

A Comprehensive Review of Wastewater Treatment Using Adsorption Process onto Low-Cost Adsorbents

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Abstract

To remove pollutants from its aqueous solutions, adsorption methods are commonly utilized. Adsorption of contaminants such as urea, heavy metals, dyes and other organic pollutants has drawn a lot of attention recently. Low-cost adsorbent is one of the most important factors that influence the adsorption techniques. Various attempts have been studied to prepare low cost adsorbent surfaces to enhance the adsorption efficiency, as well as their chemical and physical features. Using natural materials as adsorbent surfaces have brought significant attention because they were considered as low-cost and eco-friendly adsorbent. The important research areas linked to low-cost adsorbents to remove pollutants from its aqueous solutions and their effects on the adsorption process are reviewed in this paper. This paper provides readers better understood of adsorption process with the potential work using low cost-adsorbent surfaces.

Keywords: adsorption, wastewater, pollutants, urea, dyes, heavy metals.

1. Introduction

Besides the technology developments, the extreme use of chemicals, industrial developments, the fast rise of agriculture, As a result of the world's ever-growing population. All of that have participated by environmental pollution in the soil, water, and air [1]. Because of these human activities, annually huge amounts of organic and inorganic toxic materials are discharged to the water which is effected people health and other creatures especially the marine eco-system [2]. There are many marine living species that have been distinct because of water pollution [3]. Seeing this, it is very necessary to remove these pollutants from water to protect human health and stop the distinction of more creatures [4]. Pollutants are organic, inorganic, or organometallic harmful substances such as dye, heavy metals, and pharmaceutical materials [5]. Extensive works have been done to remove them from water by using various approaches and the most popular method is the adsorption process [6]. This is because the adsorption process is an eco-friendly method, easy to apply, its materials are available and not expensive [7]. On the other hand, adsorption method has demonstrated very high efficiency to remove pollutants as will discuss in the next sections. In recent times, various prepared materials have been investigated as adsorbent surfaces to remove water pollutants [8]. The criteria of using the adsorbent are to obtain an adsorbent surface with remarkable characteristics such as being cheap and not harmful to the environment [9, 10]. Natural materials, agricultural waste, and industrial waste are considered the most common adsorbent surfaces of water pollutants that can be used. This is because these materials are low-cost and eco-friendly adsorbent hence they have been used by many researchers [11, 12]. Figure 1 shows a schematic depiction of wastewater pollution and the most used treatment methods [13].

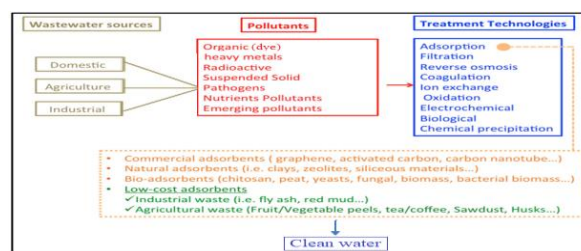


Figure 1: schematic depiction of wastewater pollution and the most used treatment methods.

Several new wastewater treatment technologies have recently been considered. However, scientists are interested in fast, efficient, and low-cost techniques to limit the known dangers of certain chemical processes. When it comes to wastewater treatment, there are many options as shown in figure 1, but adsorption is becoming more popular than other treatments methodologies [14]. There is currently an up-to-date publication on adsorption for wastewater treatment in the last twelve years that is shown in Figure 2. A number of researchers have recently devoted themselves to the search for large surface areas adsorbents, low cost, and environmental friendliness. By using nano-sized materials, the researchers were able to discover effective adsorbents for removing dyes, Toxic heavy metals, organic compounds, and other pollutants from wastewater [15].

