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Research Article



Feasibility study of bentonite for efficient copper removal from Qayyarah refinery wastewater: Optimization of operating conditions and industrial effluent analysis

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Abstract

The purpose of the study is to evaluate the feasibility of using a locally available natural material, i.e. bentonite, to adsorb copper from the industrial wastewater of the refinery, and to identify the optimal conditions for removing copper, which are pH, dose of the adsorbent, contact time, and concentration of the adsorbed ion. Using program (145 Copper, Porphyrin) for estimating copper, the sample is prepared by filling the cell with (10mL) of the sample, then adding the contents of the bag of reagent (Copper Masking), then shaking the cell well and leaving it aside to complete the dissolution of the reagent. Also, another cell is prepared containing (10mL) of the sample, and the contents of one bag of (Porphyrin 1 Reagent Powder) are added to it for each cell. The two cells were shaken well and the second reagent (Porphyrin 2 Reagent Powder) was added to them, one bag for each cell, then the two cells were shaken well and left for (3 minutes) until a pink color appeared for a short period and turned to yellow if copper was present in the sample. The spectrometer has been reset using a blank sample and the concentration of copper was recorded in units ($\mu\text{g/L}$). High effectiveness in copper removal was obtained by the produced adsorbate, Bentonite, at the ideal pH = (6), optimal dosage of the adsorbent (250 mg/L), optimal concentration of the adsorbate (prepared ion solution) (0.08 mg/L), and optimal contact duration (30 min). Under these circumstances, 94% of the copper was extracted. The present investigation found that the amounts of copper in the industrial effluent from the Qayyarah refinery varied from (0.2 - 0.023 mg/l)

Keywords: Bentonite, Copper removal, Industrial wastewater, Adsorption, Optimization

Introduction

The environmental harm produced by people is becoming worse as technology develops and the industrial sector grows, making this a difficult time for all living things on Earth. Unsustainable use of water, air, and soil by a wide range of physical, chemical, and biological factors—the latter having the greatest impact on the aquatic environment—contributes to the degradation of natural resources and aquatic organisms that exist in equilibrium (Osibanjo and Gbadebo, 2011).

Wastewater from refining crude oil is a major environmental hazard because it pollutes both aquatic and terrestrial ecosystems, changing their physical, chemical, and biological features compared to those of unpolluted ecosystems. Rivers

get contaminated with acids, alkalis, dyes, hydrocarbon compounds, hazardous salts, and fats from industrial waste discharged by the chemical, mining, agricultural, and oil sectors. There are two primary groups of minerals found in soil: Physical and mechanical weathering of rocks yields primary minerals, which are themselves composed of minerals that are very resistant to weathering. Quartz, mica, and feldspar are the most significant main minerals, and they make up the bulk of the soil's coarse (sandy and silty) texture (Li et al., 2018).

The second category is secondary minerals, which are minerals that are formed as a result of weathering processes that occur with primary minerals. Secondary minerals form the soft part (clay part) of the soil, the most important of which

are various clay minerals, oxides, and carbonates, and because of the small particles in clay minerals, they will have large surface areas, and thus a high ability to adsorb environmental pollutants, including heavy metals, in aquatic media (Gong et al., 2018). Flexible mineral soil was found in the year (1890) in the earth's crust in the Benton and is considered one of the clays of volcanic origin. The first person to discover this clay mineral was Knight in the year 1890 in the Port-Benton, Wyoming region in America. The highly colloidal and plastic clay deposits found in this region date back to the Cretaceous period. Bentonite was known industrially in the year (1940) as (Wyoming material) and until 1967 the term bentonite was not known to geologists and specialists in the field of minerals, this term was applied to many clays without referring to the formula for bentonite, especially those that contain a high percentage of the mineral montmorillonite. Bentonite is an impure clay composed mainly of montmorillonite, bentonite deposits were discovered 35 kilometers south of the pumping station (H3) in the Western Desert in Iraq, it consists of the highly crystalline mineral montmorillonite (Ca, Na) with a percentage ranging between (55%-85%). During the last twenty years of the last century, the properties of montmorillonite were studied intensively, and this resulted in its introduction into new uses, especially in the chemical and environmental fields. The exploitation of bentonite in Iraq began in the 1970s in "Qara Tabba" and other areas, and extraction continued until the discovery of new deposits in the Western Desert regions in the 1980s (Al-Ghanimi and Al-Zubaidi, 2020). The high efficiency of bentonite clay in removing various pollutants is due to the positively charged edges present on the surface of the bentonite, which are caused by some broken bonds in the lattice structure of the clay crystal, which contains positive silicon ions. The particles also attract and clump together when the negatively charged sides collide with the positively charged edges. Also, increasing the density of suspended particles leads to an increase in the number of collisions between particles, which increases the efficiency of Felting (Ibrahim and Al-Bidry, 2020).

