University of Tehran Press

Pollution

Print ISSN: 2383-451X
Online ISSN: 2383-4501

https://jpoll.ut.ac.ir/

Removal of Cd(II) Ions from Aqueous Solutions using adsorption By Bentonite Clay and Study the Adsorption Thermodynamics

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Article Info

Article type:
Research Article

Article history:
Received: 30 Dec 2022
Revised: 18 Feb 2023
Accepted: 06 May 2023

Keywords:
Adsorption
Bentonite
Cd(II) ions
Langmuir and Freundlich models
Thermodynamic

ABSTRACT

cadmium usually enter the environment and water resources through wastewater, released by various industries, and may have adverse effects. The current study employs surface of bentonite clay available locally in order to remove cadmium In solutions contaminated with this type of ions, in order to research on a surface with a high ability to adsorption of cadmium (II) ions, study Some factors affect the adsorption process on bentonite clay, such as contact time, pH the solution, Adsorbent particle size, Initial concentration of solutions and temperature of the solution were examined in the a batch process mode. The amount of adsorbed Cd (II) increased with height temperature, the optimum adsorption pH was about 6.5. Under this condition, the percent removal was 95.17%. The adsorption isotherms were studied and the results of adsorption processes were more fitted with Friendlich model rather than Langmuir adsorption model. Thermodynamic study showed that, ΔH was endothermic, ΔG is found to be negative That is, the process is automatic and ΔS was found to be positive. The current study also involves practical application using bentonite to get rid of Cd(II) ions to from wastewater of Hamdan's station of the Basra- iraq, The results indicate high affinity (97.84%) removal of Cd(II) ions.

Cite this article: Y. Hammood, A., Kassim Mohammed, I., and A. majeed, A. (2023). Removal of Cd(II) Ions from Aqueous Solutions using adsorption By Bentonite Clay and Study the Adsorption Thermodynamics. *Pollution*, 9 (3), 994-1005. https://doi.org/10.22059/poll.2023.353137.1741

DOI: https://doi.org/10.22059/poll.2023.353137.1741

INTRODUCTION

The main objective for the Wastewater Pollution Treatment in all sectors of industry, reconnoitring, agriculture and waste treatmentis the reduction of toxicity such as Organic compounds, Dyes and heavy metal. Many industrial Operations in the coating industry, heavy metals for metalworking and their wastewater must be treated before unloaded (Kurniawan *et al*., 2006; Karami *et al*., 2021; Orooji *et al*., 2021). Some of these metals are recognized to be toxic such as cadmium, mercury ,copper, chromium, lead, selenium and silver. When this metal reaches the environment, it will make up a threat to human health (Rao *et al*., 2010). Heavy metals are considered dangerous and toxic environmental pollutants, as they are stable and do not decompose, and this leads to their accumulation in microorganisms, plants and aquatic organisms, which in turn are transmitted to humans through the food chain and thus lead to multiple health problems for humans (Briffa *et al*., 2020). Cadmium is one of the most toxic elements and is not necessary for the life of living organisms. It is found in low concentrations in natural conditions, but human activities have led to an increase in its levels in all continents. Cadmium contamination

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of water and food causes serious health problems for humans, including impaired kidney functions and osteomalacia as a result of a disturbance in the calcium cycle in the body, as well as anemia, high blood pressure and an enlarged heart (Javadian *et al*., 2015). In the last years, increasing consciousness of the environmental effect of heavy metals has led to a request for the purify of industrial wastewater before to release into natural waters. This has resulted in the introduction of more stringent water pollution dominance legislation (Waly *et al*., 2010).

Processes of treating organic and inorganic pollutants from the aquatic environment include many techniques sedimentation, chemical oxidation, coagulation, reverse osmosis, ion exchange, photocatalysis and other technologies. Natural materials or low-cost materials produced through agricultural and industrial processes can be considered as low-cost adsorbents and can be used to remove heavy elements, It is necessary to compare the adsorbent materials in terms of cost. The adsorption method is a simple and inexpensive technique legislation (Khulbe and Matsuura, 2018; Ghanbariand and Salavati-Niasari, 2021). The Most common method for Cd (II), removal from industrial wastewater are natural materials were also reported as potential industrial adsorption media (Kareem *et al*.,2008; Mahmoud, *et al*. 2021), activated carbons (Abbas, 2020; Lakshmikandhan and Ramadevi, 2019) and also clay as adsorbing surface for the To get rid of Cd (II) from contaminated water (Jing *et al*.,2020; Galindo, et al. 2013).

