

Application of Solid Waste As An Adsorbent For Capture of ^{137}Cs , ^{85}Sr and ^{131}I From Environmental Water

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**Application of solid waste as an adsorbent for capture of ^{137}Cs , ^{85}Sr and ^{131}I
from environmental water**

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Abstract

Cesium, strontium, and iodine radionuclides are produced from uranium fission and easily soluble in water through accidents or leakages, as those occurred at Chernobyl, Three Mile Island, and Fukushima. Thus, developing an economical and effective process for removing these radioisotopes from real water is a progressively important issue. Hence, adsorption of ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides was achieved using solid waste adsorbent. The solid waste adsorbent is granular activated carbon (GAC) gained from the wasted household water filters (from the second stage). After the adsorption process, the gained data illustrated that the percentages of uptake for ^{137}Cs , ^{85}Sr , and ^{131}I were 87.6, 85.6, and 82.7 %, respectively. Application of GAC for the decontamination of real water as groundwater, river water, tap water, and seawater was achieved. The findings revealed that GAC has the ability to be employed effectively as a hopeful material for the decontamination of radioactive ^{137}Cs , ^{85}Sr , and ^{131}I from the environmental water.

Keywords: Solid waste adsorbent; ^{137}Cs , ^{85}Sr and ^{131}I radionuclides; Environmental Water

1. Introduction

Environmental and health issues produced by radioactive waste attracted attention worldwide since the wide application of nuclear science started from the early stages of the twentieth century [1]. The radioactive waste is produced from the research reactors, radiochemical labs., industrial activities, and nuclear medicine.

At the period of Chernobyl nuclear accident, the clouds released from the reactor polluted extensive zones as they were transported by the winds, not only in the surrounding area but also in areas towards the northern hemisphere. Water-soluble radionuclides as cesium, iodine, and strontium, contaminated food products, and aquatic ecosystems. Even with the relatively short half life of radioiodine (^{131}I), the occurrence of thyroid cancer in children existing in contaminated areas rose significantly. Consequent phases of environmental contamination were primarily due to ^{90}Sr and ^{137}Cs concentrated in human bone and muscles, respectively, due to their similarity to calcium and potassium, respectively, which led to their classification as cancerogenic.

After Fukushima accident in Japan, radioactive waste treatment has become a rising interest. At Fukushima Daiichi Nuclear Power Plant, 15PBq of ^{137}Cs , 18PBq of ^{134}Cs , 0.14PBq of ^{90}Sr , 2.0PBq of ^{85}Sr , and 160PBq of ^{131}I were released to the surroundings to date, and radioisotopes from the reactors were noticed in Greece and Russia [2]. The scavenging of radiocesium is very complicated owing to the lack of knowledge of the behavior of ultra-trace ions that compete with the high amount of monovalent cations.

Different kinds of sorbents are utilized for treating radioactive waste. Low-cost adsorbents as alum industrial waste [3], eggshell material [4], brick kiln waste [5], marble dust [6], hydroxyapatite [7], and perlite [8] offer a probable alternative to present materials for scavenging of ions. In this investigation, the adsorptive properties of one such low-cost

adsorbent (solid waste from consumed household water purifiers) is evaluated for ^{137}Cs , ^{85}Sr , and ^{131}I removal.

Various technologies are employed to remove the radioisotopes from radioactive wastewater, including adsorption [9–11], impregnation [12], ion exchange [13–16], biological methods [4, 17], and membrane [18]. Adsorption is an economical and highly effective method supplied the accurate adsorbents are employed in an environment that favors adsorption.

In this work, an economical method was employed to ^{137}Cs , ^{85}Sr , and ^{131}I removal from environmental water (groundwater, river water, and tap water) using solid waste gained from the wasted household water filters.

2. Experimental

2.1. Chemicals and reagents

Cesium chloride, strontiumchloride and potassium iodide salts were purchased from Merck (Merck D-6100 Darmstadt, Germany). The radioactive tracer ^{131}I and ^{85}Sr were gained from the second Egyptian Nuclear Reactor, and ^{137}Cs was purchased from Amersham. The solutions pH was adjusted by NaOH and/or HCl which were attained from El-NasrCo. All chemicals employed in this investigation have analytical grade purity. Aqueous solutions of 100mg/L cesium, strontium, and iodide ions were gained by dissolving their salts in bidistilledwater then labeled with ^{137}Cs , ^{85}Sr , and ^{131}I .