Mineral elements are found naturally in the Earth's crust, and their concentrations vary from one spot to another depending on the nature of the formation of the Earth's crust and the effect of weather factors on its erosion and the release of their naturally occurring elements. The increasing industrial activity has a clear impact on the concentrations of a wide range of trace metal elements, causing pollution of ecosystems with them and in a severe way since the year 1900. As a result of human activity in various areas of life, especially in mining, electric power production stations, oil refineries, agricultural fertilizer production, household sewage, dye industry, automobile exhaust emissions, and others. The term trace elements refers to mineral elements whose concentrations record small and specific values in nature, in addition to the specific need for them in the bodies of living organisms, especially nutritious ones, they are located between the second

and sixth groups of the periodic table, most of them are semi-metallic elements that exhibit metallic properties. They contribute to building living systems, and their high concentrations determine the ability of living organisms to grow and reproduce.

Copper enters aquatic environments from specific and non-specific sources, with toxic concentrations recorded in water bodies and their sediments, it poses a major threat to ecosystems because it does not decompose or break down, with toxic concentrations recorded in the bodies of living organisms and accumulated in the food web. Thus, it is considered a serious and critical problem that affects the health of ecosystems and their life. The study of the effect of copper in river water and its sediments is receiving widespread attention day after day due to the importance of aquatic habitats and the impact of their pollution on human life and well-being, as large quantities of wastewater are produced in the refining of crude oil and the production of electrical energy from black oil or natural gas, and cause the release of toxic concentrations of heavy elements cause deterioration in the quality of water and wetlands. A study of the water characteristics of the Yangtze River in Jiangsu Province in China, showed pollution of the river water and precipitation with toxic concentrations of copper resulting from the cooling water of the electric power plant, recording toxic concentrations of most elements, and copper was one of the most abundant elements in the river sediment.

Heavy metals may be extracted using a wide variety of processes, including chemical precipitation, flotation, coagulation, collecting, ion exchange, and membrane filtering. All of these techniques have their uses, but they can only be used in certain situations and produce a lot of waste. One of the most promising treatment methods is a combination of physical and chemical methods, as well as living organisms; using algae, bacteria, and fungi is one of the best future methods due to their ability to absorb high concentrations of heavy elements relative to their biomass, effectively removing toxic elements from ecosystems at low cost and in significant concentrations. The objective of current study is to evaluate the feasibility of using a locally available natural material, Iraqi bentonite, to adsorb copper ions from the refinery company's industrial wastewater. Identify the optimal conditions for removing copper, which are pH, dose of adsorbent, contact time, and concentration of the adsorbed ion.

Materials and methods

Study site

The study was conducted at Tikrit University, College of Education laboratories and the Central Laboratories Department. Samples were taken from the wastewater collection area in Qayyarah refinery, as shown in Fig. 1.

