration of positive and negative charges in the solution (e.g., hydronium and hydroxide ions) changes and affects the rate of sorption. In an acidic environment, a large volume of active sites are covered by H^+ ions, so a small number of pollutants can be adsorbed on the sorbent surface. Generally, in an acidic environment, the removal percentage of cationic pollutants is reduced. In other words, in an alkaline environment, the concentration of OH^- ions in the solution increases, which prohibits the sorption of anionic contaminants in such pH values, as there is a severe competition between the OH^- ions and anionic contaminants. Therefore, there will be an electrostatic repulsion force between H^+ ions and cationic contaminants at a low pH, and on the contrary, at a high pH, electrostatic repulsion forces are created between OH^- ions and anionic ions. Moreover, there is no competition between cationic contaminants and OH^- ions at a high pH, leading to an enhanced sorption efficiency [41]. Another critical variable in the sorption of pollutants is temperature, and this factor affects the economic viability of the process. Hence, sorption at an ambient temperature is economically preferred [85]. Moreover, the pollutant concentration in the solution is another factor that should be considered. Previous research has illustrated that the sorption efficiency declines with enhancing the pollutant concentration, owing to the low ratio of pollutants to active sites at low concentrations of the pollutant [86]. The dosage of sorbents is another crucial variable in removing contaminants. By enhancing the sorbent dosage, the number of active sites for trapping the adsorbate enhances. With the help of the sorbent dosage, the utmost sorption capacity is obtained. According to previous investigations, the uptake efficiency enhances with enhancing the sorbent concentration [87]. The rate of the sorption process is affected significantly by the contact time, so the highest change in sorption efficiency occurs at primary times. At the equilibrium time, the sorption rate is constant, illustrating that the sorbent sites are saturated with pollutants, and no additional ions can be adsorbed [88]. Moreover, the stirring speed minimizes the resistance of mass transfer. Moreover, it specifies the energy required for stirring the mix. Generally, the rate of the sorption process increases with enhancing the stirring rate. A high stirring speed leads to vortex creation in the solution and declining solution homogeneity. Moreover, additional stirring decreases the contact time between sorbent particles and contaminants, which leads to a reduced sorption rate [89].

8. Reusability

One of the most significant factors affecting the economic feasibility of the uptake process is the adsorbent's reusability, which is related to its chemical and mechanical stability. Separating nanoparticles from the solution is difficult, owing to their small particle size. Nevertheless, a variety of nanosorbents have demonstrated significant recyclability after several reuse cycles. To regenerate the nanosorbent or biosorbent, several reagents have been utilized so far, including EDTA, NaOH, acetone, ethanol and some acids such as HCl and acetic acid [6]. Table 3 reports some previous works on this subject. Accordingly, poly(N-isopropylacrylamide)/Fe_3O_4 has significant stability, as its removal efficiency for oil removal from wastewater declined from 97 to 90% (i.e., 7.2% reduction in removal efficiency) after seven reuse cycles [90]. Moreover, corn stalk biomass/chitosan/Fe_3O_4 biocomposite showed remarkable recyclability in the elimination of chloramphenicol, as its removal efficiency reduced from 95.49 to 88.52% (i.e., 7.3% reduction in removal efficiency) after five steps [91]. However, among the conducted studies, sodium alginate/magnetic modified raw aquatic weed biocomposite had a low stability, and its removal efficiency declined from 89 to 38% after six cycles (i.e., 57% reduction in its stability), demonstrating a great decrease in its sorption efficiency [45].

Table 3. Comparing the recyclability of various sorbents (biocomposites and nanocomposites) in removing organic pollutants.

Adsorbent	Contaminant	RE (%)	RE (%) after n Reuse Cycles	Ref.
Poly(N-isopropylacrylamide)/Fe_3O_4	Oil/water emulsion	About 97	*n* = 7; RE = about 90%	[90]
Corn stalk biomass/chitosan/Fe_3O_4 biocomposite	Chloramphenicol	95.49	*n* = 5; RE = 88.52%	[91]
$CuCoFe_2O_4$@Chitosan nanocompoiste	Tetracycline	93.07	*n* = 4; RE = 82.16%	[92]
Magnetic nano-silica	Oily wastewater	85	*n* = 5; RE = 80%	[93]
Poly methyl vinyl ketone/aniline/GO	Direct Red 81	89	*n* = 7; RE = 70%	[94]
SiO_2/$MnFe_2O_4$/ZIF-8	Malachite green	99	*n* = 5; RE = 93.5%	[73]
SiO_2/$MnFe_2O_4$/ZIF-8	Methyl red	93	*n* = 5; RE = 86.5%	[73]
Magnetic modified raw aquatic weed biocomposite	MB	99.12	*n* = 6; RE = 65.6%	[45]
Sodium alginate/magnetic modified raw aquatic weed biocomposite	MB	89	*n* = 6; RE = 38%	[45]