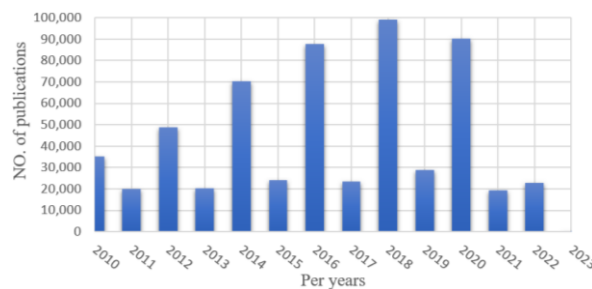


Figure 2: The annual number of research articles published in Google Scholar with the keywords "adsorption & wastewater treatment" from 2010 to 2023.

This paper will focus on the most widely used low-cost adsorbent surfaces for water treatment in the past and present, as well as a

discussion of the results of numerous adsorption studies, to remove the most common water contaminants, such as dyes, heavy metal waste, pharmaceuticals, organic and inorganic wastes.

2. Water Pollutant materials

Pollutants, which are substances such as elements, molecules, or particles involved in pollution, can have an impact on people and plants. Pollutants can enter the environment through a variety of mechanisms, both naturally and because of human activity. The most common human activities involve the use of water for industrial, agricultural, and domestic purposes. These three activities use freshwater that is treated with various chemicals before being discharged into the environment as toxic wastewater that threatens ecosystems [16, 17]. There will be an enormous amount of pollutants discharged into the environment as a result of these activities, including heavy metals, synthetic dyes, and new developing toxins [18, 19]. Some of the most common pollutants are reviewed below sections.

2.1. Organic pollutants (dyes and other compounds)

Nowadays, various types of organic pollutants are contaminating water, and the most famous ones are dyes and phenols [20, 21]. Hence, there are more than ten thousand types of manufactured dyes that are used for different purposes, such as textiles, painting, colored papers, etc. Some of these dyes are water-soluble and others are insoluble. When these dyes contaminate water, it causes color change, which is not acceptable. Furthermore, they are toxic and affect human health [22]. According to numerous studies, dyes from wastewater can be effectively removed using a variety of novel low-cost adsorbent materials. Table 1 summarizes the adsorption ability of low-cost adsorbent surfaces for different organic contaminants such as dyes, phenols, etc.

Phenols are organic molecules produced as by-products of several industrial processes, such as pulp, pesticides, and paper manufacturing [23]. Additionally, phenols cause the bad smell of water. Water contaminated with phenols must be treated before it may be discharged into the environment since it is harmful.

Table 1. The effectiveness of various adsorbent in adsorption of organic compounds

Organic pollutants	Adsorbent surfaces	Isotherm model	Efficacy in adsorbing mg/g	Ref.
phenol	corn cob activated carbon	Thomas, Adams-Bohart, & Wolborska	8.570	24
Rhodamine	Fe\montmorillonite	Langmuir & Freundlich	57	25
Atrazine	Graphene oxide	Langmuir & Freundlich	81	26
tetracycline and copper	biochar	Temkin	91.6	27
phenol	activated carbon from rice husk	Langmuir	179.81	28
acid yellow 199	Activated carbon from lemon wood	Langmuir	85.51	29
methylene blue	activated carbon from wood of cherry tree	Langmuir	41.49	30

2.2. Inorganic pollutants (heavy metals)

Heavy metals are the most hazard materials that contaminating water due to their high toxicity even at

very low percentages [24]. Furthermore, they are found in the wastewater produced by several industries such as batteries, ceramics, photographic and paint manufacturing. The wastewater of these industries contains various heavy metals for example lead, arsenic, chromium, cadmium, nickel, manganese, and mercury; hence they are found in higher amount than the allowed level [25, 26]. As it is known, these pollutants are not safe for human health and marine creatures even at low concentrations. Another safety issue with heavy metals that they are precipitate in the human bodies with the time since it is hard for the bodies to get rid of them. The inorganic metal ions are strong toxicants since they are non-biodegradable and carcinogenic. Worryingly, the concentrations of heavy metal ions in water systems are higher than the limits set by the World Health Organization. This means that many diseases are caused by this, as mentioned in several studies [27-29]. Therefore, heavy metal pollutants have to be removed from wastewater before discharging them to the environment. There have been several recent studies on the use of various sorption materials to remove inorganic metals from water media. A summary of heavy metal adsorption capacities by different sorbents is presented in table 2.