2.2. Adsorbent Preparation

The solid waste adsorbent is granular activated carbon, GAC, gained from the wasted household water filter (from the second stage). It was renewed by 1.0mol/L nitric acid

followed by hydrogen peroxide (2 g solid:1 mL HNO₃ and/or 1 mL H₂O₂) to eliminate any dirt or metal ions adsorbed on its surface. GAC was dried for 3h at 90°C.

2.3. Batch distribution studies

Distribution coefficients (K_d) of ¹³⁷Cs, ⁸⁵Sr, and ¹³¹I radionuclides in solutions on GAC were estimated batch wisely. A series of 0.1g of GAC was shaken with 5.0mL of the radioactive solution of ¹³⁷Cs, ⁸⁵Sr, and/or ¹³¹I for 3.0h and at pH of range from 5.0 to 10.0 for ⁸⁵Sr, and ¹³⁷Cs radionuclides, while for ¹³¹I, the pH range from 1.0 to 5. Centrifugation of mixtures was applied after equilibration then 3mL of the sorbate was analyzed. The radioactivity of ¹³⁷Cs, ⁸⁵Sr, and ¹³¹I radionuclides was measured utilizing NaI scintillation detector connected to the multi-channel analyzer, Genne 2000, USA. All measurements were executed in duplicates. The K_d of each radionuclide was calculated by Eq. (1).

$$K_d \left(mL / g \right) = \frac{A_o - A_e}{A_e} \times \frac{V}{m} \quad (1)$$

where K_d is the distribution coefficient, mL/g, A_o and A_e are the counting of liquid phase (cpm) before adsorption and after equilibration, respectively. V (mL) is the liquid volume, and m (g) is the GAC mass.

2.4. Kinetic studies

To assess the sorption kinetics, the batch was conducted by shaking 0.1g of GAC with 5.0mL of the radioactive solution of ¹³⁷Cs, ⁸⁵Sr, and/or ¹³¹I at different interval times (1–180 min), and optimum pH value. After separation of both phases, the activity was detected in the supernatant as previously mentioned. The quantity sorbed, q_t (mg/g), was estimated as follow;

$$q_t \text{ (mg / g)} = \frac{A_o - A_t}{A_o} \times C_o \times \frac{V}{m} \quad (3)$$

where C_o (mg/L) is the initial ion concentration, it was analyzed by Atomic Absorption Spectrophotometer (Buck Scientific) model 210 VGP, USA. A_o , A_t , m and V were identified above.

2.5. Equilibrium isotherm studies

Concentrations range from 50 to 500 mg/L of ions were employed to investigate the isotherm for the ions onto GAC. A 0.1g of GAC was mixed with 5.0mL of cesium, strontium and/or iodide ions, labeled by their radionuclides. The pH was adjusted to the optimum value (pH 10 for cesium and strontium ions and pH 5.0 for iodide ions), then the mixture was agitated. The activity of the filtrate was determined. The quantity sorbed of ions at equilibrium, q_e (mg/g), was estimated using the previous equation after replacing A_t by A_e .

3. Results and Discussions

3.1. Effect of pH

The pH is the more significant variable governing the ions sorption onto sorbents. This is relatively sorbed since hydroxyl or hydrogen ions themselves powerfully compete with ions. The pH effect on sorption of anions differs from sorption of cations, therefore, the pH influence on the capture of $^{137}\text{Cs}^+$ and $^{85}\text{Sr}^{2+}$ cations was employed at high pH (ranged from 5.0 to 10.0) to minimize the sorption competition between the cations and hydrogen ions. While for ^{131}I , low pH (ranged from 1.0 to 5) was selected to diminish the sorption competition with OH^- ions, the outcomes are illustrated by Fig. 1. For Cs^+ and Sr^{2+} cations, the data confirmed that the percent uptake and the distribution coefficient of both radionuclides rise by raising the pH value. The maximum percent uptake for both

radionuclides was attained at pH 10.0 (87.6 and 85.6% for $^{137}\text{Cs}^+$ and $^{85}\text{Sr}^{2+}$, respectively) as indicated by Fig. 1(a) and (b). While the maximum percent uptake of $^{131}\text{I}^-$ was obtained at pH 1.0 (82.7%) as exposed by Fig. 1(c). Hence, optimum pH was favorite at pH 10.0 for Cs^+ and Sr^{2+} , while for I^- , the optimum pH was favored at pH 1.0.