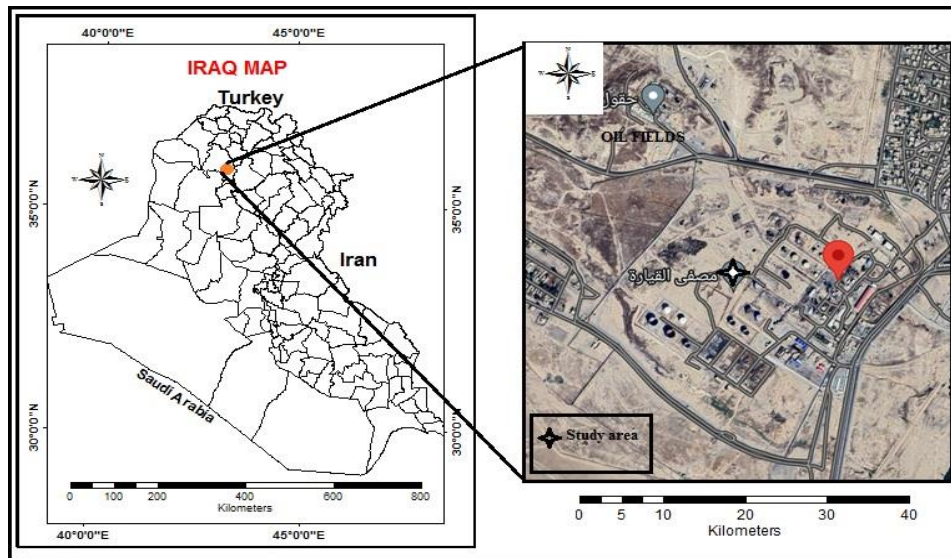


Fig.1 Location of Qayyarah refinery

In order to preserve the properties of the sample until laboratory tests were conducted on it, it was stored in the laboratory at a temperature of 4°C. The preliminary treatment processes for wastewater included the following:

- 1- Filtering wastewater from suspended particles using filter paper with a pore size of (0.45 μm), as in the picture (3-2).
- 2- Digest the filtered wastewater by adding (1 mL) of concentrated nitric acid to each (50 mL) of sample water.



Fig. 2 Wastewater collection area

The concentration of copper was estimated using the Porphyrin Method. This method is considered one of the best and most accurate methods used to estimate copper concentration, as this method is not affected by interfering substances and does not require an extraction or concentration process for the sample before analysis. Work is done to remove the interfering

substances using the Copper Masking reagent, where the porphyrin index produces a yellow complex with copper ions and then measures at a wavelength of 425 nm a, 1979.

Materials and method

Using the program (145 Copper, Porphyrin) for the determination of copper, the sample is prepared by filling the cell with (10mL) of the sample, then adding the contents of the bag of reagent (Copper Masking), then shaking the cell well and leaving it aside to complete the dissolution of the reagent. Also, another cell is prepared containing (10mL) of the sample and the contents of one bag of Porphyrin 1 Reagent Powder are added to it for each cell. The two cells are shaken well and the second reagent (Porphyrin 2 Reagent Powder) is added to them, one bag for each cell, then the two cells are shaken well. They were left for (3 minutes) until a pink color appeared for a short time and turned to yellow if copper was present in the sample. The spectrometer was reset using a blank sample and the copper concentration was recorded in units (μg/L).

Preparation of Iraqi nano-bentonite

The Iraqi bentonite clay was ground using a dry grinding method using a Silver Crest-Germany electric grinder for four hours (divided into 10 minutes × 24 grinding times), the reason for this is the high temperature inside the mill, largely as a result of the friction of the bentonite with the inner surface of the mill. Picture (3-13) shows Iraqi bentonite before and after grinding. Bentonite nanoparticles were manufactured using the co-precipitation method. After the process of grinding the bentonite and obtaining the powder, the bentonite powder is sieved using a sieve with a mesh size of (75 μm), and (20 g) of the bentonite powder is dissolved in (100 mL) of hydrochloric acid (12 molar), then the solution is heated and mixed. Using a magnetic stirrer at a temperature of (70°C) for

two hours at a speed of (300 rpm). The solution is then washed with distilled water several times until the pH becomes neutral. The solution is to dry in the oven at a temperature of (100°C) for 5 hours. After drying, the product is crushed using a mortar several times to obtain bentonite powder. The calcination process is then performed at a temperature of (600°C) for one hour, which aims to eliminate the chlorine bond (Cl-bound) on the bentonite and thus obtain a product in the form of bentonite nanoparticles. Finally, the powder is ground using an electric grinder at a speed of (100 rpm) for 30 minutes and then sieved (Hashemian et al., 2015; Sirait and Manalu, 2018).