Table 2. Summary of heavy metal adsorption capacities by different low-cost sorbents.

Heavy metal pollutants	Adsorbent surfaces	Isotherm model	% Removal efficacy	Source
Zn, Pb, Cd, Mn & Cu	fly ash	Freundlich	39, 28, 74, 42 & 71.	38
Co, Cr & Ni.	Coir pith	Langmuir & Freundlich	12.82, 11.56 & 15.95.	39
Ni, Cu & Pb.	rice husk	Freundlich, Langmuir & Temkin	74.5, 77.5, & 100.	40
Cd, Cr, Cu, Pb, Ni, & Zn	Soil	Langmuir & Freundlich	35.65, 26.5, 27.6, 34.69, 32.9, 34.46	41
Ni, Cd, Cu, & Zn	Corn	Freundlich	87, 85, 64, & 90	42
Pb, As, & Cd	activated carbon from pistachio shell	Langmuir	14, 15, & 12	43

3. Adsorption process

The concept of an adsorption process means attracting specific materials from the liquid or gas phase toward the solid phase, which leads to increasing the concentration of materials on the solid phase surface [44]. This review covers the solid-liquid adsorption phenomenon; hence the solid phase called the adsorbent, where the adsorption process happens, and the dissolved materials in the liquid phase called the adsorbate, which is adsorbed on the solid surface of the adsorbent. In 1785, the scientist Lowitz was the first to observe the solid-liquid adsorption phenomenon between wood charcoal and colored water [30]. Then, in 1881, Kayser was the first to use the term adsorption [31]. Up to date, numerous research has been done regarding the solid-liquid

adsorption phenomenon to remove water pollutants [32, 33].

Thus, there are two types of adsorptions: physical adsorption and chemical adsorption. Physical adsorption happens by weak physical forces such as van der Waals forces between the adsorbent surface and the adsorbate molecules. This type of adsorption is weak and reversible, so it is possible to re-activate the adsorbent surface and use it again. Chemical adsorption, on the other hand, occurs through stronger chemical bonds and is irreversible. It is definitely stronger than physical adsorption, but, in this case, it is hard to re-activate the adsorbent surface and use it again [34]. Adsorbed species are attached to the solid surface by physiochemical interactions, as shown in Figure 3.

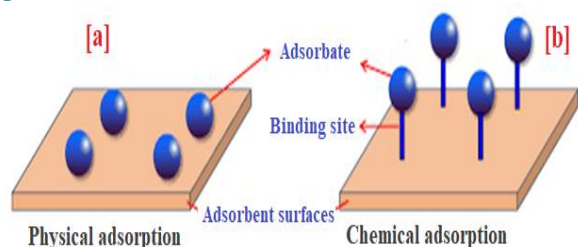


Figure 3: An adsorption system for removing pollutants by (a) physical adsorption (b) chemical adsorption.

4. Physical properties of adsorbent surfaces

Choosing a suitable adsorbent to remove specific pollutants is an important point. However, there are a few factors controlling it. The first point is the surface area of the adsorbent. Hence, the adsorbent works better if it has a bigger surface area. This depends on the porosity of the adsorbent because a porous structure increases the surface area [35, 36]. Figure 4 shows an animated chart of an adsorbent that has a porous structure with different pore sizes and how it captures the adsorbate molecules. The second factor is the adsorption time, which means the time needed to reach the equilibrium of the adsorption process. The shorter time required to reach equilibrium as it is a better adsorbent because almost all these adsorbent surfaces are used in industry with flowing liquids, so a good adsorbent should have the ability to adsorb the pollutants quickly. Other significant parameters determine the quality of the adsorbent surface, such as the cost since the adsorbent has to be available and cheap. The adsorbent also has to be environmentally friendly, so it does not harm the ecosystem. According to the literature, they are going to review some important adsorbents that have shown excellent results for removing water pollutants [37].