the present study assessed of using bentonite clay as an adsorbent to remove Cd (II) ions, it examined the cadmium removal percentage from synthetic samples. Herein different adsorption parameters would be

investigated including effect of cadmium concentrations, of pH, adsorbent amount, contact time, and primary cadmium concentration . All of the adsorption tests were implemented by batch adsorption system at room temperature. Further, an atomic absorption spectrometer. Then, Langmuir and Freundlich's isotherms were compared. The thermodynamic parameters of adsorption were calculated by using Van't Hoff 's equations. This study primarily aimed to provide a method for cadmium ion adsorption from samples using bentonite clay as the adsorbents.

EXPERIMENTAL SECTION

Preparation of the adsorbent surface (Bentonite)

Natural Bentonite clay models were obtained from the Ministry of Industry and Minerals , General Company for Geological Survey and Mining in Baghdad, Iraq. First, the models were washed to get rid of dirt using deionized distilled water, The models were dried using an oven at120 °C for 3 h, Grinding the powder for the purpose of obtaining fine particles by means of an electric grinder into fine particles and sifting the ground powder using laboratory sieves of different sizes ranging from 75 to 300 μm, and finally the sample powder was kept in containers to be used as an experiment primer.

Chemical reagent

Metal salts used was from the facility (merk). Standard solutions were prepared of Cd (II) ions with concentrations within the range (0.02 - 2.20 mg/L) these concentrations areproportional to the sensitivity of Flame Atomic Absorption Spectrophotometer (FAAS) used for measurement.

Adsorption studies

The batch method was used to study the adsorption process. The adsorption was performed using 100 ml conical stopper flasks by adding 50 mL From Cd(II) ions at a concentration 20 mg/L to 0.5 g of bentonite powder with a grain size of 75 μm. Experiments were carried out at laboratory temperature with shaking for the required time at 120 rpm. Samples were separated by a centrifuge. The models were measured in the solutions were estimated by a flame atomic absorption spectrophotometer (GBC SCINTIFIC EQUIPMNT SensAA). Equations No.)1(and)2(were used to find the removal ratio and the adsorption capacity by calculating the concentrations of ions in the solution before and after the adsorption (Jain *et al*., 2019).

$$Qe = V(C_0 - C_e)/m \qquad (1)$$

$$removal\ \% = [(C_0 - Ce)/C_0] \times 100 \qquad (2)$$

Where; Qe is the adsorption capacity (mg/g), V is the total volume of adsorbate (L), m is the adsorbent weight (g), C_0 is the Initial concentration of the adsorbate solution in mg.L^{-1}, Ce is the concentration of of the solution adsorbate in mg.L^{-1} after adsorption.

RESULTS AND DISCUSSION

FT-IR spectroscopy

Aquantity of bentonite powder was taken and measured with an infrared spectrometer (FTIR) type shimadzu (4000-400 cm- 1) for the purpose of diagnosing the functional groups present in the composition of bentonite powder The FTIR spectra as explains in Fig (1) We notice the emergence of strong band in the range (3621-3400 cm^{-1}) resulting from tretching vibration of (OH) group, while group (si = o) showed the stretching vibration at the site1033.85cm^{-1}, the band at 530.42 cm^{-1} can be attributed to the stretching vibration Fe–O group and the band around 796.60cm^{-1} vibration is attributed to the stretching of the Al–O group (Silverstein *et al*., 2005).

Effect of contact time

Determine the contact time necessary to reach the equilibrium state at different time has been studied in the range from (5-180min) at 25°C. Figure (2) shows an a increment in the percentage of removal with the time passing before Access to equilibrium state. The rate of ions removal was higher in the beginning of the first minutes of adsorption, due to the availability of large surface area on the surface of bentonite prepped to the adsorption of elemental ionsand then the increase was turned slowly until reaching saturation state. This is due to occupancy or filling of the adsorbent molecules of the most effective sites for adsorption on the surfaces of Bentonite (Hammood *et al*.,2021; Pehlivan, *et al*. 2009). Equilibrium time was attained at 45min. The removal percentage of ions on bentonite powder was worth 95.19% and the value of the equilibrium adsorption capacity (1.90mg/g).

