

Use of Zeolite to Reduce the Activity of Cesium -137 in the Liquid Waste**Aamir Abdullah Mohammed¹, Hayder Saleem Hussain¹, Salam Kudhair Abdullah²**¹Physics Department, College of Science, University of Baghdad, Baghdad, Iraq²Ministry of Science and Technology, Baghdad, Iraq

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Abstract

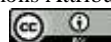
Prepared zeolite type A was used for the removal of cesium ions from aqueous solution. The experimental data were analyzed by Langmuir, Freundlich isotherms. Various parameters, such as contact time, zeolite weight, pH, and initial concentration, were studied. The results indicated that the highest removal efficiency was 95.53% at (2h time, 0.04 g weight, and pH=6.8). The results also showed that the Freundlich model fits well with the experimental results and is better than the Langmuir model.

Key words*Isotherm, zeolite, cesium.***Article info.***Received: Sep. 2020**Accepted: Jan. 2021**Published: Jun. 2021***استخدام الزيولايت لتقليل النشاط الإشعاعي للـسيزيوم-137 في النفايات السائلة**عامر عبدالله محمد¹، حيدر سليم حسين¹، سلام خضير عبدالله²¹قسم الفيزياء، كلية العلوم، جامعة بغداد، بغداد، العراق²وزارة العلوم والتكنولوجيا، بغداد، العراق**الخلاصة**

تم استخدام الزيولايت المحضر نوع A لازالة الايونات من المحلول المائي. تم تحليل البيانات العملية باستخدام نموذجي لانكبير وفريندلش بالاضافة الى دراسة تأثير العوامل المختلفة على عملية الازالة مثل زمن الاتصال و وزن الزيولايت و الاس الهيدروجيني والتركيز الابتدائي. بينت النتائج ان اكبر كفاءة ازالة كانت 95.53% عند زمن اتصال ساعتين ووزن 0.04 غرام للمادة المازة و 6.8 للأس الهيدروجيني للمحلول. كذلك بينت النتائج ان نموذج فريندلش يتلائم بشكل جيد مع النتائج العملية وبشكل افضل من نموذج لانكبير.

Introduction

The development of nuclear science and technology, nuclear weapons testing, electricity generation in nuclear power plants and the use of radionuclides in research and medicine have resulted in increased output of both radionuclide-containing liquid and solid waste and the release of artificial radionuclides into the environment. Radiocesium ¹³⁴Cs and ¹³⁷Cs are one of the most common products radioisotopes of nuclear fission products that have a relatively long half-life and are considered a dangerous element due to their high solubility, making it easy to transfer to the ambience [1,2].



Sorption was an efficient separation method for a wide variety of uses. Using activated carbon as sorbent is costly, an alternative cheap sorbent capable of dramatically reducing the cost of a sorption system has always been looked for [3]. Active separation techniques, e.g. solvent extraction, ion exchange using inorganic and organic ion exchangers for extracting radionuclides from liquid radioactive waste have been developed. Water treatment processes most commonly used to eliminate radionuclides (Cs, Sr, U, Pu) are those based on co-precipitation and sorption [4]. Many researchers reported using zeolite to extract Cs⁺ ions [5, 6]. Ion exchange properties of zeolites have received great attention, especially for application in radioactive liquid waste treatment [7]. Rashid (2009) [8] studied the ability of clay minerals to absorb radioactive cesium from polluted liquid samples taken from Al-Tuwaitha site. Radio cesium sorption has been investigated for employing various Iraqi raw materials (bentonite, porcellante, kaolin) as well as zeolite.

Experimental work

Prepared Zeolite -A was used in the present work to remove cesium ions from the liquid waste. It was hydrothermally synthesized using sodium aluminum as the source of the alumina. Sodium metasilicate was used as source of silica, and sodium hydroxide as a source of alkaline and cation. Zeolite-A was synthesized by adding 0.94 g of NaOH to 30 g of distilled water as the source of Na₂O. The solution was then divided into two halves.

The first half was mixed with 2.43 g of sodium aluminate used as an aluminum source, while the second half (15 g of NaOH solution) was used to dissolve 3.224 g of sodium metasilicate. The silicate solution was poured into the aluminium-containing solution. The mixture was added to fill two thirds of an autoclave volume, sealed tightly and left in an oven at 100 ° C for 3 hours. The solid was isolated from the solution by filtration. A pH meter was used to test the solution's pH as it was washed with water; washing is stopped when pH was less than 9 [8].