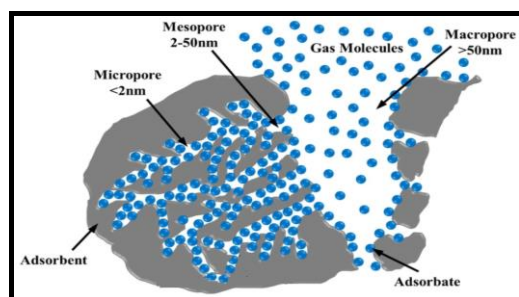


Figure 4: Animated diagram of adsorbent porous structure.

5. Excellent low-cost adsorbent surfaces

Low-cost adsorbents have been extensively studied for the removal of a wide range of water contaminants from aqueous solutions. Natural waste materials such as industrial and agricultural waste can all be used as inexpensive adsorbents, which might be promising adsorbents for environmental and water purification. This section will discuss some of the most essential adsorbent surfaces used to get rid of heavy metals, dyes, and urea, which are both organic and inorganic contaminants. Most often, activated carbon is used as a toxin adsorbent. The high surface area, pore volume, and porosity of activated carbon all contribute to its high adsorption capacity. This is dependent on the manner of activation and the sort of raw materials employed. For years, scientists have been on the lookout for more cost-effective and efficient ways to employ agricultural waste and industrial by-products as adsorbents in their quest for the perfect sorbent. Low-cost adsorbents include natural materials like clay, zeolites, chitosan, and some waste products from industrial operations like fly ash, red mud, and sludge.

5.1. Clay

Clay was used as a cheap and available adsorbent surface for heavy metals. Abid's group employed clay surface to extract cadmium and chromium from their aqueous solutions via the batch method and studied the effect of pH on adsorption efficiency. Clay minerals' adsorption capabilities come from their high surface area and exchange capacities. Because clay has a negative charge, it can attract positively charged adsorbates. They have compared natural clay with purified clay and proved that natural clay absorbs chromium ions in very high quantities. Interestingly, the adsorption isotherms were fit for both the Freundlich and Langmuir isotherms [38]. Among clays, montmorillonite has the most surface area and the greatest cation exchange capacity. In terms of cost, activated carbon is regarded to be more expensive. Therefore, a number of experiments have been undertaken utilizing montmorillonite to demonstrate its ability to remove contaminants from aqueous solutions [39-41]. A growing amount of attention has been paid to clay minerals, including kaolinite and bentonite, which have the ability to absorb both inorganic and organic molecules.

Mengdie Niu et al. reported on the adsorption of Cr (III), Pb (II), and Cd (II) ions from an aqueous solution using three surfaces: a synthesized sulfate aluminate cement-bentonite, pure bentonite, and pure SAC. They found that the adsorption efficiency of SAC-bentonite for Cr (III) was close to that of pure bentonite or pure SAC, whereas the adsorption efficiency for Pb (II) and Cd (II) was notably higher than that of bentonite or SAC. When the ratio of bentonite to SAC was 1:1, the removal efficiency of SAC-bentonite for Cr (III), Pb (II), and Cd (II) was 99.96%, 99.84%, and 99.72%, respectively [42]. A porous surface of Nigerian bentonite was utilized by Kovo et al., for the removal of heavy metals like Ni (II) and Mn (II) from binary systems. Scatchard plot analysis revealed the heterogeneous nature of bentonite clay. High adsorption capacities of both ions were obtained, 200 mg/g and

