

## *Evaluation of Zeolite Efficiency for Removal of Cesium Ions from Seawater*

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### **Abstract**

Cesium is a dangerous radioactive material which is a problem to marine life when it leaks into seawater. Zeolites are substances for removing cesium because they absorb cationic elements and are widely used due to their simplicity, high efficiency and low cost. The objective of this study was to investigate the efficiency of NaCl treated zeolite for removal of cesium ion from synthetic seawater. This study was carried out by performing batch experiments. A synthetic seawater solution of 100 mL containing 80 mg L<sup>-1</sup> of non-radioactive cesium was mixed with NaCl treated zeolite of two sizes, 300-424  $\mu\text{m}$ . and 425-850  $\mu\text{m}$ . The sorbent weight per solution volume ratios used in the experiment were 5, 10, 15, and 20 g. per 100 mL of seawater. The contact times were 10, 20, 30, 60 and 120 minutes. The effect of zeolite size was an important factor in cesium removal. Zeolite with a size of 300-424  $\mu\text{m}$ . showed greater efficiency than that with a size of 425-850  $\mu\text{m}$ . The optimum sorbent weight per solution volume ratio was 15 g. per 100 mL of seawater. The best contact time was 60 minutes with more than 80% uptake occurring through rapid adsorption. These results show that NaCl treated zeolite is a promising material for removing cesium from contaminated seawater.

Keywords: Adsorption, Zeolite, Cesium removal

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## **1. Introduction**

Releasing radiation into the environment is a serious problem because radiation can produce harmful effects on living organisms. The release of large amounts of radioactive elements into aquatic environment can be accumulated by biota and are cycled through the food web to eventually threaten human health, ranging from minor early skin lesions to cancer (Shaw & Bell, 1994).

Cesium is the most plentiful radionuclide in nuclear fission products that is accidentally released. It has a long half-life of about 30 years and is considered a hazardous element to the environment. Currently, among many techniques used for the treatment of radioactive waste, the most common ones are chemical precipitation, ion-exchange, adsorption and reverse osmosis. The ion exchange process is considered one of the more effective methods for the removal of radioisotopes from liquid waste. Ion exchange is a chemical treatment process used to remove unwanted ionic species from wastewater. It is basically a simple process based on reversible interchange of ions between liquid and solid with no permanent changes in the structure of the solid.

Zeolite is the focus in this study because it has high cation-exchange capacities and selectivity for many radioactive materials (El-Kamash, 2008) & (Yasmen & Maysoun, 2011). This experiment aims to study the appropriate conditions for removal of cesium and strontium contamination in artificial seawater using a batch experiment.

## **2. Materials and methods**

### **2.1 Adsorbents**

Selected zeolite with particle size of 300-424  $\mu\text{m}$  and 425-850  $\mu\text{m}$  was obtained by crushing zeolite and using sediment sieves with different mesh sizes ranging between 300-424  $\mu\text{m}$  and 425-850  $\mu\text{m}$ .

### **2.2 Preparation of treated NaCl zeolite particles as adsorbents**

First, reflux 3 g. of zeolite with 100 mL of 1.0 M NaCl at room temperature for 24 hours. After 24 hours, rinse the mixture and wash zeolite with 300 mL of deionized water. Then, activate the zeolite by mechanical shaking with 25 mL of methanol for 60 minutes. Filter and dry the zeolite at 50 °C for 3 h. Then, the activated zeolite samples were ready to be used in the experiment.

### **2.3 Artificial seawater**

The artificial seawater was prepared by dissolving commercial sea salt in deionized water. An appropriate amount of commercial sea salt was added into deionized water to obtain a salinity of 35 ppt. A digital salinity probe was used to measure salinities in the artificial seawater. In addition, a pH value was recorded in order to be used as additional information of the experiment.

## 2.4 Chemical preparation

A solution of 80 mg/l of cesium chloride was prepared by dissolving 0.1014 gram of cesium chloride (MW 168.35) in 1,000 ml of artificial seawater with salinity of 35 ppt.