Sirait and Manalu, (2018) examined the crystal structure of ground bentonite with the addition of hydrochloric acid by X-ray diffraction technique, which showed the hexagonal structure of bentonite with a diameter size of (19.17 nm). The acidic reactions of soluble solvents such as hydrochloric acid lead to increased openness of the pores so that the surface area of the bentonite nanoparticle becomes larger, and the smaller the adsorbed pore size, the greater the surface area and thus the greater the number of particles (Bath et al., 2012).

Treatment of copper

Copper (Cu) content was determined using a visible spectrophotometer (HACH DR3900). This metal was selected since there were no prior studies including it at the research location, and it is also one of the most prevalent elements found in the wastewater produced by Northern Refineries Company. For the second station, located upstream from the

mouth of the canal in the Tigris River, where the adsorption process occurred under the influence of conditions (dose of the adsorbent, pH, ion concentration, contact time), a copper ion solution was prepared with the following proportions: (0.05, 0.09, 0.08, 0.07) mg/L. These concentrations were chosen because they represent the same concentrations present in the study samples during the study period. After closing the nozzle of each beaker with Parafilm, the adsorbate (prepared ion solution) for copper was studied, and the beakers were placed on the shaker for varying amounts of time (5, 15, 25, 30) and mixing speed (180 rpm), after which the solid adsorbent materials were separated by filtering the mixtures through filter paper (0.45 m). The resultant extract is then collected and put through a visual spectrophotometer (Wu & Maskaly, 2017). The following tables display the results of a series of tests performed at room temperature; these results reveal a significant amount of the element of the study was removed from the device readings.

An off-the-shelf program (Statistical Package for the Social Sciences, SPSS-USA-Version-21) was used to do statistical analysis on the copper collected for the research. To assess statistically significant variations between the means of copper values, we used analysis of variance with Duncan's Multiple Range Test, setting the significance level at P 0.05.

Results and Discussion

Results of copper treatment

Table 1. Shows the removal percentage with the change in pH for the adsorption process of copper ions on the surface of Bentonite

No.	Element	pH	Adsorbent Dose (mg/L)	Contact time (min)	Prepared solution (mg/L)	Concentration after treatment	Removal percentage %
1	Copper	4	50	5	0.05	0.04	20%
2	Copper	5	50	5	0.05	0.034	32%
3	Copper	6	50	5	0.05	0.024	52%
4	Copper	9	50	5	0.05	0.0285	43%

Table 2. Shows the removal percentage with changes in the dose of the adsorbent for the adsorption process of copper ions on the surface of Bentonite

No.	Element	pH	Adsorbent Dose (mg/L)	Contact time (min)	Prepared solution (mg/L)	Concentration after treatment	Removal percentage %
1	Copper	6	100	5	0.05	0.022	56%
2	Copper	6	200	5	0.05	0.0165	67%
3	Copper	6	250	5	0.05	0.0115	77%

Table 3. Shows the removal percentage with changes in the concentration of ions of the element adsorbed on the surface of Bentonite

No.	Element	pH	Adsorbent Dose (mg/L)	Contact time (min)	Prepared solution (mg/L)	Concentration after treatment	Removal percentage %
1	Copper	6	250	5	0.09	0.0189	79%
2	Copper	6	250	5	0.08	0.0128	84%
3	Copper	6	250	5	0.07	0.0133	81%