166.7 mg/g, respectively. During the desorption experiment, they mentioned that more than 90% of both ions were taken out of the bentonite using 0.1M HCL as a desorbing agent [43]. Another study used natural bentonite surface to remove Cu (II), Zn(II) and Co(II) ions from water medias by the adsorption process. Chemical analysis of metal oxides that present in clay was done as follows: SiO₂ (71.90), Al₂O₃ (13.85), Fe₂O₃ (0.68), TiO₂ (0.09), CaO (2.42), MgO (1.27), Na₂O (0.39), K₂O (1.62), and 7.50 percent ignition loss were found. From the experiments, it is shown that the bentonite is more sensitive toward pH changes, Consequently, the negative charges rise as the pH of the solution increases. And as a result, the amount of metal cations adsorbed increases with rising pH and decreased with increase in temperature [44]. Recently, work on the removal of azo dyes from industrial waste was done by Paredes-Quevedo et al. [45] In this work, crude bentonite was utilized as an adsorbent surface to take off cationic dye (basic red 46) from a contaminant solution. They discovered that the adsorption capacity was 594 mg/g at 25 °C and pH = 7. Compared to other materials in the literature, this natural crude bentonite can remove 94% of BR-46 in just 10 minutes, under mild conditions and normal atmospheric pressure Chinoune et al. [46] have identified that bentonite adsorption can be improved by synthesizing magnesium hydroxide coated bentonite surfaces to increase the adsorption capacity of dyes like Procion blue and Remazol brilliant blue from aqueous solutions compared to pure bentonite. The maximum adsorption of dyes was determined to be 98.2% for Remazol brilliant blue and 87.8% for Procion blue.

A highly active mesoporous heterogeneous surface consisting of kaolinite was tested for Acid Black (1) dye adsorption by Kakavandi et al. [47]. Adsorption experiments were carried out in order to establish the equilibrium point and simulate its isotherm. Langmuir's model experiment shows that dye molecules have been adsorbed on surfaces with varying adsorption energies. The adsorption capacity for AB1 dye on NZVI@kaolinite was found to be 24.5 mg/g, indicating that the prepared surface has high performance toward removing pollutants.

Other studies [48] used kaolinite for purification of wastewater from heavy metals like pb and compared its adsorption efficiency to other surfaces called activated phosphate rock and a mixture of kaolinite with APR. Pb removal rates with absorbent kaolinite decreased from 51.85 to 6.91%. On the contrary, they increased with APR and (kaolinite +APR) from 97.15 to 99.35% and 81.74 to 98.18%, respectively.

5.2. Silica Gel

Silica gel consists from aggregation silicon dioxide to form high porous and amorphous granules having various particle sizes. It has very high surface area ranged between 250 m²/g and 900 m²/g this is due to the high porosity. Because of that silica gel considers an excellent adsorbent surface for both gases and soluble pollutants. Therefore it uses in many applications such as material drier, moisture indicator and purification of materials by column chromatography [49, 50]. Figure 5 depicts the

SEM image of silica gel demonstrating the various particle sizes and high porosity of their particles [51].

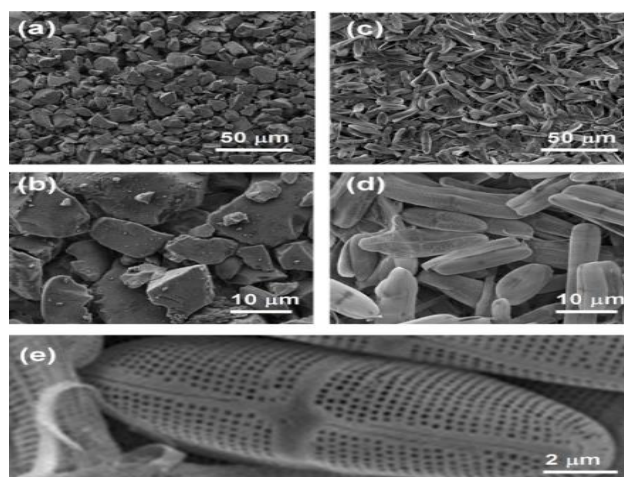


Figure 5: SEM photos of porous materials. a, b are silica gel and c, e are diatom biosilicas.

