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Treatment of Radioactive Liquid Waste Using Bentonite

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Abstract

Radioactive liquid waste contaminated with cesium-137 found in the radiochemistry laboratories at Tuwaitha site, south of Baghdad, was treated in this work. Bentonite was used as a sorbent material for the removal of radioactive cesium-137 from liquid waste by ion exchange method. The results indicated that the best removal efficiency obtained was 95.13% with experimental conditions of 2 h mixture time, 0.04 g sorbent mass, and pH=10 for the radioactive liquid. It was found that the experimental results match well with Langmuir and Freundlich models, with better matching with the latter.

Keywords: Cesium-137, Bentonite, Isotherm.

معالجة النفايات المشعة السائلة باستخدام البنتونايت العراقي

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الخلاصة

تم في هذا البحث معالجة النفايات المشعة السائلة الملوثة بالسيزيوم−137 الموجودة في مختبرات الكيمياء الاشعاعية في موقع التويثة الذي يقع جنوب بغداد, حيث تم استخدام البنتونايت كمادة مازة لازالة السيزيوم − 137 المشع من النفايات السائلة بطريقة التبادل الايوني. بينت النتائج ان افضل كفاءة ازالة تم الحصول عليها هي 95,13% مع الظروف التجريبية (زمن خلط ساعتين, كتلة المادة المازة (0,04) غرام والأس الهيدروجيني pH=10 للسائل المشع). لقد وجد ان النتائج العملية تتطابق بشكل جيد مع نموذجي لانكماير و فريندلش وانها تتطابق مع نموذج فريندلش بشكل افضل من نموذج لانكماير.

1- Introduction

Rising urbanization, growth, industrialization, and other human activities have been causing increased environmental pollution in the world [1]. These contaminants pose serious threats to human environment. As one of the most important fission radionuclides in reactors, a great attention has been always directed to cesium-137 (half - life 30 years) due to its gamma radiation. It is hazardous for human health and environment [2]. There are increased endeavors that aim at disconnecting the dissolvable radioactive contaminants from watery waste through fixing them onto solid materials that can be discarded in a repository [3, 4]. Radiations can create destructive impacts on living life forms. Because of ignorance and unplanned conditions, various instances of damage, ranging from minor

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early skin injuries to delayed bone cancer and leukemia were reported among radiologists and others who were exposed to excessive amounts of radiations. When the first definitive "maximum permissible" levels of exposure to radiation were instituted, their general acceptance have led to a marked decrease in the incidence of radiation injuries [5]. Handling radioactive fluid waste includes the utilization of sundry methods such as filtration, precipitation, sorption, ion exchange, evaporation and/or membrane separation to meet the requirements, both for the release of cleaned effluents into nature and the conditioning of waste concentrates for disposal. New and improved substances and processes are under study and development in different countries [6]. Among these processes, inorganic ion exchange is a more adequate and suitable candidate for the mild or low radioactive wastewater due to its specific selectivity and radiation and thermic stabilities [7]. Smectic clays, always known as bentonites and utilized in a variety of industrial applications, are the most widely used and most interesting group of clay minerals [8]. Bentonite is described as an octahedral layer of aluminum atoms fixed through two tetrahedral layers of silicon atoms. It has a net negative electric charge because of the isomorphic exchange of Al^{3+} with Fe^{2+} and Mg^{2+} in the octahedral locations and Si^{4+} with Al^{3+} in the tetrahedral locations, which is balanced via the cations such as Na^{+} and Ca^{2+} existing between the layers and surrounding the edges. Natural bentonite becomes alkaline, with pH of 8 - 10, when hydrated with water. It is hydrophilic in nature as it is firmly hydrated by water. This clarifies why bentonite has a high water absorption capacity. Bentonite additionally has an extraordinary cation exchange capacity, bonding capacity, and plasticity, along with a powerful tendency to intereact with organic compounds [9]. Sulaymon *et al*., studied the treatment of cesium-contaminated radioactive liquid waste. The adsorbent materials used for extracting ^{137}Cs were bentonite and modified bentonite, with removal efficiencies of 85% and 91.8 %, respectively [2].

