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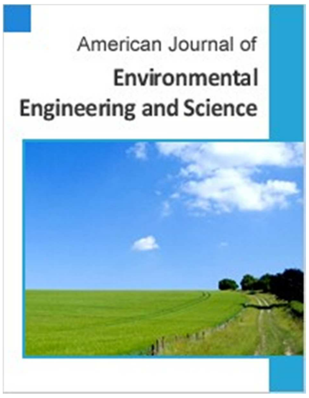

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Assessment of Uptake Coefficient for Potassium-40 Removal from Polluted Water by Packed-Bed Ion Exchange Resins

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Abstract

The removal of potassium-40 from liquid solution polluted with potassium chloride using a cation-ion exchange resin was studied in a packed bed. The efficiency of dowex resin to remove potassium-40 from synthesis wastewater has been tested at different amounts of the resin bed. The kinetic equation has been found to fit well for the study of exchange dynamics. The uptake constant ($p=0.0318\ cm^{-1}$) has been calculated with the help of the kinetic model. The results revealed that the use of cation-ion exchange resin in a packed bed for potassium-40 separation is a promising technique for polluted water treatment in view of the fact that potassium-40 ion removal rates up to 61% were obtained.

1. Introduction

Environmental pollution brought out by rapid industrialization and population explosion is becoming the most challenging threat to the living beings throughput the world. Mostly the areas situated around industrial belts are under stress due to the continuous disposal of untreated wastes from various industries [1]. Effluent limits have become stricter in recent years causing the chemical process industries to upgrade their wastewater. To meet these limits, plants have to employ sophisticated chemical and physical processes because biological treatment alone is not adequate [2].

There are several methods to treat the metal contaminated effluent such as precipitation, ion-exchange, and adsorption, etc., but the selection of the wastewater treatment methods is based on the concentration of waste and the cost of treatment [3, 4].

Ion exchange removes ions from the aqueous phase by the exchange of cations or anions between the contaminants and the exchange medium. Ion exchange material may consist of resins made from synthetic organic materials that contain ionic functional groups to which exchangeable ions are attached. They also may be inorganic and natural polymeric materials [5, 6].

2. Experimental Work

2.1. Materials and Methods

Most ion exchange and adsorption processes are carried out in columns in order to take advantage of the multiple equilibrium steps created by this operation [7]. In order to prepare the radioactive solution of K-40, Known amount of potassium chloride (KCl) is dissolved in a known volume of distilled water, In fact that a small fraction of (0.012%) of natural potassium is radioactive (K-40). The processes of radioactive decay of K-40 produce gamma rays at one energy (1.461 MeV), with a specific activity (31 kBq/kg) of K-40. The long half-life ($T_{1/2}$) of K-40 (1.227×10^9 yr) assists to measure the radioactivity solution at any time after preparation because the K-40 decay is stable [8]. The mixture was stirred for 15 minutes by a magnetic stirrer. The suspended solids were removed from the solution by precipitation and filtration through filter papers. The solution is added to a known amount of ion exchange bed (dowex resin) (Figure 1). A liquates of experiments work available (1500- 2000) ml of radioactive solution are added each time to the column and allowed to pass through it. The flow rate selected is about (~3 mL/min) and adjusted with a screw cap.

The gamma-ray activity concentration of natural potassium in the samples was measured before and after treatment by the gamma-ray spectroscopy system. The apparatus used in this study is illustrated graphically in Figure 2. The ion exchange column consists of a graduated glass cylinder (diameter 2.3 cm) filled with the resin bed to the desired depth. The most efficient ion exchange materials are used for further efficiency characterization. One of the characterization criterions is the depth of the resin, which ranges between (16.85 – 31.3) cm, while the other conditions remain unchanged (Q = 3 ml/min, Di = 23mm, Concentration of ions in solution (46.7) Bq/L)