The solid was dried with a filter in the oven. The samples were analyzed with X-ray diffractometer to test zeolite's crystallinity. Additionally, Scanning Electron Microscope (SEM) with EDX Unit was used to determine the morphology and components of the zeolite structure (Figs. 1, 2 and Table 1).

In gamma spectrometry analysis, a qualitative study of radionuclides was conducted with a high-purity germanium detector (HPGe with (65 per cent) relative efficiency.

The isotherm models used in the present work were model Langmuir and model Freundlich [9].

The isothermic equation Langmuir is defined by [3]:

$$C_e/q_e = 1/(q_m b) + C_e/q_m \quad (1)$$

where q_e and C_e are amount of substance adsorbed and the concentration at equilibrium respectively. q_m and b are Langmuir constants.

The Freundlich equation is represented as follows [10]:

$$\text{Log } q_e = (1/n) \text{ log } C_e + \text{ log } K_f \quad (2)$$

where; (K_f , $1/n$) are Freundlich constants.

The removal efficiency was calculated as follows [10, 11]:

$$\text{Removal eff.} = (C_0 - C_e / C_0) * 100\% \quad (3)$$

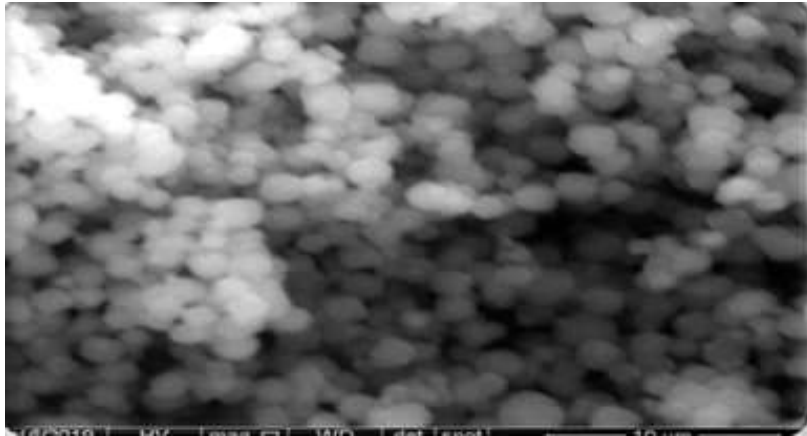


Fig.1: SEM of zeolite.

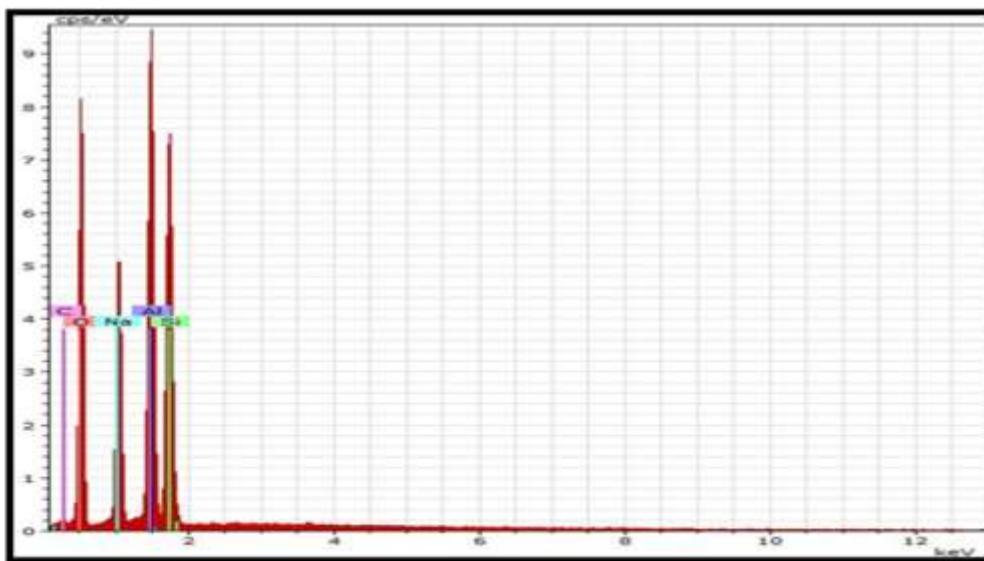


Fig.2: EDX spectrum of zeolite.