Table 4. Shows the removal percentage with change in contact time for the adsorption process of copper ions on the surface of Bentonite

No.	Element	pH	Adsorbent Dose (mg/L)	Contact time (min)	Prepared solution (mg/L)	Concentration after treatment	Removal percentage %
1	Copper	6	250	15	0.08	0.0112	86%
2	Copper	6	250	25	0.08	0.0096	88%
3	Copper	6	250	30	0.08	0.0048	94%

Optimal conditions for removal

The prepared adsorbate, Bentonite, achieved high efficiency in removing copper at the optimal pH = (6) and at the optimal dose of the adsorbent (250 mg/L), the concentration of the adsorbate (the prepared ion solution) (0.08 mg/L), and the contact time (30 min). The removal percentage of the copper element was 94% under the influence of these conditions. The results of the current study showed that the concentrations of the copper element ranged between (0.2-0.023 mg/L) in the industrial wastewater at Qayyarah refinery.

Copper affects the nervous and digestive systems, the kidneys, the liver, and hemolysis. Copper ions interact with inorganic ligands, forming complexes that are absorbed from insoluble particles in the aqueous solution. Therefore, their presence in water in high concentrations reduces the concentrations of inorganic ions, which are an essential component of total dissolved salts and total hardness. This is reflected in the electrical conductivity, where there is a direct relationship between them, and this is confirmed by the results of statistical analysis. As it is known, living organisms need copper in small quantities, so its presence in the soil is a pollutant and has negative effects on groundwater, it has a strong effect and relationship with dissolved organic carbon, and there is a positive relationship between organic carbon and copper concentrations and reduces the amount of dissolved oxygen in water as a result of decomposition reactions of organic materials used in oxidation. It has an effective impact on acidity, electrical conductivity, and temperatures. The results of this study agree with the findings of the scientists that copper ions bind to polar organic compounds containing carboxyl, amine, or hydroxide groups to form complexes dissolved in water, unlike non-polarized organic compounds that do not tend to bind to copper ions. Therefore, the

percentage of copper decreases in the presence of petroleum-based organic compounds, and this is statistically consistent. Adsorption is an exothermic reaction, and the process of adsorption of heavy elements reduces the temperature rise, and the optimum temperature at which the highest adsorption occurs is that the temperature rises to the optimum temperature at which the highest adsorption occurs, and is due to the fact that temperatures help to increase the kinetic energy of the adsorbate molecules, which helps in the separation of these molecules from the surface of the adsorbent, as well as the contact time between the adsorbent and adsorbate, and the optimal adsorption time is that the percentage of removal of the copper element increases with increasing contact time and then remains constant until equilibrium is achieved.

Based on the interaction of light energy with metal nanoparticles, this treatment has become of great interest due to its high and broad photocatalytic activities. Photocatalytic activities are based on the destruction of bacteria and organic materials by reacting with hydroxyl radicals. The materials used in nano catalysts are usually inorganic materials such as semiconductors and metal oxides. However, to be considered a nano photocatalyst, they must follow certain requirements to be harmless and have their concentration in water and air below the maximum permissible value for the formation of agglomerations, coagulation, and sedimentation and for the formation of safe normal particles. Nanomembranes are responsible for separating particles from wastewater. They can be very effective in removing dyes, heavy metals, and other contaminants. The nanomaterials used as nanomembranes are nanotubes, nanoribbons, and nanofibers. Conventional water treatment sometimes cannot be very effective in removing some contaminants, such as metals and microorganisms. Another problem is the formation of disinfection by-products (DPBs), which pose a risk to human health. DPBs result from

the reaction of chemical disinfectants with organic substances and inorganic ions present in water, which are added to the secondary effluent in order to prevent bioindication. Antimicrobial nanomaterials, for example, do not typically produce DPBs because they are not strong oxidants, so they can be important disinfectants and improve water treatment.