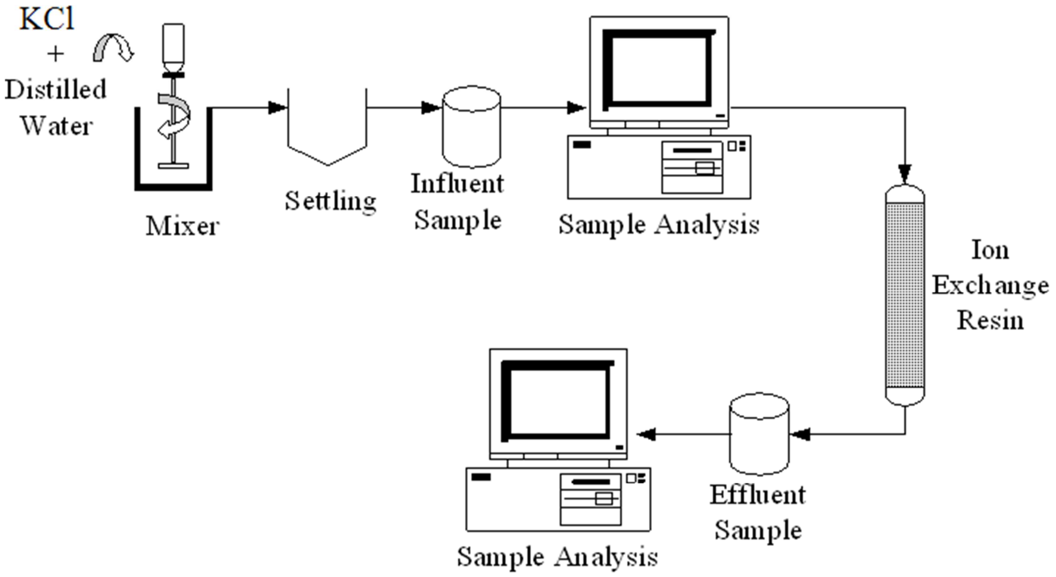

Figure 1. *Schematic diagram of the apparatus used in the study.*

A substance that enters the region has three possible faces (Figure 2): some of it may leave the region unchanged, some of it may accumulate within the boundary, and some of it may be converted to some other substance. The following materials balance equation is written for each substance of interest [9, 10]:

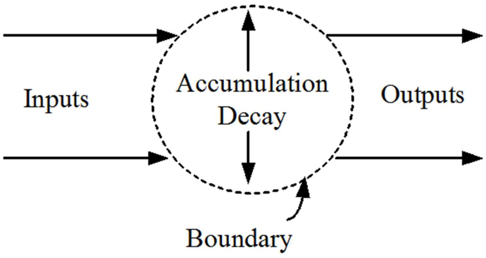

Figure 2. *Materials balance diagram.*

Input Rate = Output Rate + Decay Rate + Accumulation Rate

A common problem in experimental work is to obtain a mathematical relationship y = f(x) between two variables x and y by fitting a curve to points in the plane corresponding to various experimentally determined values of x and y, say [11]:

$$(x_1, y_1), (x_2, y_2), \ldots\ldots.(x_n, y_n)$$

The derivation of the law of exponential change in the effluent concentration as a function of resin depth utilized is adopted as outlined in reference [12]. The procedure for solving the initial value problem is illustrated below:

The effluent K-40 activity concentration (C) from a resin bed with a depth of (y) is decreased at a rate that is proportional to the activity initially present (C_o). If the activity initially present (C_o) is known, the effluent K-40 activity concentration (C) can be found as a function of resin depth (y) by solving the initial value problem as can be seen in formula (1):

Differential equation:

$$\frac{dC}{dy} = p*C \tag{1}$$

Initial condition: $C = C_o$ when $y = 0$.

where p is the uptake coefficient. To find the nonzero solution, divide both sides of formula (1) by (C), yields formula (2):

$$\frac{1}{C}\frac{dC}{dy} = p \tag{2}$$

Integrating both sides of formula (2) yields:

$$\ln\left|C\right| = p*y + cons\tan t \tag{3}$$

Exponentiation of formula (3):

$$\left|C\right| = e^{p*y+Cons\tan t} \tag{4}$$

$$\left|C\right| = e^{cons\tan t}e^{py} \tag{5}$$

Eliminating absolute values in formula (5):

$$C = \pm e^{cons\tan t}e^{py} = Ae^{py} \tag{6}$$

the value of A in formula 6 is found for the initial value problem by solving for A when $C=C_o$ at $y=0$:

$$C_o = Ae^{p*0} \tag{7}$$

The solution of the initial value problem is the simplified form of the second order partial differential equation for penetration of sorbable solutes through a permeable bed is [13]:

$$C = C_o\, e^{-py} \tag{8}$$

in which C is the concentration in the bed at depth y, C_o is the concentration at the surface, and p is the linear uptake coefficient.