The purpose of the present work is to purify the radioactive liquid waste contaminated with cesium-137 using bentonite obtained from Iraqi sources. The adsorption of Cs^{+}, in addition to the effects of some factors such as shaking time, sorbent mass, pH, and the activity concentration, were studied in the present work.

2- Materials and methods

2.1- Sorbent: The adsorption of radioactive cesium onto Iraqi bentonite was studied. Scanning Electron Microscope (SEM, INSPECT S50) was used for surface characterization of bentonite. It can be described as a microscope that produces magnified real images of solid specimens using a high energy electron beam. It is attached with an EDX unit, which is used to obtain more precise details by concentrating on small parts of the sample (bentonite in this study) to determine their composition. The SEM micrograph of bentonite is shown in Figure-1 and its components are listed in Table-1. The results show that the main elements of bentonite are O, Si, Ca, C and Al), with O having the highest content (58.80 % of weight).

Table 1-Betonite components.

Element	Weight %
O	58.80
Si	14.08
Ca	7.43
C	7.14
Al	4.56
S	2.48
Fe	2.32
Mg	1.72
Na	1.10
P	0.20
Cl	0.17

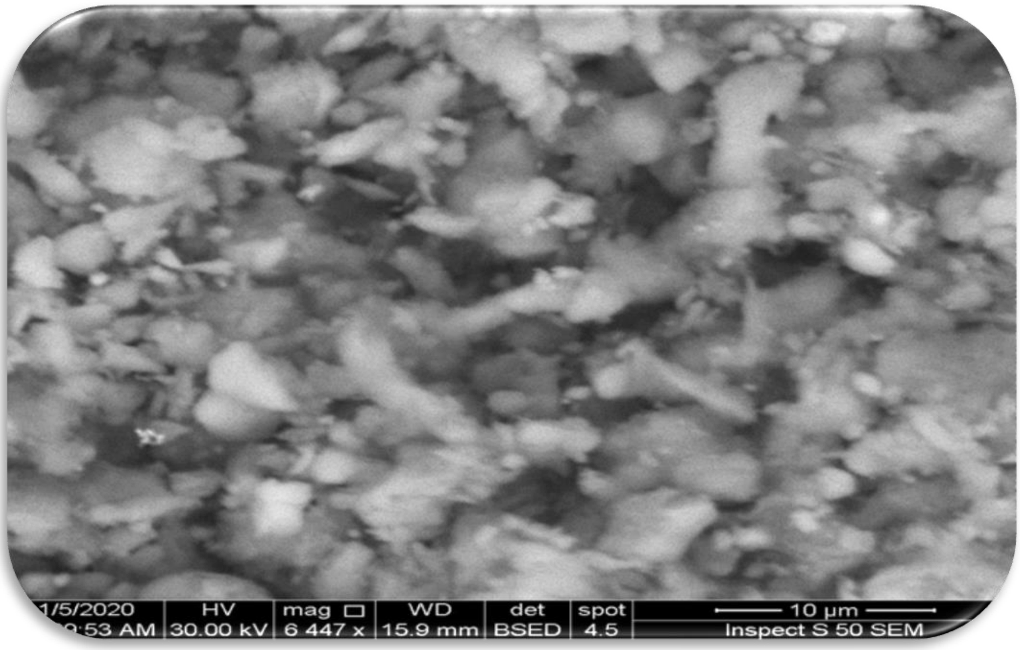

Figure 1-SEM for bentonite (scale 10 μm).

2.2- Sorbate: Contaminated liquid was collected from the radiochemistry laboratories in Al-Tuwaytha site. The sorbate liquid used in this work is Cs-137.

2.3- Equipment

The qualitative analysis of radionuclides was performed via gamma spectrometry analysis using high-purity germanium detector (HPGe) with 65% relative efficiency, 1.95 keV energy resolution, and 1.33 MeV of ^{60}Co isotope. Roller mixer was used to mix the sorbent and sorbate materials. Sensitive balance (Denver instrument - made in Germany) was utilized to measure weight of samples.