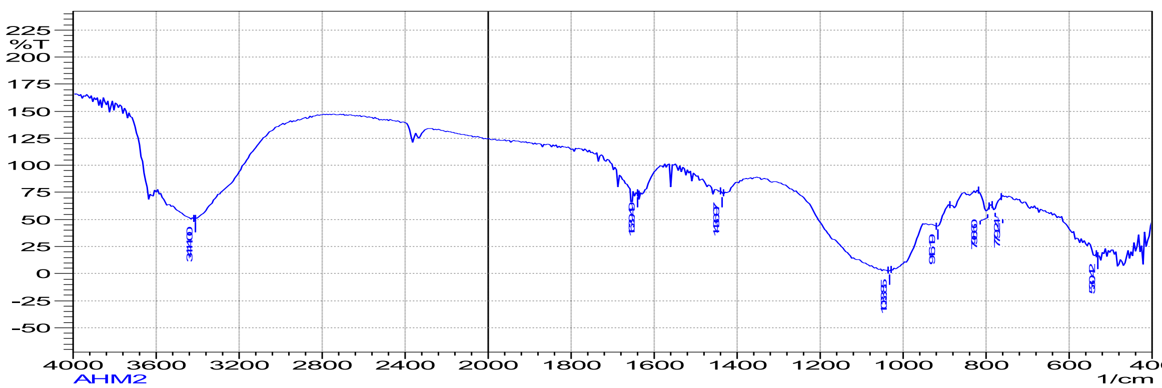

Fig. 1. FT- IR spectrum of Bentonite

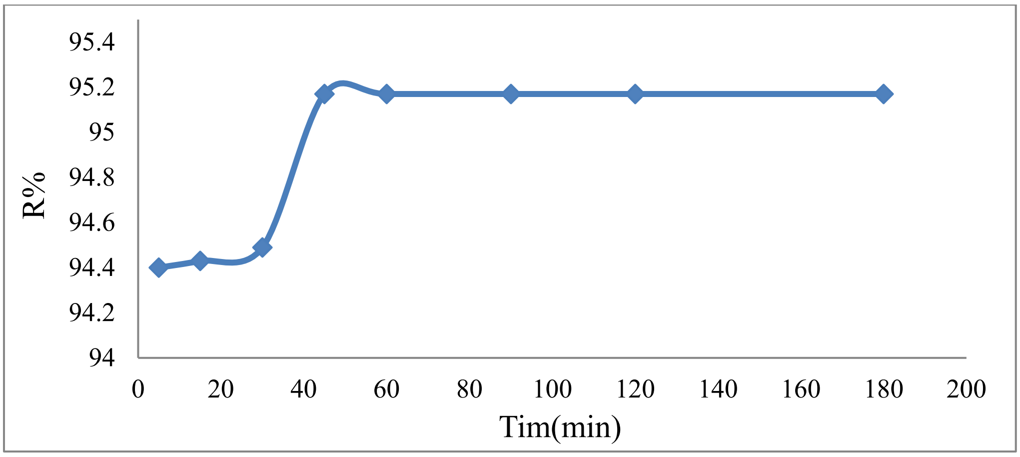

Fig. 2. Effect of contact time on adsorption of Cd(II) ion on to Bentonite

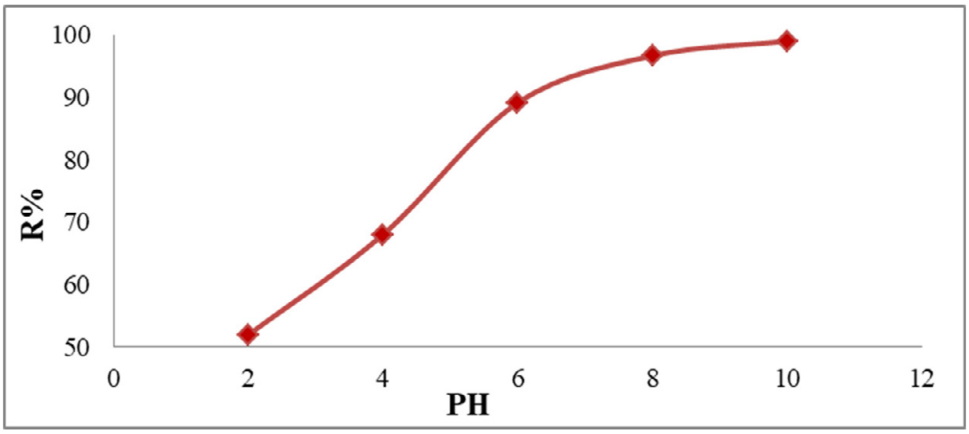

Fig. 3. Effect of pH on Cd(II) adsorption on Bentonite

Effect of pH

The Changing the pH value has effect on the adsorption process when its value changes, it may lead to an increase or decrease in the amount of adsorption or remain unchanged. Lots of variables can be involved in this process, such as the chemical condition of the adsorbate, the adsorbent surface, and the solvent (Adamson and Gast, 2001). The change of the pH value was studied within the range (2, 4, 6, 8 and 10) using (20 mg/L) from Cd(II) ions and contact time of (45 min) and warmly 25°C, Figure (3) shows the changing the pH .

From the above figure, we notice an rise in the percentage removal with to rise the pH. At low pH values there is an abundance of hydrogen ions (H+) will vie with Cd(II) ions On the sites of negative charges on the adsorbent surface, then the percentage of removal decreases, on the contrary, high pH values will lead to a reduction the hydrogen (H+) ions in the solution, thus reducing competition with Cd(II) ions on the negative charge sites available on surface of Bentonite and favoring the adsorption of cations on the surface, which raises values the percentage of removal, on the other hand, the increase in the pH may change the nature of the surface towards inhibiting its activity and then a decrease in the adsorption of ions accordingly (Lund, 1994), All study experiments were performed at pH 6.5 to avert interference of the sedimentation process with the adsorption process, which may play a role in the ion adsorption process.