## 2.5 Batch sorption experiment

The experiments were performed in a batch reactor at room temperature with continuous stirring at 160 rpm.

## 2.6 Sorption isotherm

NaCl treated zeolite of 5 g., 10 g., 15 g., and 20 g. of were left in contact with 100 ml of cesium chloride solution (80 mg/l) at pH values of 8.4. The samples were filtered with No.5 filter paper to avoid any precipitation before the ICP-MS measurement.

The analysis of the isotherm data is important to develop an equation which accurately represents the results and which could be used for design purposes. In order to investigate the sorption isotherm, two equilibrium models were analyzed: Langmuir and Freundlich isotherm equations. These two isotherm models were derived and used for gas adsorption by microporous adsorbents, and then extended to solute adsorption from aqueous solutions. The Langmuir model is obtained under the ideal assumption of a totally homogeneous adsorption surface, where the Freundlich isotherm is suitable for a highly heterogeneous surface (Hui, Chao & Kot, 2005).

### 2.6.1 Langmuir isotherm

The Langmuir sorption isotherm is the best known of all isotherms describing sorption and it has been successfully applied to many sorption processes. It is represented as:

$$q_e = q_m \frac{bC_e}{1+bC_e} \quad (1)$$

In the formula,  $C_e$  is the equilibrium aqueous metal ions concentration (mg/l),  $q_e$  the amount of metal ions adsorbed per gram of adsorbent at equilibrium (mg/g),  $q_m$  and  $b$  is the Langmuir constants related to the maximum adsorption capacity and energy of adsorption, respectively. The values of  $q_m$  (mg/g) and  $b$  ( $\text{mg}^{-1}$ ) can be determined from the linear plot of  $C_e/q_e$  versus  $C_e$ .

### 2.6.2 Freundlich isotherm

The Freundlich isotherm is most frequently used to describe the adsorption of inorganic and organic components in solution. This fairly satisfactory empirical isotherm can be used for a non-ideal sorption that involves heterogeneous sorption and is expressed as:

$$\log q_e = \log K + \frac{1}{n} \log C_e \quad (2)$$

In the formula,  $K$  is briefly an indicator of the adsorption capacity and  $1/n$  the adsorption intensity. The magnitude of the exponent  $1/n$  gives an indication of the favorability of adsorption. Values of  $n$ , where  $n > 1$  represent a favorable adsorption condition. By plotting  $\log q_e$  versus  $\log C_e$ , values of  $K$  and  $n$  can be determined from the slope and intercept of the plot.

## 2.7 Removal efficiency

In order to obtain the removal efficiency of metal ions by the adsorbent, one must consider the percent removal as:

$$\text{Removal efficiency} = \frac{C_0 - C_e}{C_0} \times 100 \quad (3)$$

$C_0$  is the initial metal ion concentration (mg/l), and  $C_e$  the equilibrium metal ion concentration (mg/l)

## 3. Results and discussion

### 3.1 Characterization

The zeolite sample must be analyzed for chemical composition and crystal structures of the zeolite sample, since the ion exchange characteristics of any zeolite are dependent on the chemical composition and crystal structures of zeolite. Chemical compositions were analyzed by X-Ray Fluorescence Spectrometer (XRF) and crystal structures were identified from X-ray diffraction (XRD). The result of chemical composition of zeolite is shown in Table 1.

**Table 1**

*Chemical composition of the zeolite sample*

Element	Concentration (% wt)
SiO <sub>2</sub>	78.00
Al <sub>2</sub> O <sub>3</sub>	12.60
CaO	3.50
K <sub>2</sub> O	3.11
Fe <sub>2</sub> O <sub>3</sub>	1.32
MgO	0.73
Na <sub>2</sub> O	0.38
TiO <sub>2</sub>	0.19
BaO	-
SrO	-

### 3.2 Effect of particle size

Figure 1 shows relationship between size of zeolite and percentage removal of cesium. Two sizes of Zeolite were prepared, 300-424  $\mu\text{m}$  and 425-850  $\mu\text{m}$ , then shaken in 100 ml cesium contaminated seawater for 120 min. In general, the smaller size has a greater efficiency than the larger size because it has larger surface areas of its particles (Suzuki, Ozawa, Ochi, Chikuma & Watanabe, 2013). Findings showed that zeolite of the size 300-424  $\mu\text{m}$  could adsorb cesium ions from seawater (74.38%) better than at a size of 425-850  $\mu\text{m}$  (67.51%).