The non-linear equation (such as formula 8) can be converted to linear one by means of the natural logarithm rules that simplified arithmetic by replacing multiplication by addition. For any positive numbers a and x and for any exponent n, the rules of arithmetic for logarithms are [12]:

$$\mathrm{Ln}\,(ax) = \mathrm{Ln}\,(a) + \mathrm{Ln}\,(x) \tag{9}$$

$$Ln\left(\frac{a}{x}\right) = Ln(a) - Ln(x) \tag{10}$$

$$\mathrm{Ln}\,(x^n) = n\,\mathrm{Ln}\,(x) \tag{11}$$

The natural logarithm function has an inverse function, the exponential function of x as shown in Equation (12) [9]:

$$\exp\,(x) = \mathrm{Ln}^{-1}(x) \tag{12}$$

When equations (2) and (4) are applied to equation (1), becomes:

$$\mathrm{Ln}\,(C) = \mathrm{Ln}\,(C_o) - p\,y \tag{13}$$

Regression analysis (Least Square Estimate) is used to quantify the linear relationship between two sets of measurements (Ln(C) and y) taken on a single population. Regression analysis finds an estimate of the line of best fit [13]:

$$Y = \bar{a} + \bar{b}X \tag{14}$$

through the scattered points on the graph. To estimate the slope $\bar{b}$ (equal to –p in equation (6)), the below formula was used [13]:

$$\bar{b} = \frac{n\sum_{i=1}^{i=n} X_iY_i - \sum_{i=1}^{i=n} Y_i \sum_{i=1}^{i=n} X_i}{n\sum_{i=1}^{i=n} X_i^2 - (\sum_{i=1}^{i=n} X_i)^2} \tag{15}$$

where n represent a number of points.

Since the line of best fit always passes through the means of X and Y, $\bar{X}$ and $\bar{Y}$, the estimate of the constant $\bar{a}$ can be found by substituting them into the equation to give formula 16:

$$a = \frac{1}{n}(\sum_{i=1}^{i=n} Y_i - b\sum_{i=1}^{i=n} X_i) \tag{16}$$

A computer program written in Quick Basic language is prepared to estimate the regression coefficients (a, b). The flow chart is shown in Figure 3.

To test whether two sets of measurements taken on a single population are linearly associated. The correlation coefficient (R) is given by the formula (17) [14]:

$$R = \frac{\sum_{i=1}^{i=n} (X_i - \bar{X})(Y_i - \bar{Y})}{\left[\sum_{i=1}^{i=n} (X_i - \bar{X})^2 (Y_i - \bar{Y})^2\right]^{0.5}} \tag{17}$$

The correlation coefficient varies from –1 (perfect negative correlation) through 0 (no correlation) up to a value 1 (perfect positive correlation). So the correlation is carried out in a computer package.