Adsorbent particle size

Three different adsorbent particle sizes were used to study the adsorption (75, 120, and 300 µm) at constant concentrations and temperature (25°C).

Shows Figure (4) that the adsorption percentage rise with decreasing size of the adsorbent particles, Where we note that at a size of (75 µm) the percentage of removal is (95.17%), while The percentage of ion removal decreased at a minute size (300 µm) of the adsorbent material (92.84%).

The main reason for the rise in the percentage of adsorption when the reduction in the size of the particles of the adsorbent material bentonite is due to the fact that the reduction in the size of the particles leads to an increase in the surface area available for adsorption, therefore increase in the active sites on which adsorption occurs (Zouboulis *et al*., 2002).

Effect of initial concentration of Cd(II) ions

The change in the concentration of Cd(II) ions was studied using a concentration in the range (5-40 mg/L) at a temperature of (25 °C) as shown in Figures (5).

It was found through the results that when the concentrations decrease, the percentage of removal increases, Where the percentage of removal was (93.28%) when concentration of (40mg/l), and these percentages increased to be (97.02%) when using concentration of (5mg/l).

The main reason for this increase in the removal percentage with a decrease in concentration isreturns to the high values of the initial concentration contain relatively large numbers of ions of the elements under study , thus occupy the largest number of active sites on Bentonite, which

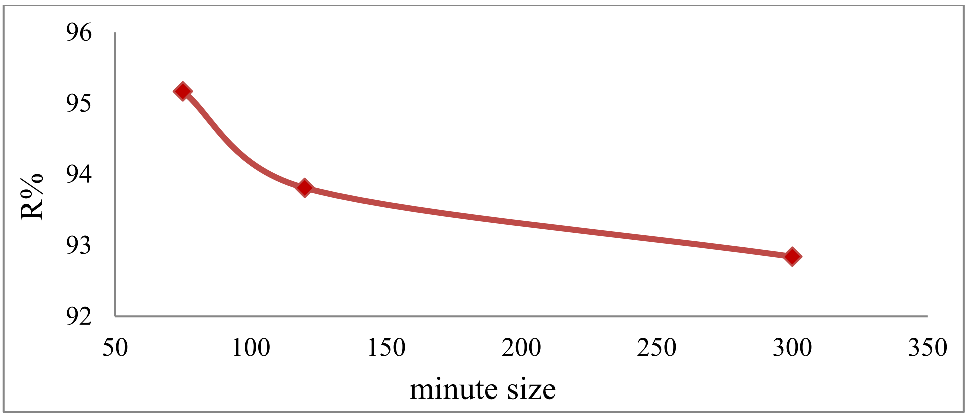

Fig. 4. Relationship of the percentage of removal to the size of the adsorbent particle.