Access to drinking water is an important factor in preventing people from contracting water-borne diseases, which are the leading cause of death worldwide, especially in developing countries. Diarrhea, as a result of this condition, kills approximately 2.2 million people every year and particularly affects children under the age of five. In the absence of safe drinking water, many health problems can occur and lead to health and economic problems. Some studies evaluate the ability of nanomaterials to remove pollutants from water. Nanomaterials have been studied as an alternative to removing metals, microorganisms, and oils from polluted water. Nanomaterials have also been developed for water desalination as a possibility to use seawater as a drinking source. Heavy metals and some ions have high toxicity, and this property can cause real damage to humans and ecosystems, the magnetic nanofluids act as a coagulant in water and as a potential absorber of heavy metals. The results showed that the metal removal efficiency increased to nearly 100% when the pH was also increased from 4 to 8, these heavy metals were Cu^{+2} (Hatamie et al., 2016).

Copper nanomaterials can be used in water treatment, there are two different applications for that: using copper nanomaterials as a type of absorbent material and using them as a photocatalyst that converts pollutants into less toxic substances. Carbon-based materials are used as adsorbates, e.g., carbon nanotubes, and have a high surface area to volume ratio and pore size distribution. With these properties, it offers an important adsorption capacity compared to granular or powdered carbon. Another carbon-based material is graphene, as they are considered effective absorbers due to their large specific surface area and electron-rich environment. Graphene oxide has also shown high absorption capacity due to its strong functional groups. Bentonite is composed mainly of silica (60-70%), aluminum (15-20%), and a small percentage of iron. Bentonite consists mainly of the highly crystalline mineral Montmorillonite, with a percentage ranging between (55-85%) (Al-Ghanimi and Al-Zubaidi, 2020). Each particle consists of an eight-sided aluminum plate sandwiched between two tetrahedral octahedral silica sheets, meaning it consists of layers (1:2), and the chemical formula of montmorillonite is $[\text{Mg}_2 \text{Al}_{10} \text{Si}_{60} (\text{OH})_{12} (\text{Na}, \text{Ca})]$. Montmorillonite belongs to the smectite group, the main smectite minerals are: Calcium montmorillonite, Sodium montmorillonite, Saponite which is Magnesium montmorillonite, Nontronite which is Iron montmorillonite, Beidellite which is aluminum montmorillonite. The most common are calcium and sodium montmorillonite. Calcium montmorillonite can be found in many parts of the world while sodium montmorillonite is relatively rare. Between the three layers of montmorillonite, there are water molecules that can be repelled without

destroying the internal atomic structure of this mineral when heated to a temperature not exceeding 100°C . The montmorillonite crystal is negatively charged due to the substitutions in the octahedral layer, as magnesium ions (Mg^{+2}) are replaced by aluminum ions (Al^{+3}), and (Al^{+3}) ions are replaced by silicon ions (Si^{+4}) in the tetrahedral layer, which is why the bentonite crystal is negatively charged, and this negative charge is balanced by replaceable positive ions. The reason for researchers' interest in the mineral montmorillonite is that this mineral possesses a number of physical and chemical properties that are not found in other minerals (Viggori and Hellata, 2003). These properties are: it has a fine, granular size, diversity in its internal chemical composition, diversity in the type of replaceable ions and surface charge, and the structural characteristics of this mineral. The mineral can react with organic and inorganic solvents, has a large and chemically effective surface area, and also has a large ion exchange capacity. In addition to montmorillonite, bentonite contains other minerals such as quartz, calcite, feldspar, and biotite.

One of the main reasons why bentonite has been preferred for various applications is because of the effectiveness and low cost of these clays, and the availability of these clays in various parts of the world. Among the common uses of bentonite are the following:

Bentonite is used as a good adsorbent for removing pesticides, herbicides and heavy metals from aqueous media (Nwosu et al., 2018). Bentonite has also been used as a drilling fluid for many years. Drilling mud must have a high viscosity and thixotropic property, which prevents the sedimentation of fine materials and the freezing of cuttings during drilling oil wells. It is used as a bleaching agent for oils through adsorption to remove some dyes and colors. Calcium bentonite is also used in catalytic cracking of petroleum in petrochemical processing, while sodium bentonite is used in drying oils (Xavier et al., 2014).