5.3. Granular activated alumina

Safwat and Minerva have investigated the efficiency of granular activated alumina to adsorb urea from wastewater [52]. To investigate the adsorption kinetics and processes, the experimental data of the adsorption process was fitted to several kinetic models. The equilibrium data were then analyzed by fitting them to several two- and three-parameter isotherm models. The elimination efficiency of urea was improved as the pH value increased, as they have proved. They also demonstrated that the highest adsorption efficiency was 24% at pH = 9. Adsorption of urea onto activated alumina may be described by pseudo-second-order kinetics, according to kinetic studies. The Redlich-Peterson isotherm for activated alumina might be used to express the adsorption isotherms, according to equilibrium research. SEM was used to analyze adsorbents, and the findings revealed the presence of adsorption. Figure 6 shows graphical abstract of adsorption of urea on the granular activated alumina and compares the results with activated carbon.

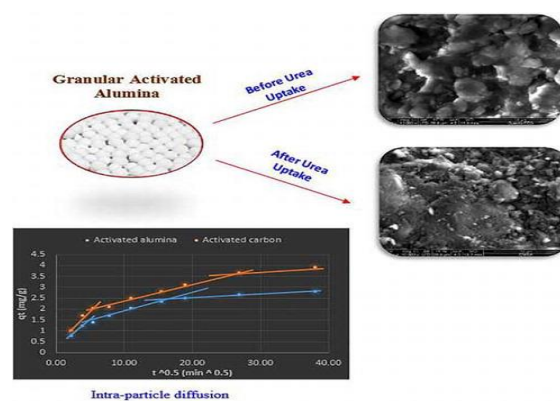


Figure 6: Adsorption of urea on the granular activated alumina and compared them with activated carbon [52].

5.4. Zeolites

Schaif's group has successfully used zeolites as an adsorbent surface to remove urea from its aqueous solutions [53]. Zeolite was used as microporous materials'

physical and chemical characteristics were altered in a systematic way (acidity, grain size, and pore size). The initial urea concentrations of the solutions are similar to those detected in the blood of healthy people and patients with renal failure. The adsorption data obtained at 37 °C reveal that urea adsorption is selective and is dependent not only on the size of the channels but also on the interactions between the chemicals and the zeolite lattices. Since, it was demonstrated that the adsorption of urea onto zeolites was comparable or even better than active carbon.

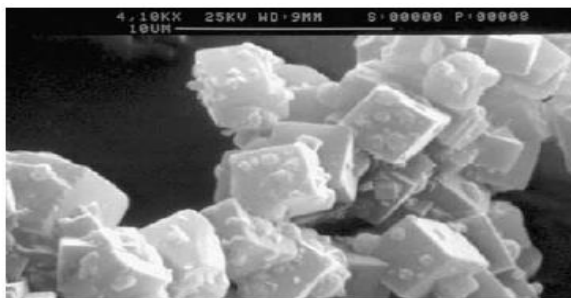


Figure 7: SEM image of zeolite [68].

6. The view from the future

Waste materials can be used in environmental applications and can be substitutes for activated carbon in the removal of undesirable materials from wastewater or the treatment of water pollution. Low-cost and easy-to-find waste materials are available. So, it's likely that soon more people will use low-cost adsorbents to get rid of heavy metal ions.

2. Conclusion

Rapid technology has led to an increase in the use of many toxic materials such as heavy metals, dyes, and other organic compounds, resulting in a severe global environmental hazard. There have been various efforts to develop an appropriate corrective solution due to its toxic nature. To remove these pollutants, several traditional approaches have been employed, such as precipitation, membrane separation, ion exchange, and adsorption processes, in the past. However, these approaches have proven to have several disadvantages owing to a number of challenges, costs, and limitations. Thus, the adsorption process is one of them, and it is particularly beneficial and effective for removing pollutants. For example, silica, clays, activated granulated alumina, and zeolites are extremely effective in adsorbing pollutants such as heavy metals, dyes, and urea from wastewater, and they could be widely used as adsorbent surfaces due to their excellent effectiveness and low-cost considerations.

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