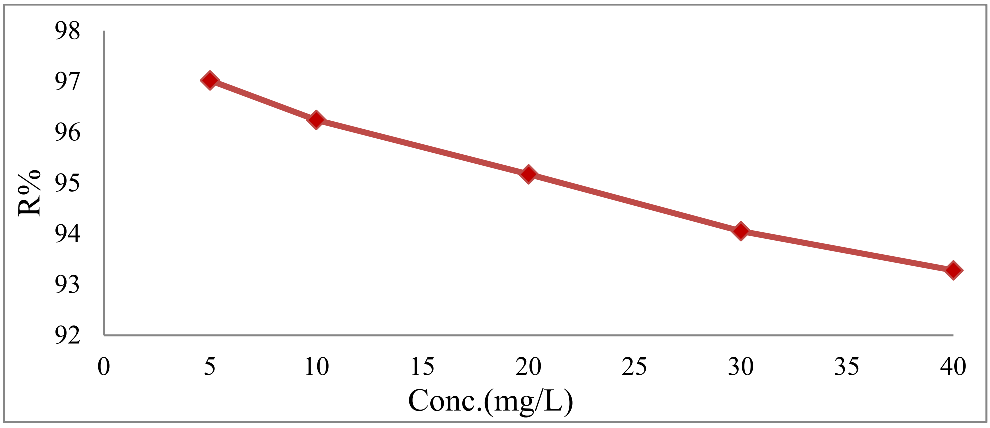

Fig. 5. Effect of initial concentration on adsorption of Cd(II)

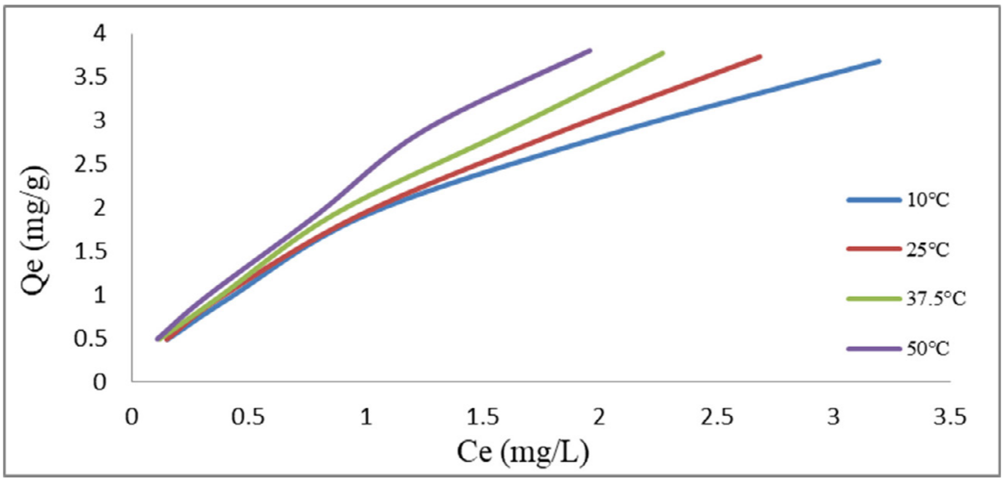

Fig. 6. Adsorption isotherm of Cd(II) on Bentonite surface at different temperatures

Table 1. Results of Freundlich and Langmuir adsorption isotherms

	Freundlich constants			Langmuir constants		
Temp. (°C)	**n**	**Kf**	**R^2**	**Q_m (mg/g)**	**b (L/mg)**	**R^2**
10.0	1.483	1.758	0.996	5.734	0.522	0.976
25.0	1.418	1.890	0.999	6.312	0.491	0.963
37.5	1.432	2.082	0.997	6.538	0.533	0.897
50.0	1.385	2.341	0.997	7.069	0.552	0.891

makes the percentage of adsorbed ions less than the remaining free ions in the solution, but in the case of low-concentration solutions, the number of ions dispersed in the aqueous solution is equivalent or approximately equal to the active groups present on the adsorbing surface, so the adsorption percentage increases (Hmood and Jassim, 2013).

Adsorption isotherm

The adsorption was studied at temperatures (10.0, 25.0, 37.5, 50°C), and a graphic relationship was drawn between the amount of the adsorbent substance (Qe) and the concentration at equilibrium (Ce) to get the general shape of the adsorption isotherms, as shown in Figures (6) .

We notice from Figures (6) that the general form of isotherms at different temperatures studied indicates that they are of class (L-type) according to (Giles) classification (Giles *et al.*, 1960).