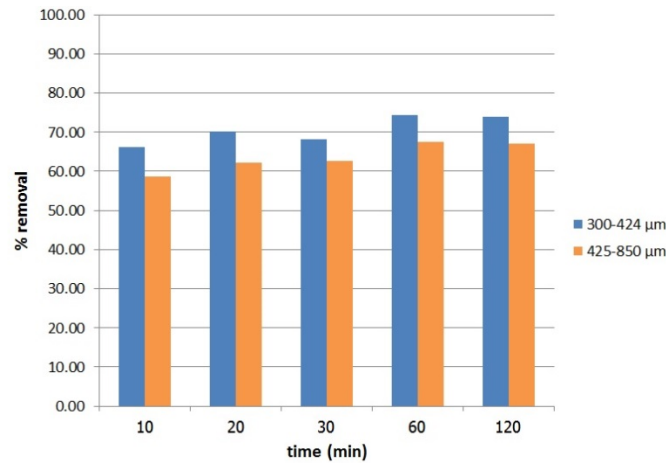


Figure 1: removal capacity comparison by size of zeolite

### 3.3 Effect of shaking time

The effect of shaking time was studied; 15 g. of zeolite was shaken in 100 ml seawater for different periods ranging from 10 minutes to 2 hours. The result in Figure 2 shows that zeolite of 300-424  $\mu\text{m}$  has a fast adsorption of cesium in seawater, more than 80%, within 20 minutes, followed by steady adsorption.

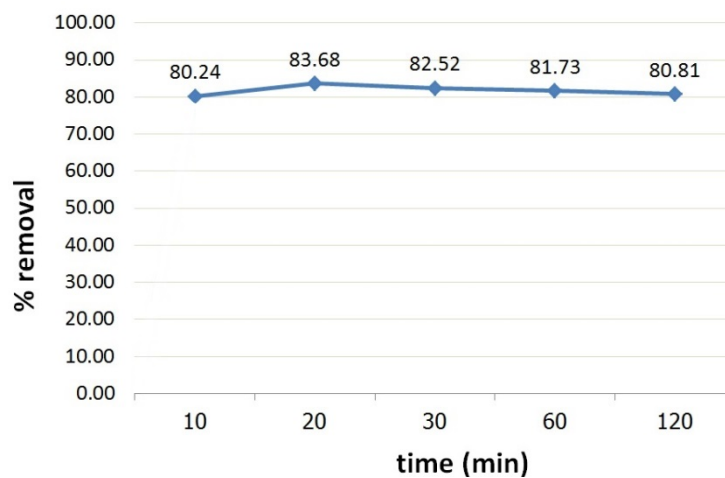


Figure 2: Time dependence of Cs uptake

### 3.4 Effect of adsorbent weight

The different amounts of zeolite used in this study were 5 g, 10 g, 15 g, and 20 g, shaken in 100 ml. seawater solution for 2 hours. These factors were expected to show the optimum adsorbent weights that could be used to achieve high removal efficiency. Figure 3 shows that the efficiency of using 300-424  $\mu\text{m}$  zeolite for 15 g. was similar to that using 20 g. zeolite ( $p\text{-value} > 0.05$ ) for the removal of cesium from seawater. For this reason, the optimum adsorbent in this study was 15 g/100 ml.

