Treatment of low level radioactive waste containing ^{134}Cs radionuclide using modified natural clay

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1. Background and Goal of the present work

Radioactive waste-related environmental and health issues are receiving more and more attention globally. One of the nuclear fission products found in radioactive waste that poses the most significant problems is radioactive cesium. The main goal of this paper is to use modified bentonite made from 8-hydroxyquinoline to remove the radioactive ^{134}Cs from an aqueous solution.

2. General experimental, methods, and materials

2.1. Materials

Merck supplied the 8-hydroxyquinoline and cesium chloride salt. In order to gain ^{134}Cs, a precise weight of high purity cesium chloride (20 mg) that was wrapped in aluminium foil and exposed to radiation for 48 hours in the second Egyptian research reactor at Inshas facility was used. The irradiated sample was kept as a stock solution after dissolving in 5 mL of 1.0 M HCl. A high resolution NaI(Tl) γ-ray spectrometry model 802- 3X3 with pulse height multi-channel analyzer (McA), Canberra, USA was used to radiometrically measure the gamma activity of radioactive cesium.

2.2. Preparation of sorbent material

Before usage, natural bentonite was powdered, sieved, cleaned, and dried for two hours at 110°C. The suspension of 30 g of bentonite in 4.7 pH deionized water was combined with 8-hydroxyquinoline. For the immobilization of 8-hydroxyquinoline onto bentonite, the mixture was agitated for 24 hours. Filtration was employed to separate the solid phase, which was then rinsed three times with deionized water. Prior to usage, it was sieved, powdered, and dried in a 60°C oven for 24 hours.

2.3. Sorption experiments

The influence of pH, contact time, and metal ion concentrations on the removal of ^{134}Cs radioisotopes from the aqueous solutions were studied by the batch method to calculate the distribution coefficients and quantity sorbed of ^{134}Cs on HQ–Bent. The initial and equilibrium activities, A_i and A_e of ^{134}Cs were determined by high resolution NaI(Tl) γ-ray spectrometry.

3. Results and discussion

3.1. Characterization of the prepared sorbent

The elemental analysis of bentonite has been carried out to indicate that silica and alumina make up the majority of the material with 54.9 and 21.5%, respectively, along with minor amounts of metal oxides including iron, potassium, titanium, sulfur, magnesium, and calcium oxides as displayed in Fig. 1.

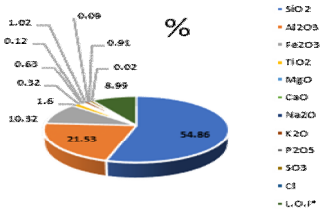

Fig. 1 *Elemental analysis of bentonite*

3.2. Effect of pH on the sorption process

The more important factor influencing the sorption process is pH, since hydrogen ions themselves fiercely compete with the positive ions, this is only relatively sorbed. The results are displayed in Fig. 2, and the data exposed that increasing pH increases the sorption percentage of radioactive cesium. Reduced competition between the proton and the ions is to blame for this. According the figure, the maximum sorption for ^{134}Cs was 88.24%. The optimum pH was selected at pH 7.8.

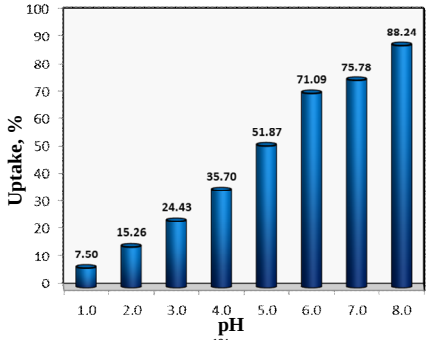

Fig. 2*. Effect of pH on removal of radioactive^{134}Cs by HQ–Bent at room temp. and 10-4 mol/L*

3.3. Effect of contact time at different concentrations

According to Fig. 3, the sorption rate is initially rapid for the first 25 to 30 min and then it gradually declines with longer contact times as a result of the sorbent's active sites becoming less active. The rate-limiting stage in the sorption process was studied using the experimental results. Additionally, knowledge of the kinetics of radioisotopes uptake is necessary for choosing the ideal operational settings for large-scale metal removal operations. Also, the clearance percentage increased when the concentration was raised.