The results of the study of the effect of temperature were used in applying the mathematical equation isotherm Freundlich and Langmuir for adsorption,Equation (2) represents Freundlich adsorption equation, while equation (3) represents Langmuir adsorption equation (Babadi *et al.*, 2018). The Freundlich and Lanckmeier empirical constants and correlation coefficient (R^2) shown in Table (1)

were calculated.

$$L0g\ Qe = log\ Kf + 1/n\ l0g\ Ce \qquad (2)$$

Where Kf is Friendlich constant and n is A fixed value (rank) ranging from (2-8) and Relies on the type of adsorbent surface, the nature of the adsorbate and the temperature.

$$Ce/Qe = 1/Qmb + Ce/Qm \qquad (3)$$

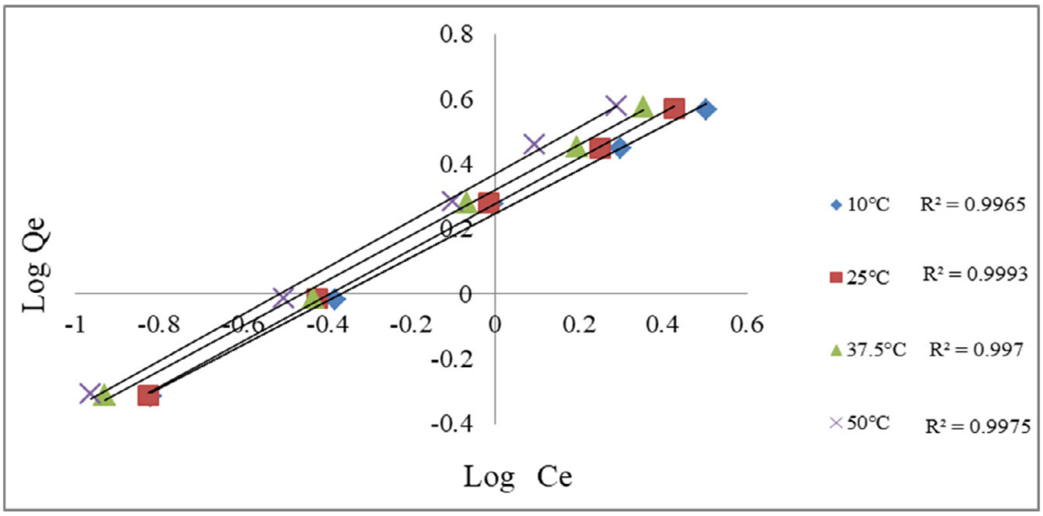

Fig. 7. Freundlich adsoprion isotherms for adsorpion of Cd(II) over Bentonite at different temperatures

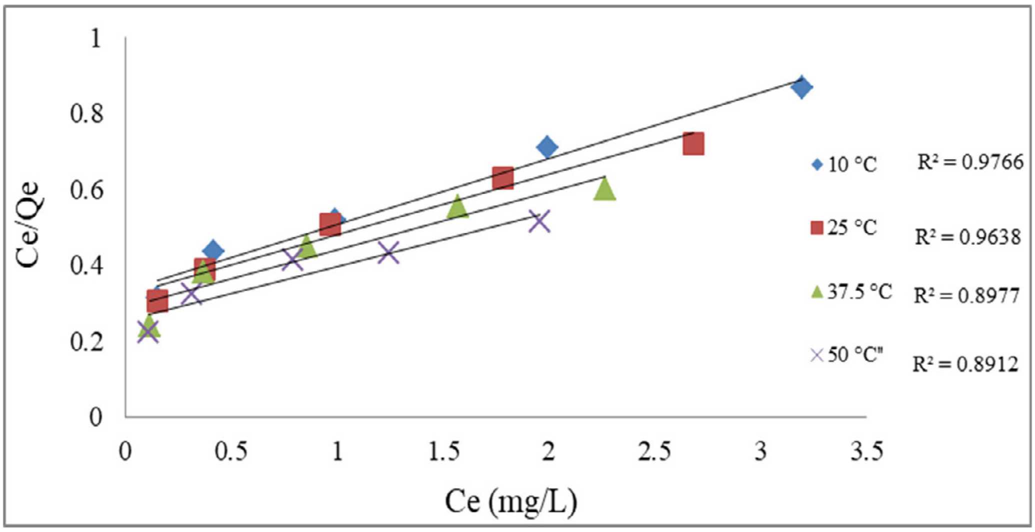

Fig. 8. Langmuir adsorption isotherms for adsorpion of Cd(II) over Bentonite at different temperatures

Where Q_m is the maximum adsorption capacity when the adsorption surface is completely saturated (mg/g) and b is The Langmuir constant is related to the adsorption energy or the equilibrium constant.