Fig. 1

3.2. Effect of contact time

Figure 2 displays that the uptake of ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides using GAC rose by increasing time. The quantity sorbed also was computed using Eq. (3), the outcomes outlined that the quantity sorbed rose gradually till reaching the equilibrium to be 4.3, 4.27, and 4.14 mg/g for ^{137}Cs , ^{85}Sr , and ^{131}I , respectively. The sorption rate is high at the beginning, then it is progressively reduced probably because of exhaustion of the available sorption sites and the increase of boundary-layer thickness.

Fig. 2

3.3. Equilibrium isotherm studies

For the optimizing design of a sorption system, it is essential to set the most proper correlations of equilibrium outcomes. The isotherm remains considerable from theoretical and experimental points of view. The knowledge of isotherm nature and its parameters makes it probable to calculate the equilibrium quantity sorbed of adsorbate concentrations outside those employed in this investigation, particularly more diluted ones; moreover, it is needful to scale-up and design the sorption equipment. The adsorption isotherm of ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides using GAC is displayed in Fig. 3.

Fig. 3

3.4. Langmuir model

Langmuir isotherm proposed that all the sites have the same energy and a monolayer ion coverage over a homogeneous surface [19]. Langmuir equation is illustrated as follows.

$$\frac{C_e}{q_e} = \frac{1}{bQ} + \left(\frac{C_e}{Q}\right) \quad (4)$$

where Langmuir constants Q and b describe the capacity, mg/g, and sorption energy, respectively. Plotting of C_e/q_e and C_e gives a high value of correlation coefficient, $R^2 \geq 0.95$, this denotes that Langmuir theory effectively defined the sorption data of ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides using GAC, Fig. 4. Table 1 represents values of Langmuir parameters; the Q values of ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides were 21.7, 21.2, and 20.1 mg/g, respectively. According to these values, the sorption onto GAC follows the order $\text{Cs}^+ > \text{Sr}^{2+} > \text{I}^-$ which is agreed with the order gained from the above studies.

Fig. 4

Table 1

3.5. Freundlich isotherm model

The experimental outcomes can be fitted over a large scale of concentrations by Freundlich description. It provides an expression including the active sites energy and their distribution, and the surface heterogeneity. Freundlich equation is formatted as follows.

$$\log q_e = \frac{1}{n} \log C_e + \log k$$

where k and n refer to the capacity and adsorption intensity, q_e and C_e are defined above. The k and n values are enumerated from intercept and slope, respectively, of $\log q_e$ against $\log C_e$ plot as exposed in Fig. 5. The outcomes illustrated that $1/n < 1$ for all ions as represented by Table 2. This denotes concentration-dependent sorption onto GAC. The R^2 value is less than

that of Langmuir model; therefore, Langmuir isotherm is more applicable for sorption of ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides by GAC.

Fig. 5

Table 2

3.6. Comparison of sorption capacity of Cs^+ , Sr^{2+} and I^- onto different sorbents

The monolayer-capacity of Cs^+ , Sr^{2+} , and I^- by GAC was compared with other sorbents gained in the literature; the outcomes are stated in Table 3. GAC has much higher sorption for the three ions than several materials. It denotes that GAC is recommended to be a promising adsorbent for the capture of Cs^+ , Sr^{2+} , and I^- from the aqueous phase.

Table 3

3.7. Real application study

The qualification of GAC adsorbent for decontaminating solutions containing ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides was tested. The GAC was applied effectively in the capture of ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides in spiked tap water and environmental water samples gained by adding $2\mu\text{g/mL}$ from Cs(I) , Sr(II) , and I(I) ions with the respective radiotracer element. A 10 mL solution was shaken for equilibrium time with 0.2 g of GAC adsorbent at pH 9.0–10.0 for strontium and cesium ions and at pH 1.0–2.0 for iodide ions. Both phases were detached and the filtrate was radiometrically analyzed. Employment of GAC has exposed that ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides can be wonderfully separated from various samples with percent removal in the range of 90.0 to 96.0%, 92.0 to 98.0%, and 72.4 to 85.0 for ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides, respectively, with just one treatment. Table 4 denotes that GAC adsorbent was effectively utilized for the capture of ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides from real water. As for seawater, the removal percentage of ^{137}Cs , ^{85}Sr , and ^{131}I

was low related to other types of water, as seawater contains a high percentage of salts and other ions, which impede sorption of ^{137}Cs , ^{85}Sr , and ^{131}I . From the above data, it can establish that GAC adsorbent can be applied positively as a hopeful material for the decontamination of radioactive ^{137}Cs , ^{85}Sr and ^{131}I from the environment.