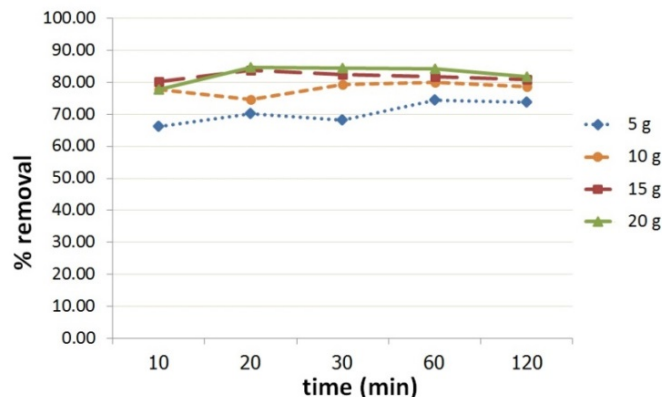


Figure 3: Relationship between adsorbent weight and percent cesium removal

### 3.5 Sorption isotherm study

Figure 4 and Figure 5 show the correlation coefficients ( $R^2$ ) of cesium adsorption isotherms by zeolite in Langmuir's equation and Freundlich's equation were 0.9555 and 0.9606 respectively. The adsorption isotherm data was fitted to both the Freundlich and Langmuir isotherm equations ( $R^2 > 0.9$ ). However, the linear Langmuir adsorption equation of cesium showed a negative value as indicated in Table 2. Thus, adsorbent better fit the Freundlich's equation as shown by the higher regression coefficient.

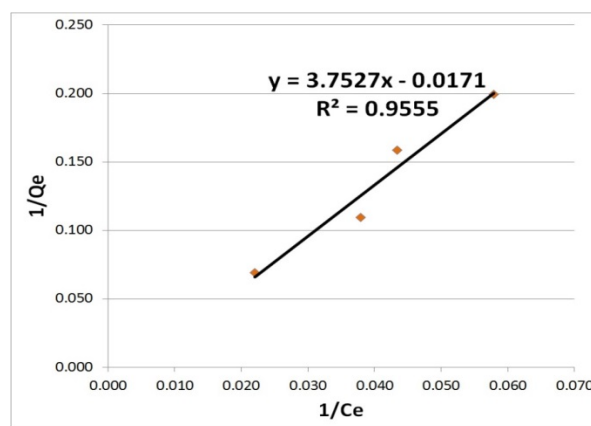


Figure 4: Linear Langmuir adsorption isotherm of Cesium

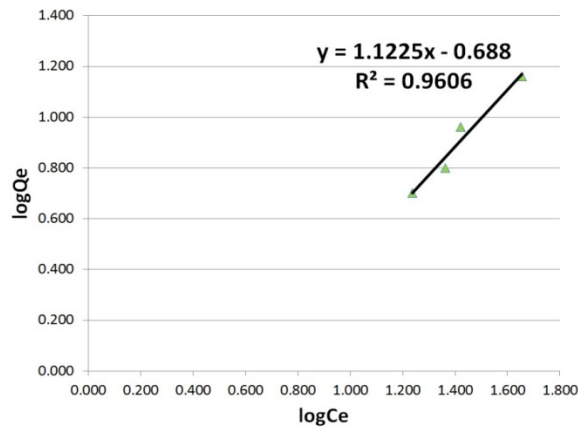


Figure 5: Linear Freundlich adsorption isotherm of Cesium

**Table 2**

*Isotherm parameters for adsorption of cesium onto zeolite*

Isotherm model	Isotherm parameters	Metal cesium
Langmuir isotherm	Qmax (mg/g)	0.2664
	b	-219.51
	R <sup>2</sup>	0.9555
Freundlich isotherm	K (mg/g)	0.205
	1/n	1.1225
	R <sup>2</sup>	0.9606

#### 4. Conclusions

The zeolite had increased removal capacity when the size was small. The optimum zeolite used was 15 g/100 ml and the maximum removal efficiency of cesium was 83.68%. Zeolite had rapid adsorption of cesium in seawater, more than 80% within 20 minutes. The adsorption isotherm data best fits the Freundlich model with a maximum adsorption capacity of 0.205 mg/g.

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