Through the values of (R2) shown in the above table, it becomes clear that the Freundlich equation is the most applicable to the adsorption process because there is a linear relationship between the values of *LogQe* against the *Log Ce* as shown in the tables (1) and Figures (7), while the results showed the exclusion of the Langmuir equation for the absence of a linear relationship when plotting the *(Ce/Qe)* versus *(Ce)* values as Appear in Table (1) and Figure (8).

Table(2) summarizes the results of other similar studies. As shown, The maximum adsorption capacities calculated by the Langmuir model were 7.069mg/g.The results were desirable and acceptable compared to the other research, which is somewhat similar to the results of Ebrahim and Mohammed(2012) and Sepehr and Tosan(2016). The results also indicate the possibility of removing cadmium from aqueous solutions using Bentonite clay.

Table 2. Comparison between the maximum cadmium adsorption by various adsorbents

Type of adsorbent	Heavy metals	Maximum adsorption Capacity (mg. g^{-1})	Absorption conditions	References
Activated Carbon		3.002	pH=4	
Kaolinite	Cadmium	1.837	C_0=60mg L^{-1}	(Jenan et al.,2010)
			time=24hr	
	Chromium (III)	0.704	pH=3	
red Kaolin	Iron (III)	1.453	C_0=50mg L^{-1}	(Khdeem ,2010)
	Cadmium	1.686	time=120min	
	Lead	2.90	pH=4.9	
Activated biocarbon	Cadmium	2.88	C_0=100 mg L^{-1}	(Singanan ,2011)
			time=48hr	
			pH=6	(Ebrahim &
Rice Husk	Cadmium	7.692	C_0=25mg L^{-1}	Mohammed,2012)
			time=100min	
			pH=6	
Iraqi date pit	Cadmium	1.57	C_0=40mg L^{-1}	(Gyaath ,2012)
			time=24hr	
	Cadmium		pH=6	(Sepehr &
Grape pruning residues		7.895	C_0=11mg L^{-1}	Tosan,2016)
			time=24hr	
	Mercury	107.75	pH=8	(Javidi Alsadi. &
sugarcane bagasse-activated carbon	Lead	0.625	C_0=5mg L^{-1}	and Esfandiari
	Cadmium	2.425	time=3hr	2019)
	Cadmium	5.12	pH=5	
natural clay	Copper	13.16	C_0=100mg L^{-1}	(Abbou et al.,2021)
	Lead	15.70	time=120min	
	Cadmium	4.34	pH=6	(Es-Said et
natural clay	Copper	4.65	C_0=200mg L^{-1}	al.,2023)
	zinc	4.47	time= 26min	
			pH=6.5	
Present study	Cadmium	7.069	C_0=20mg L^{-1}	-------
			time=45min	

Table 3. Thermodynamic functions for adsorption of Cd(II) ion

Co (mg/L)	K				- ΔG				ΔH	ΔS
	Temperature									
	283	*298*	*310.5*	*323*	*283*	*298*	*310.5*	*323*		
5	31.894	32.557	41.372	44.871	7.943	8.738	9.400	10.063	7.056	0.053
10	23.038	25.595	26.100	30.746	7.155	7.800	8.337	8.875	5.014	0.043
20	19.202	19.703	22.255	24.220	6.749	7.349	7.849	8.349	4.571	0.040
30	14.060	15.816	18.071	23.135	5.844	6.639	7.301	7.964	9.155	0.053
40	11.523	13.897	16.628	19.429	5.597	6.422	7.109	7.797	9.968	0.055

Thermodynamic studies

The thermodynamic functions were studied in the range (10-50°C) and at initial concentrations (5-40 mg/L). The results of the thermodynamic values of (free energy ΔG, enthalpy ΔH and entropy ΔS) for the adsorption process of cadmium (II) ions on the surface under study are recorded in Table (3) which were calculated through the following equations(Ge and Ma, 2015).