2.4- Adsorption isotherm: The batch process was applied to find the adsorption of cesium-137 ions upon bentonite. Five samples with different activity concentrations were used to study sorption isotherm. The isotherm models used in the present work were the Langmuir and Freundlich models.
The amount of substance adsorbed at equilibrium is [10]:

$$q_e = (C_\circ - C_e)V/W \quad (1)$$

where $C_\circ$ is the initial concentration, C_e is the concentration at equilibrium, V is the volume of fluid, and W is the weight of the adsorbent.
The removal efficiency is calculated as follows [10, 11]:

$$\text{Removal efficiency \%} = \left(\frac{C_\circ - C_e}{C_\circ}\right) \times 100\% \quad (2)$$

The Langmuir isotherm equation is represented as follows [12]:

$$\frac{C_e}{q_e} = \frac{1}{q_m\,b} + \frac{1}{q_m} C_e \quad (3)$$

where q_m and b are Langmuir constants and they can be calculated by plotting C_e / q_e against C_e.
The Freundlich equation is represented as follows [13]:

$$\text{Log}\,(q_{e)} = \frac{1}{n}\,\text{Log}\,(C_e) + \text{Log}\,(K_f) \quad (4)$$

where (K_f and 1/ n are Freundlich constants and can be found via plotting Log (q_e) against Log (C_e).

3- Results and discussion

The plot of activity versus mixing time, with a weight of 0.01 g and pH=6.8, is shown in Figure-2, where the activity decreases with increasing mixing time. In addition, the greatest removal of cesium ions was recorded during the first half hour, while the equilibrium time was 2 hours.

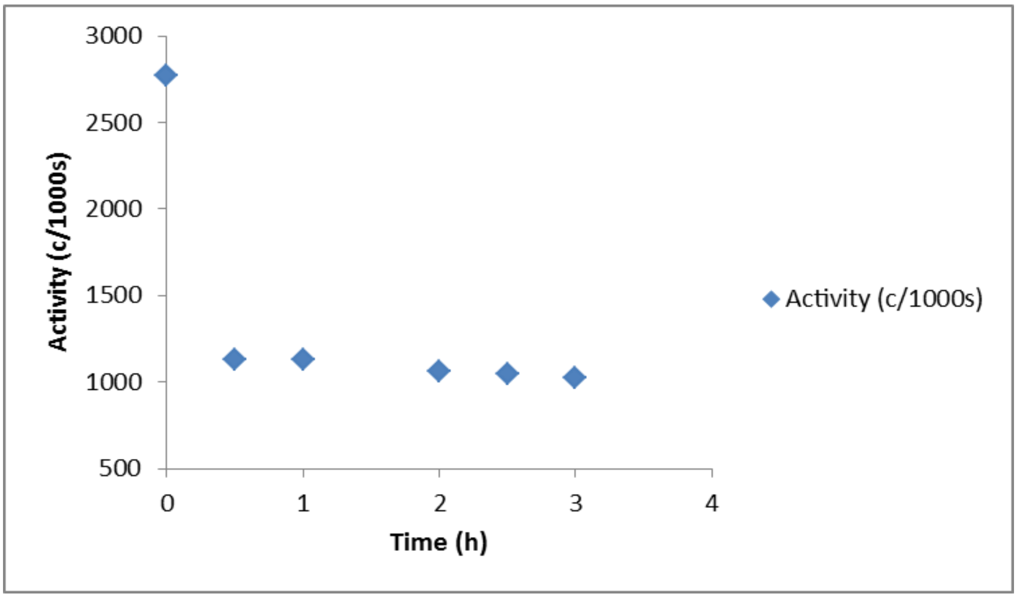

Figure 2-Effects of mixing time on the activity.

The plot of activity versus weight, with a constant time of 2 h and pH=6.8, is shown in Figure-3. The activity was decreasing with the increase in the weight of sorbent material, while the greatest removal efficiency was found at a weight of 0.04 g.