Bentonite is a pale-yellow substance that does not dissolve in water, organic or inorganic acids, or bases. Bentonite has good adsorption capacity, has many physical and chemical properties, and is one of the minerals abundantly available on Earth (Vickers, 2017). The weak link between bentonite molecules enables water to readily enter between them, which in turn causes the molecules to move apart and contributes to bentonite's extraordinary capacity for swelling by as much as a factor of eight. Bentonite's hydration is significantly influenced by where the 1:2 symmetric substitution occurs (i.e., whether the layer charge is obtained from substitution in the tetrahedral or octahedral layer). The hydration energy of the interlayer cation, the polarisation of water molecules by the interlayer cations, variations in electrostatic surface potentials due to differences in the interlayer charge sites, water activity, and the size and shape of bentonite particles are the primary factors that influence the hydration of the bentonite interlayer. Charge, hydration energy, and exchange cation size play major roles in regulating water uptake and dilution.

The rate and extent of bloating are improved by the small particle size, high surface area, and porosity provided by bentonite. When bentonite is fully hydrated, the tortuous flow path in the clay gel creates a unique thermodynamically stable hydrated state for suspension of most interlayer contamination by reacting with surfaces. In many of its uses, bentonite mineral's cation exchange and exchange reaction are crucial. Between the layers of silicate exist a variety of exchangeable cations, including sodium, calcium, and magnesium. In contrast to sodium bentonite, which has a cation exchange capacity of 80-130 mEq/100g, calcium bentonite's range is only 40-70 mEq/100g. Bentonite, in its typical form, is comprised of extremely thin, viscous, thixotropic particles that boast a very high bloating capacity, specific surface area, adsorption capacity, layer charge and exchange capacity, and adsorption capacity. To be more specific, sodium bentonite generally displays strong adsorption, bonding strength, and bleaching capacity, whereas calcium bentonite typically demonstrates high viscosity, dispersibility, bloating ability, and thixotropy. The following explains why bentonite is effective at adsorbing heavy metals: it has a porous interlayer, a high bloating capacity, a massive surface area, and a high ability to exchange positive ions. They form a weak connection with the bentonite molecules, which is another drawback of these positive ions. Since this is the case, additional positive ions may easily take their place (Alexander et al., 2019).

Conclusion

In this study, the feasibility of using locally available bentonite as an adsorbent for the removal of copper from industrial wastewater in the Qayyarah refinery was successfully evaluated. The research focused on optimizing key operational parameters, including pH, adsorbent dosage, contact time, and the concentration of the adsorbed copper ions. The results indicate that bentonite exhibited high effectiveness in removing copper from the refinery's wastewater. The following optimized conditions were identified for efficient copper removal: a pH of 6, an adsorbent dosage of 250 mg/L, a concentration of the adsorbed ion at 0.08 mg/L, and a contact duration of 30 minutes. Under these optimized conditions, an impressive 94% of copper was extracted from the wastewater, highlighting the excellent copper adsorption capacity of bentonite.

Furthermore, the analysis of industrial effluent from the Qayyarah refinery revealed a wide variation in copper concentrations, ranging from 0.2 to 0.023 mg/L. This variation underscores the importance of efficient copper removal techniques to meet regulatory standards and reduce the environmental impact of industrial wastewater discharges. This study demonstrates the potential of locally available bentonite as an effective and sustainable solution for copper removal from industrial wastewater. The optimized conditions can serve as a practical guideline for the implementation of bentonite-based adsorption systems in refinery wastewater treatment processes, contributing to environmental protection and compliance with water quality regulations.

Conflict of Interest

The author hereby declares no conflict of interest.

Consent for publication

The author declares that the work has consent for publication.

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