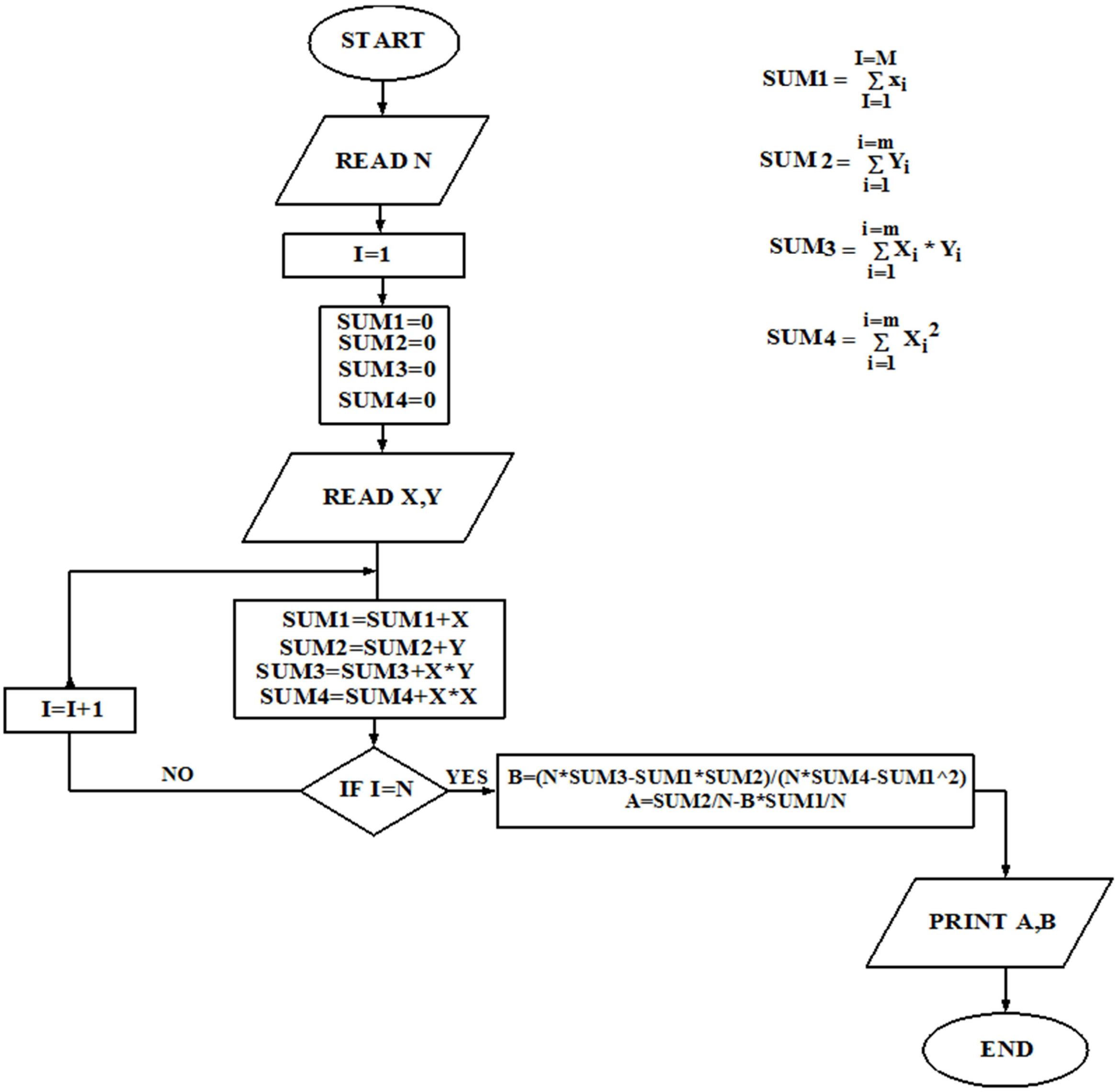

Figure 3. *Block Diagram of Computer Program written in QBasic language for Least Square Estimates.*

2.2. Samples Counting

Gamma spectrometer system based on high purity germanium detectors (HPGe) (Figure 4) was used to determine the activity concentration of K-40 in aquatic samples, before and after the passage of the radioactive solution through packed bed system. The HPGe gamma spectrometric system is composed of:

A high-purity germanium (HPGe) detector (100 cm^3) with an efficiency of 40%, relative to NaI(Tl) scintillator. The detector is surrounded by a graded-z cylindrical shield which consists of lead, cadmium, and copper with a thickness of 5 cm, 0.8cm, and 0.4 cm, respectively, which provides an efficient suppression of background gamma radiation presented at the laboratory site. The energy resolution (FWHM) achieved is 2.5 KeV at the 1.33 MeV reference transition of Co-60.

The detector is vertically mounted into a 30-litre dewar filled with liquid nitrogen. Experimental arrangement for spectra collection includes the high-voltage supplies type TC-243 (Tennelec company) bias supply (+1400 V), equipped with a remote shutdown feature and signal - processing electronics. The latter includes a spectroscopy main amplifier type TC-243 (Tennelec company) which incorporates an efficient pile-up ejector and a multi-channel buffer (MCB) which is a PC-based plug-in PCI card consisting of an 8k ADC. A personal computer analyses with 8192 channel (Tandy, USA), allow data acquisition, storage, display and online analysis of the acquired spectra. The activity of radioactive elements in the standard Canberra source is supplied by the manufacturer, the net counts per second are measured, and a standard calibration curve is presented, to be used in estimating K-40 activity in the samples before and after treatment. Every sample is counted for 3600s. Prior to the samples measurement, gamma background at the laboratory site is determined with an empty Marinelli beaker under identical measurement conditions. It has been later subtracted from the measured gamma-Ray spectra of each sample.