Table 1: Elements analysis of zeolite.

Element	Weight %
O	47.12
Si	18.43
Al	17.16
Na	11.74
C	5.55

Results and discussion

Fig.3 explains the effect of time on the activity of cesium. The adsorption occurs dramatically within the first half hour due to the fact that adsorption occurs at the beginning and the ion exchange occurs between the ions in the fluid and the adsorbent material, where the ions transported from the solution to occupy the majority of the cavities on the surface of the adsorbent, hence the decrease in the radioactivity which was minimal and almost stabilized at two hours or so-called equilibrium time.

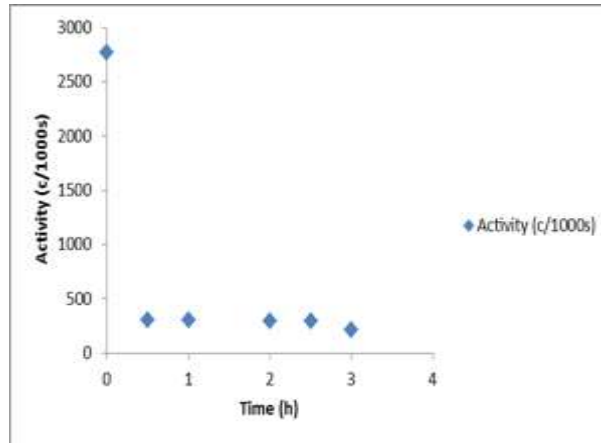


Fig.3: Effect of contact time on the removing of cesium from the solution.

Fig.4 shows the effect of weight increase on removal efficiency, where the greatest removal efficiency was at a weight of 0.04 g. The values of weight (0.005, 0.01, 0.02, 0.03, and 0.04) g were chosen in the present work.

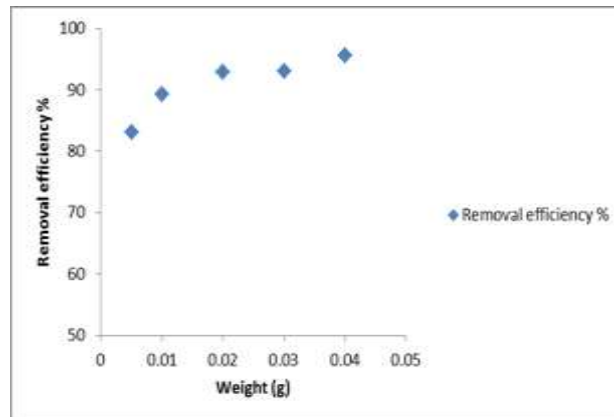


Fig.4: Effect of weight on the removal efficiency.

To investigate the effect of pH on the radioactive cesium removal efficiency, five values of pH were chosen (2, 4, 6.8, 8, and 10), where the greatest removal efficiency was at pH =6.8 as shown in Fig.5.

At low pH, the surplus H^+ present in solution with the studied element ions for active locations contributing to less element ion removal and negative surface hydroxyl groups starts to occur.

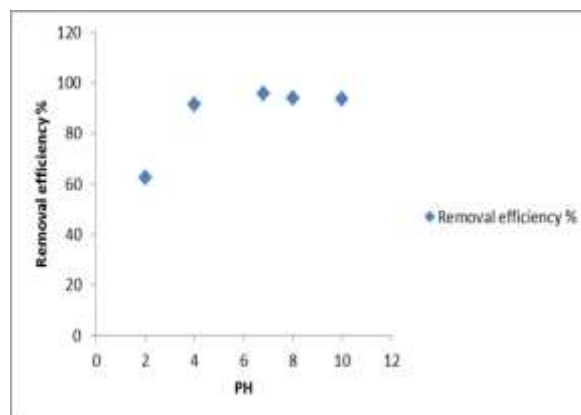


Fig.5: Effect of pH on the removal efficiency.