9. Sorption Kinetics

Kinetics are equations that describe the sorption behavior as well as the diffusion rate of pollutants in the sorbent pores. Moreover, with the help of kinetics, useful information about the sorption mechanism is attained. The future industrial utilization of a process can be determined according to the kinetic rate [95]. There are several models that can properly describe the kinetic behavior of the uptake process, including pseudo first-order (PFO), intraparticle diffusion (ID), Elovich and pseudo second-order (PSO) models. These models can determine the rate controlling stages and mechanism. They include three steps: exterior mass transfer from the suspension on the outer side of the sorbent, penetrating the adsorbate into the pores and adsorption on the sorbent. The PFO (Equation (1)), ID (Equation (2)), Elovich (Equation (3)) and PSO (Equation (4)) models are defined as follows [96]:

$$W_t = W_e\left(1 - e^{-k_1 t}\right) \tag{1}$$

$$W_t = k_i t^{1/2} + C \tag{2}$$

$$W_t = \beta \ln(\alpha\beta) + \ln(t) \tag{3}$$

$$\frac{t}{W} = \frac{t}{W_e} + \frac{1}{k_2 W_e^2} \tag{4}$$

In Equation (1), k_1 (the rate constant of the process, 1/min) is the time period for achieving the equilibrium. W_t and W_e are also the uptake capacity of a sorbent at time t and the equilibrium state (mg/g), respectively. Moreover, k_2 (g/mg·min) and k_i (mg/g·min$^{1/2}$) can be defined as the constant rate of the PSO and ID models, respectively. Further, the constant C in Equation (2) gives the boundary layer thickness. α and β are the initial sorption rate (mg/g·min) and the desorption constant (g/mg), respectively [96]. Table 4 indicates the kinetic outcomes of various biocomposites and nanocomposites on the elimination of organic compounds. A critical result of these investigations demonstrates that most sorption processes follow the PSO model, owing to its higher correlation factors. In most studies, the PSO and PFO models are utilized to investigate sorption kinetics. Another important outcome of this investigation is that, in almost all studies, the constant k_1 is greater than k_2. Nyankson et al. [97] showed that the uptake of MB using zeolite/cerium oxide occurs in three steps, and they obtained a three-step mechanism.

Table 4. Kinetic outcomes of various composites in removing organic contaminants.

Adsorbent	Pollutant	PFO Constants	PSO Constants	Elovich Constants	ID Constants	Ref.
$SiO_2/MnFe_2O_4$/ZIF-8	Malachite green	k_1 = 0.062 R^2 = 0.991	k_2 = 0.001 R^2 = 0.999	-	-	[73]
$SiO_2/MnFe_2O_4$/ZIF-8	Methyl red	k_1 = 0.160 R^2 = 0.941	k_2 = 0.001 R^2 = 0.999	-	-	[73]
Zeolite/cerium oxide	MB	k_1 = 0.00076 R^2 = 0.44513	k_2 = 1.14655 R^2 = 0.99276	-	*ki*1 = 0.10233 *ki*2 = 0.05229 *ki*3 = 0.02049	[97]
Graphene quantum dots/zeolitic imidazolate framework	Toluene	k_1 = 0.038 R^2 = 0.98	k_2 = 0.000093 R^2 = 0.95	-	-	[98]
Ni-Co-S/SDS	Nile blue	k_1 = 0.0499 R^2 = 0.9614	k_2 = 0.0011 R^2 = 0.9998	-	-	[99]
$CaCO_3$@starch/polyacrylamide /TEMPO-oxidized	Congo red	-	-	-	*ki*1 = 13.84 *ki*2 = 3.54	[100]
$CaCO_3$@starch/polyacrylamide /TEMPO-oxidized	MB	-	-	-	*ki*1 = 5.21 *ki*2 = 1.19	[100]

10. Conclusions

Eliminating organic contaminants from effluents can be carried out via various processes, among which the sorption process is considered an effective procedure, owing to many pros such as a high removal performance for any type of contaminant, a high sorption capacity and an abundance of adsorbents. Selecting a proper sorbent (i.e., one that is cost-effective and highly reusable) in this process is of great importance. The utilization of nanocomposites as well as biocomposites has received much attention, owing to their significant recyclability, high uptake capacity and low contact time. In this study, the removal performance of nanocomposites and biocomposites in removing organic pollutants was investigated. Moreover, the impact of various variables on the uptake yield as well as the sorption kinetics was fully studied. Several types of sorbents such as GO, graphite, CNTs and their composites with other materials such as magnetic nanoparticles have demonstrated remarkable specific surface areas and high sorption efficiencies for the elimination of various kinds of contaminants, especially organic compounds. According to previous works, SiO_2/$MnFe_2O_4$/ZIF-8 MOF, magnetite/MWCNTs, nano zerovalent iron, barium/Cobalt@ polyethylene glycol (PEG), graphene oxide/Fe_3O_4, kaolin/$CuFe_2O_4$ and Fe_3O_4@UiO-66 are some nanocomposites with high performances. Moreover, polyaniline/sodium alginate/*Oscillatoria* biomass, cow bones/zeolite/coconut biocomposite and AC derived from melamine and sucrose are fantastic biocomposites in the removal of organic pollutants. Moreover, the reusability study showed that poly(N-isopropylacrylamide)/Fe_3O_4 and corn stalk biomass/chitosan/Fe_3O_4 biocomposite have significant stability compared to other composites in the elimination of organic pollutants.

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