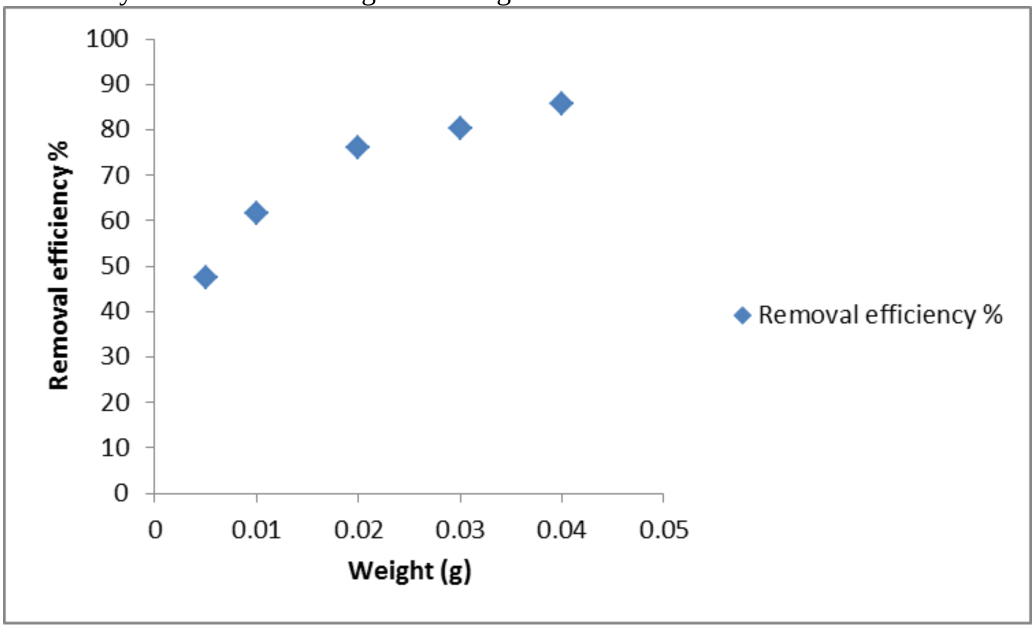

Figure. 3-Effects of weight on the activity.

Figure-4 shows the increasing removal efficiency with increasing pH. At low pH, surplus H^+ which is present in the fluid competes with the studied element ions for active locations, leading to less removal of element ions and negatively-charged surface hydroxyl groups. Therefore, the removal efficiency will decrease.

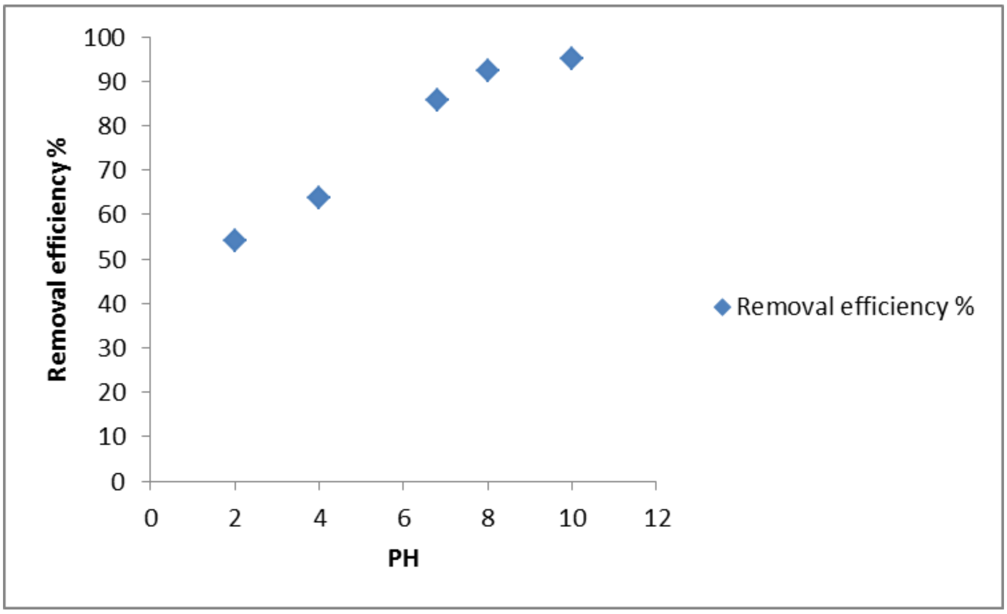

Figure 4-Effects of pH on removal efficiency.