Table 4

Conclusion

The GAC as a solid waste adsorbent gained from the wasted household water filter (from the second stage) was effectively employed for the capture of ^{137}Cs , ^{85}Sr , and ^{131}I radionuclides from the aqueous phase. The percent uptake was computed to be 87.6, 85.6, and 82.7 % for ^{137}Cs , ^{85}Sr , and ^{131}I respectively. The ^{137}Cs , ^{85}Sr , and ^{131}I capture from contaminated water was successfully achieved. From the data, it can be established that GAC adsorbent can be operated effectively as hopeful material for the decontamination of radioactive ^{137}Cs , ^{85}Sr , and ^{131}I from the environment and the radioactive liquid waste treatment.

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Conflict of Interest

The authors declare no conflict of interest

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Figures

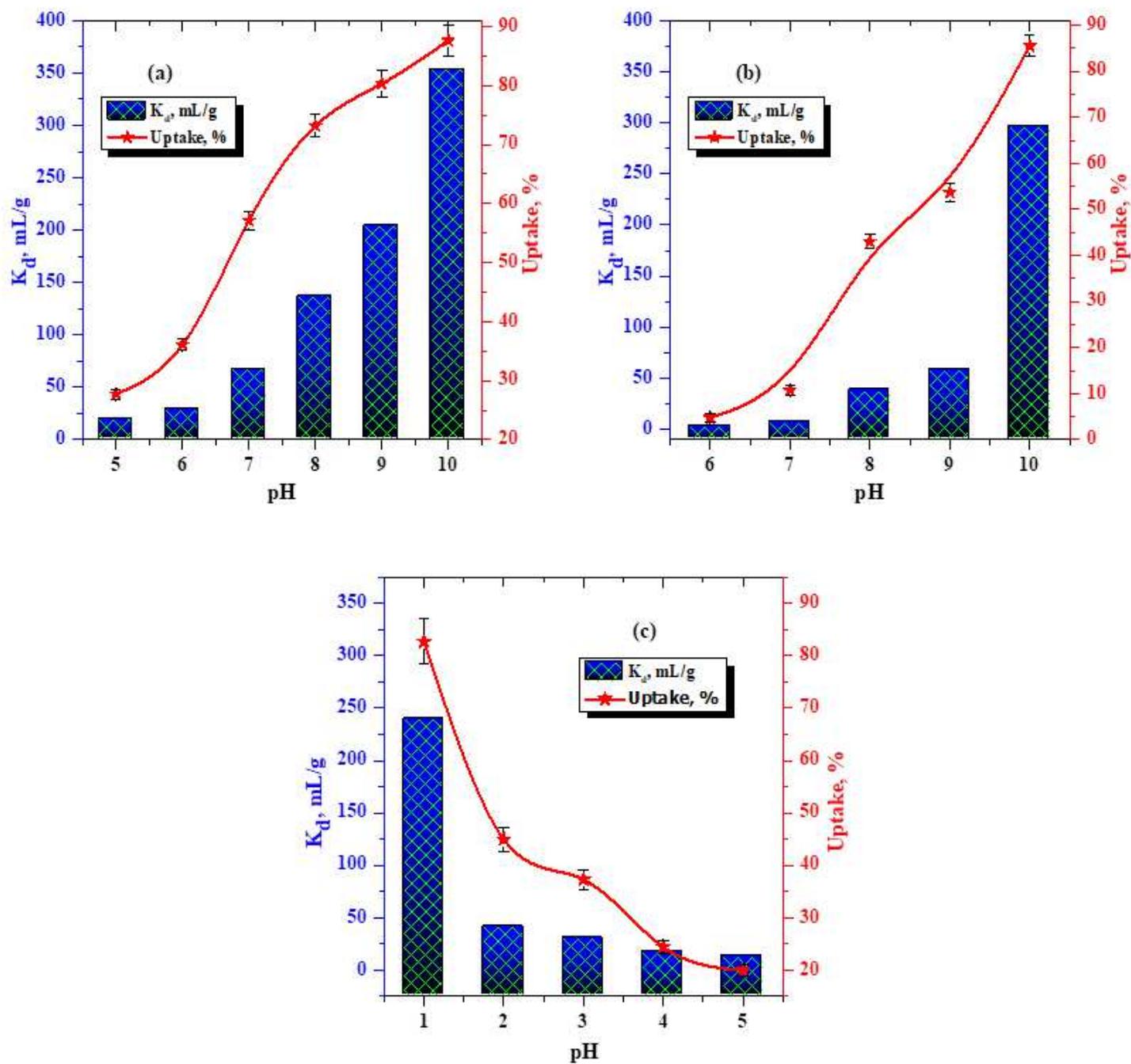


Figure 1

Effect of pH on the removal of (a) 137Cs, (b) 89Sr and (c) 131I using GAC