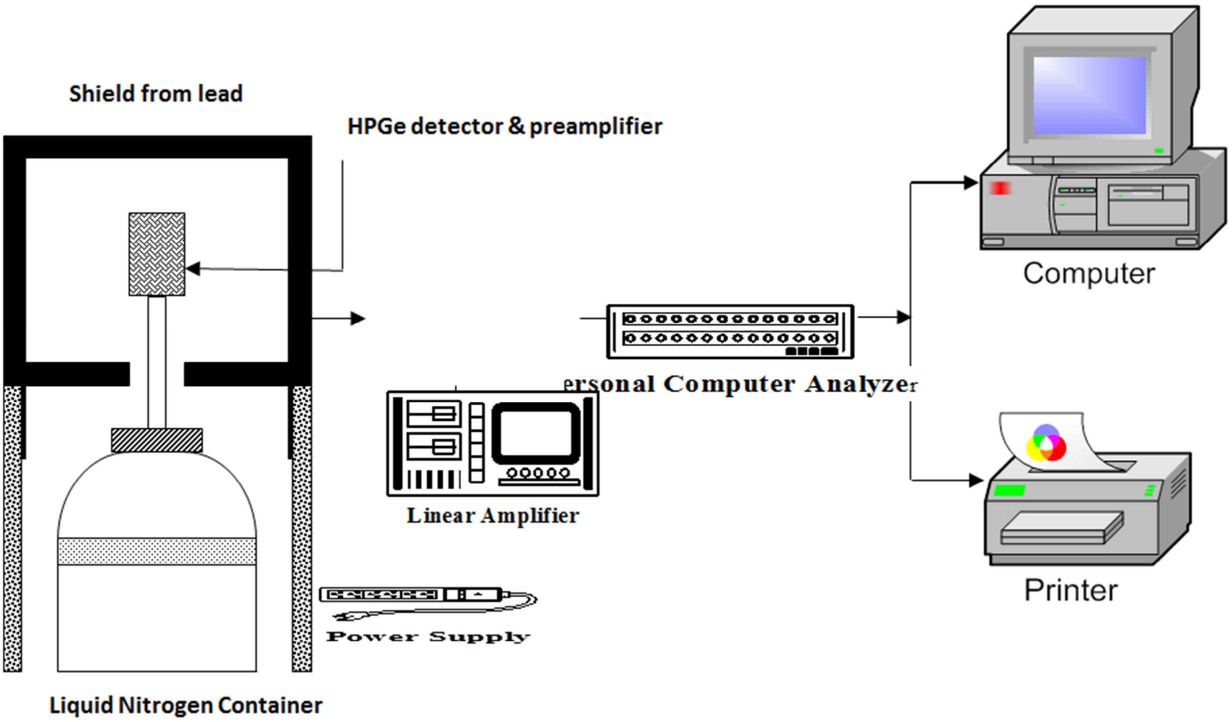

Figure 4. *Gamma ray spectrometry system.*

3. Results and Discussion

The untreated wastewater with a 46.7 Bq/L activity concentration influent of K-40 passed through a bed of resin of a certain depth from (16.85 - 31.3 cm) containing cation resin (dowex) in the hydrogen form. When the impurities in the wastewater came in contact with the resin's bed, an exchange between the ions will take place. The activity concentration of K-40 before and after treatment is given in Table 1. The uptake coefficient ($p=0.0318\ cm^{-1}$) in formula (1) was calculated by the least square method applied to the data given in Table 1. This result indicates that, for removal of K-40 by the ion exchange technology, the relationship between the influent concentration C_o (Bq/L), the effluent concentration C (Bq/L) and the resin depth y (cm) can be well represented by formula (18):

$$C = C_o\, e^{-0.0318y} \qquad (18)$$

Figure 5 shows the kinetic data of removal of K-40 as observed and predicted by equation (1), with a correlation coefficient between observed and predicted data of 99%.

Table 1. *Activity concentration of K-40 before and after treatment.*

Resin depth (cm)	Activity concentration (Bq/L)	
	Influent	Effluent
16.85	46.7	28.9
19.26	46.7	27.8
24.08	46.7	23.1
28.89	46.7	20.9
31.30	46.7	18

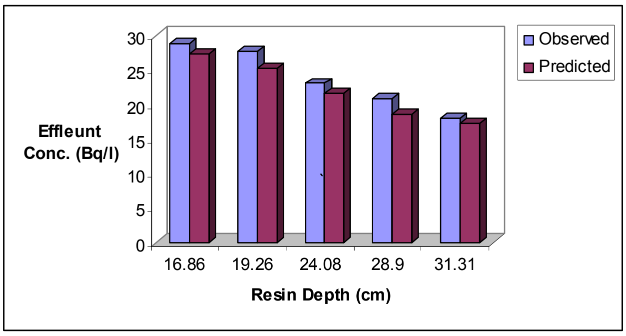

Figure 5. *Comparison between observed and predicted activity concentration of K-40 in the effluent samples.*

4. Conclusions

The use of a cation-ion exchange resin in a packed-bed was found to be an efficient technique for K-40 removal from polluted water with removal efficiency reaches 61%. Due to the same chemical behavior of the radioactive isotope of Ceasium-137 (Cs-137), this method could be effectively used to remove it from the polluted wastewater with this isotope.

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