The plot of removal efficiency (%) versus initial concentration, with time of 2 h, pH=6.8, and weight of 0.04 g, is shown in Figure-5. The removal efficiency was increased with increasing the initial concentration.

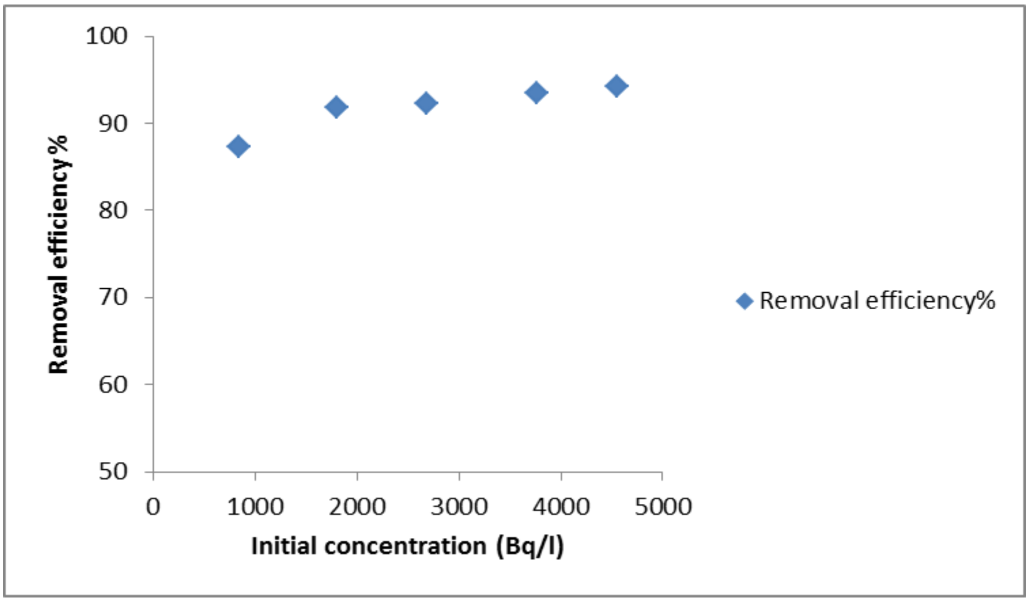

Figure 5-Effects of initial concentration on removal efficiency.

3.1- Equilibrium isotherms

Figure-6 shows the linear relationship of the isotherm Langmuir model, which includes the relationship between the equilibrium concentration and the amount of adsorbed material at equilibrium (Ce /qe against Ce).

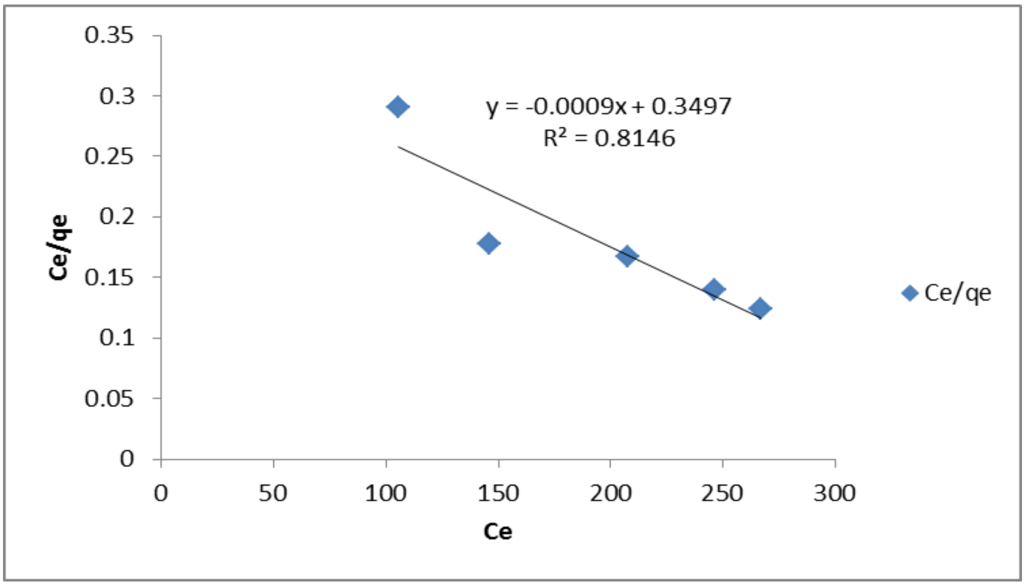

Figure 6-Langmuir plot of C_e/q_e against C_e for the adsorption of cesium-137 on bentonite.