Fig.6 explains that the removal efficiency increases with the increase in the initial concentration and then gradually stabilizes with the increase in concentration.

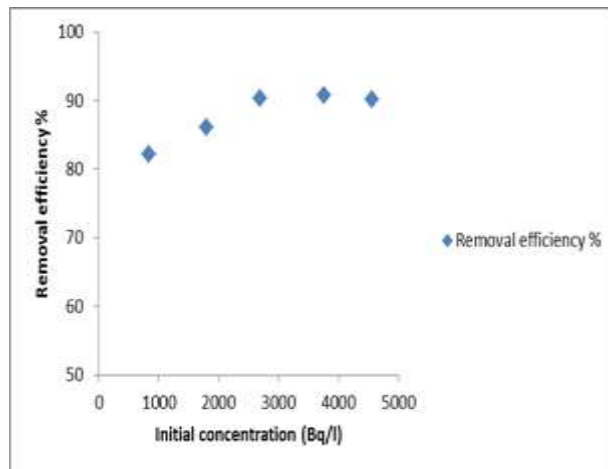


Fig.6: Effect of initial concentration on the removal efficiency of cesium.

To study equilibrium isotherm, Langmuir and Freundlich adsorption models used as shown in Figs.7 and 8.

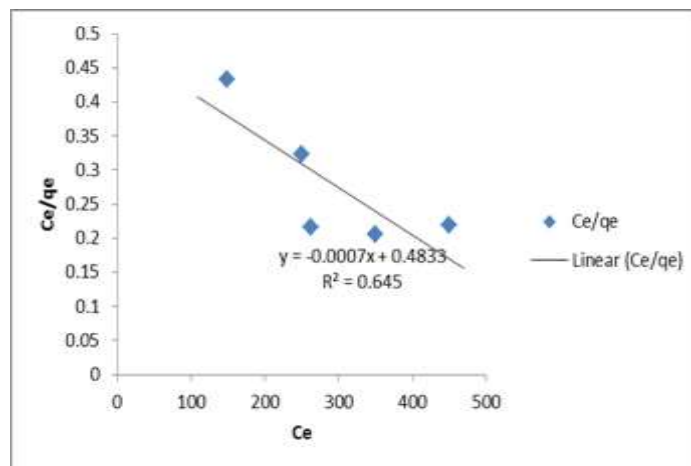


Fig.7: Langmuir isotherm model.

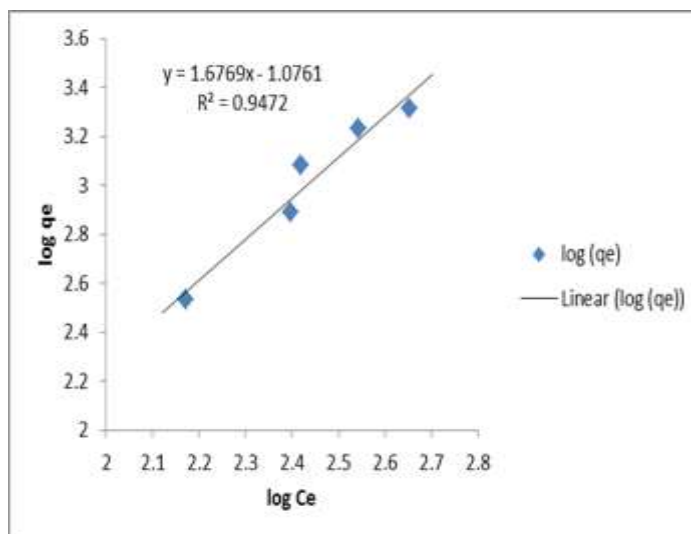


Fig.8: Freundlich isotherm model.

The values of linear correlation coefficients (R^2) were (0.5175 and 0.853) for Langmuir and Freundlich isotherm models respectively. The Langmuir constants were ($q_m = 1428.57$ and $b = -0.001448$), while Freundlich constant were ($n = 0.596338$ and $K_f = 0.340923$).

Conclusions

The results showed that the prepared zeolite type (A) was suitable for the process of adsorption or removal of cesium ions from radioactive liquid waste. In this study the volume of radioactive waste was reduced about (500) times with removal efficiency 95.53%. Therefore, this method can be considered as a very effective way to treat radioactive liquid waste.

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