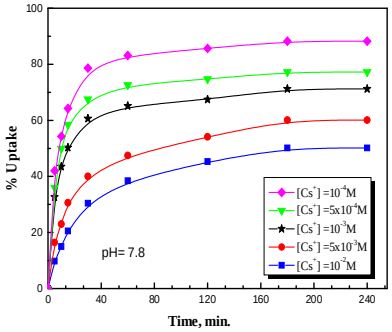

Fig. 3. Effect of contact time on sorption of ^{134}Cs at different concentrations, pH=7.8 and room temperature

3.4. Equilibrium isotherm

The isotherm studies are necessary to gain the sorption capacity of the sorbent material, scale up and build the sorption apparatus. The most often used sorption isotherms are the Langmuir and Freundlich equations.

From Langmuir isotherm, Eq. (1), the sorption capacity of radioactive ^{134}Cs onto 8HQ-Bent was computed and found to be 0.343 mmol/g as reported in Table 1. While the capacity of ^{134}Cs onto bentonite ore was 0.140 mmol/g, this means that the modification process enhanced the sorption capacity of bentonite. Fig. 4 displays the plot of Langmuir isotherm and Fig. 5 displays the plot of Freundlich isotherm.

$$\frac{1}{q_e} = \frac{1}{Q} + \frac{1}{bQ}\frac{1}{C_e} \qquad (1)$$

$$\log q_e = \frac{1}{n}\log C_e + \log k \qquad (2)$$

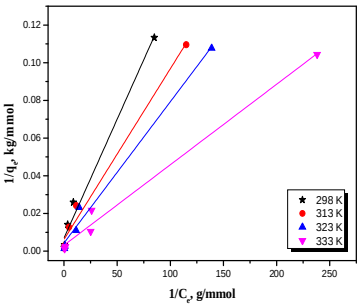

Fig. 4. Langmuir isotherm plot for sorption of ^{134}Cs using 8HQ-Bent

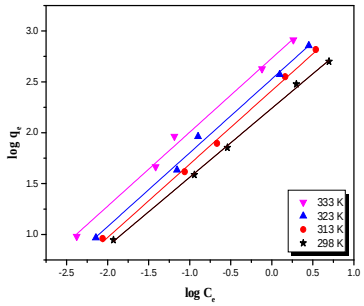

Fig. 5. Freundlich isotherm plot for sorption of ^{134}Cs using 8HQ-Bent

	Langmuir			Freundlich		
Temp., K	**Q, mmol/g**	**b, L/mmol**	R^2	R^2	n	k, (mmol/g)(mmol/L)$^{1/2}$
298	0.343	7.0	0.982	0.999	1.475	0.373
313	0.467	6.7	0.983	0.998	1.388	0.651
323	0.650	5.3	0.986	0.993	1.379	0.937
333	0.913	7.4	0.987	0.994	1.376	1.537

3.5. Comparison of sorption capacity of cesium onto different materials

Table 2 compares the ^{134}Cs ion capacity values onto 8HQ-Bent with various sorbents found in the literature. It is clear that the 8HQ-Bent has a significantly higher sorption capacity than many other sorbents. It demonstrates that 8HQ-Bent is a material that holds promise for removing radioactive cesium from liquid radioactive waste.

Sorbents	Experimental conditions	Capacity, mmol/g	References
Egyptian bentonite	pH=2	0.0002	[13]
bentonite/polyaniline	pH=7	0.18	[14]
Taiwan laterite	pH=7	0.30	[15]
montmorillonite-prussian blue	pH=7	0.42	[16]
Ammonium-pillared MMT/Fe_3O_4 composite	pH=6	0.2	[17]
Sericite	pH=5	0.05	[18]
8HQ-Bent	pH=7.8	0.343	Present work

4. Conclusion and Acknowledgement

Bentonite clay was successfully modified by 8-hydroxyquinoline and applied to remove ^{134}Cs from an aqueous solution. The Langmuir isotherm was used to calculate the sorption capacity, which was computed to be 0.343 mmol/g. According to the findings, 8HQ–Bent is a highly effective sorbent media and is suggested for removing radioactive 134Cs from radioactive waste. This paper is based upon work supported by Science, Technology & Innovation Funding Authority (STDF) under grant number 46016.