Figure-7 shows the linear relationship of the isotherm Freundlich model, which includes the relationship between log qe and log Ce.

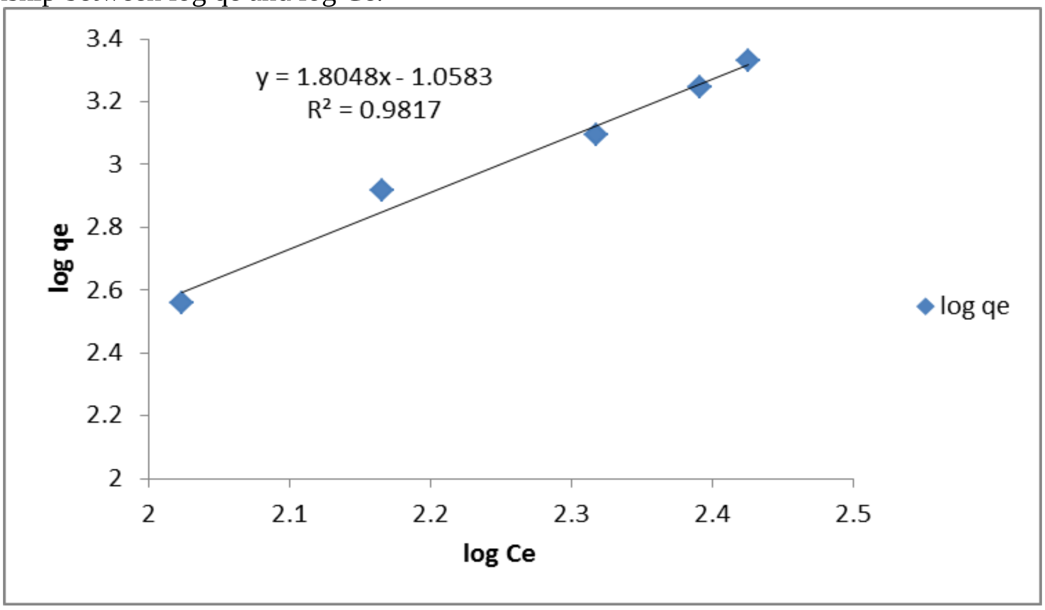

Figure 7-Freundlich plot of log q_e vs. log C_e for the adsorption of cesium-137 on bentonite.

The constants of Langmuir and Freundlich models with their correlation coefficients are listed in Table-2.

Table 2-Equilibrium isotherm model constants and correlation coefficients.

Isotherm model	**Isotherm model constant**				**Linear correlation coefficient (R^2)**
	q_m	**b**	**n**	**K_f**	
Langmuir	-1111.11	-0.0025736			0.8146
Freundlich			0.554078	0.347045	0.9817

The values of linear correlation coefficients (R^2) were 0.8146 and 0.9817 for Langmuir and Freundlich isotherm models, respectively. Freundlich model had a higher correlation coefficient than that of the Langmuir model. This implies that Freundlich model better fits the experimental results than Langmuir model.

4- Conclusions

According to the results, the present study could reach a number of conclusions:

- The equilibrium time was 2 h.
- The best weight of sorbent material to remove radioactive cesium was 0.04 g.
- The removal efficiency was directly proportional to the pH value, since the greatest removal efficiency was at pH=10, while the smallest removal efficiency was at pH=2.
- The removal efficiency was directly proportional to the initial concentrations.
- Linear correlation coefficient (R^2) values for Freundlich and Langmuir models were 0.9817 and 0.8146, respectively. Both models presented good experimental results, but Freundlich model was best suited to these results.

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