$$\Delta G = -RT \ln k \qquad (4)$$

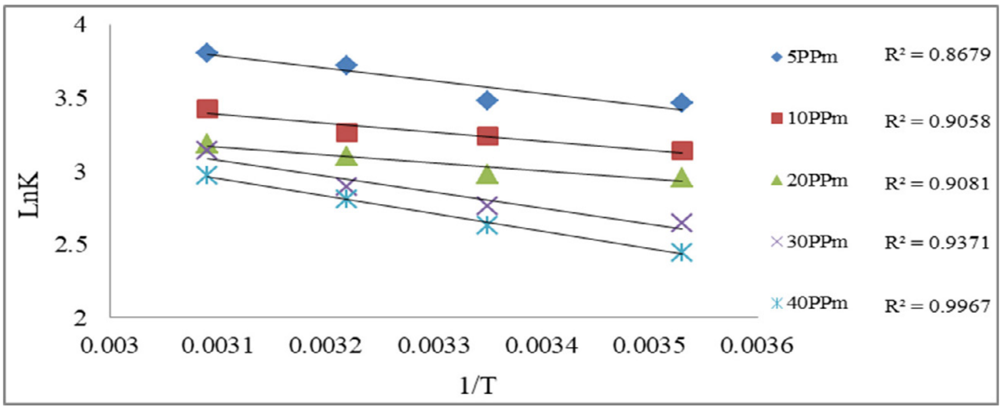

Fig. 9. plot of van·t Hoff relation ship between LnK versus 1/T

Table 4. Percentage removal of ions

Minute size (μm)	optimum conditions contact time (min)	temperature (C°)	pH	C_o (mg/l)	C_e (mg/l)	Removal%
75	45	25	7.35	0.093	0.002	97.84

$$K = Csolid / Cliquid \tag{5}$$

$$lnK = \Delta S / R - \Delta H / RT \tag{6}$$

Where DG is the change in free energy ($KJ.mol^{-1}$), K is the thermodynamic equilibrium constant for the adsorption process, C_{solid} is the Solid phase concentration (mg/L), C_{liquid} is the liquid phase concentration (mg/L), R is the general gases constant and T is absolute temperature in Kelvin (0.0083 $KJ.mol^{-1}$ K^{-1}).

The values of ΔH (KJ.mol-1) and ΔS (KJ.mol-1.k-1) were calculated using equation (6) So we get a straight line relationship whose slope represents (-ΔH / R) and the cutoff represents (ΔS/R).

Figure (9) shows the linear relationships of lnK values versus 1/(T) The correlation coefficient was (R2 = 0.8679 – 0.9967) for adsorption of hydrocarbons on the surface of porcelain.

The results show in Table (3) that the (DH) values of the cadmium ion (II) on the surface are positive, which means that the adsorption the cadmium ion (II) process is endothermic, and that the (DG) values are all negative, and this means that the adsorption is automatic (Tighadouini *et al*., 2020).

Positive values of (DS) on bentonite powder indicate that the adsorbed ions are less homogeneous on the surface than their form in solution, because the process is accompanied by diffusion (Kishani *et al*., 2021).

Application of the Method

The practical application included a process of removing cadmium ions from sewage samples from the collection center at the Hamdan main sewage treatment plant of the Directorate of sewage directorate of Basra Governorate - Iraq. The samples were collected in 2 litter plastic containers. The absorbance values of these samples were measured after digestion, using the Flame Atomic Absorption Spectrometer (F.A.A.S) to find out the concentration of cadmium ions present in these samples and then treat them with the adsorbent surface (bentonite clay) under the optimal conditions of acidity and the weight of the adsorbent material as well as the appropriate contact time.

Table (4) shows the results of practical applications using the adsorbent surface, and the results show the high susceptibility of the surface used to remove cadmium ions. The removal rates were (97.84%), which indicates the possibility of using this surface to remove cadmium ions from solutions contaminated with these elements.

CONCLUSION

The current study showed that bentonite powder has a good efficiency for adsorption of Cd (II) ions from its aqueous solutions. The surface showed improved Cd (II) ions removal abilities compared to some materials mentioned in the literature. The equilibrium data followed the Freundlich isothermal model. The results showed the highest removal rate of 95.19% for Cd(II) at pH 6.5 and 25°C. The results of the final study showed the possibility of using bentonite powder as a high-efficiency and low-cost sorbent to remove cadmium (II) from wastewater.

GRANT SUPPORT DETAILS

The present research did not receive any financial support.

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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