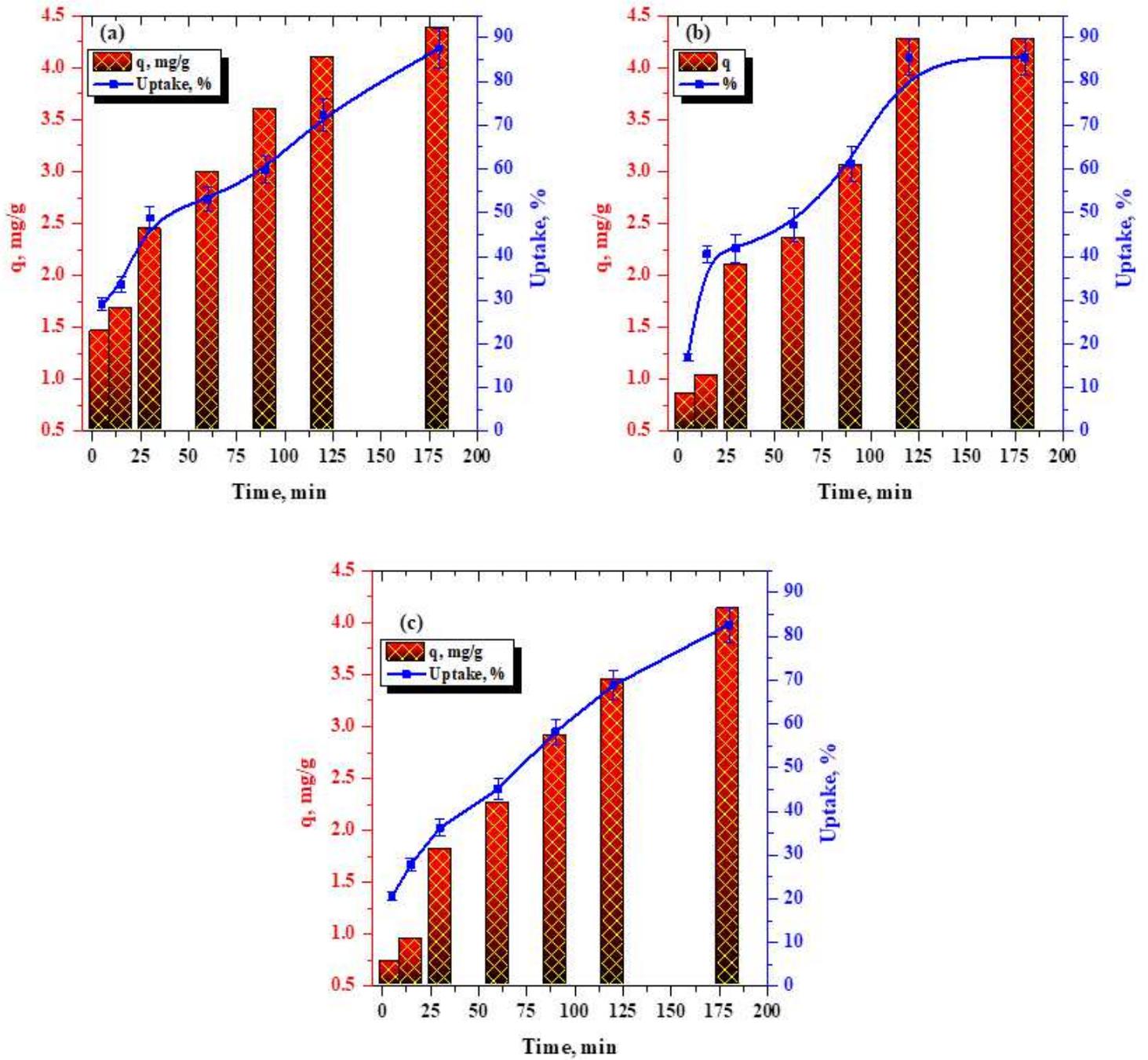


Figure 2

Effect of contact time on the removal of (a)137Cs, (b)85Sr and (c)131I using GAC

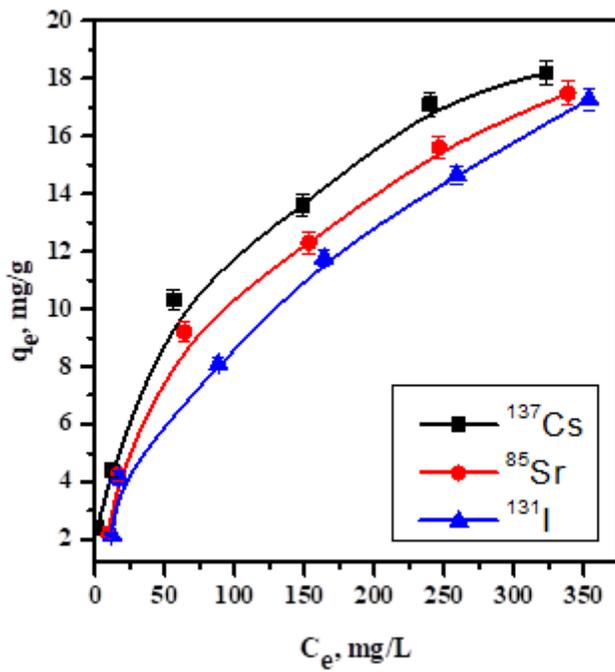


Figure 3

Sorption isotherm plot for the removal of ^{137}Cs , ^{85}Sr and ^{131}I using GAC

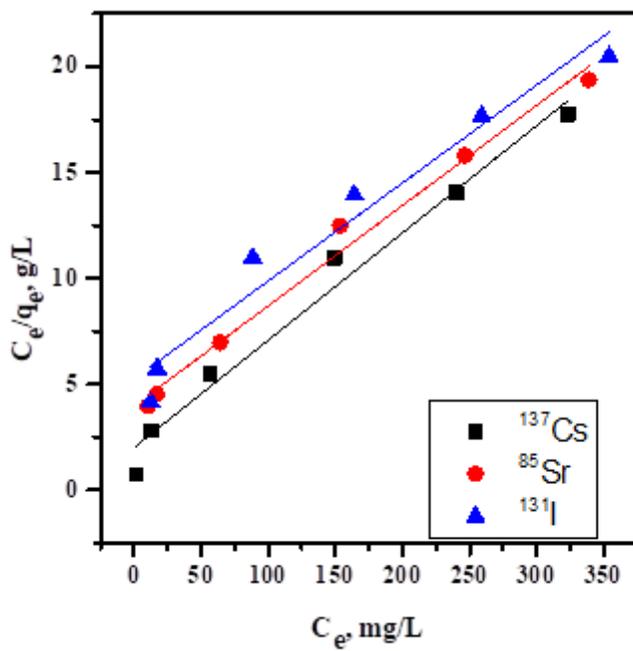


Figure 4

Langmuir plots for sorption of ^{137}Cs , ^{85}Sr and ^{131}I using GAC

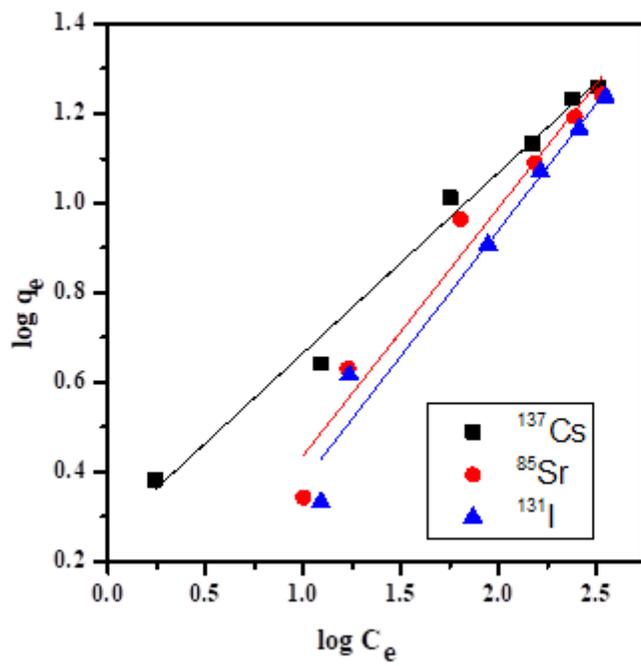


Figure 5

Freundlich plots for sorption of ^{137}Cs , ^{85}Sr and ^{131}I using GAC