ASSESSMENT OF DISTRIBUTION COEFFICIENT TO REMOVE THE RADIOACTIVE POLLUTION FROM WASTEWATER BY USING BENTONITE CLAYS

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Abstract

The technology of ion exchange is applied in batch operation for removing potassium-40 as from liquid solutions (prepared in laboratory by solved known amount of KCl in known amount of distilled water. A measured quantity of the ion exchange medium (bentonite clays) is mixed with radioactive solution containing potassium-40 in a suitable mixing vessel. The activity concentrations of liquid solution containing potassium-40 in the influent and effluent samples are measured by gamma ray spectroscopy system composed of a high purity germanium detector with 40% efficiency coupled to an 8192 channel personal computer analyzer. The amount of exchanger media required and the rate of exchange is determined by laboratory tests. The distribution coefficient for bentonite clays for removing K-40 in batch operation (0.104815 l/gm) is estimated by correlation line method, with a correlation coefficient between the observed and the predicted data of 96%. To ensure accuracy and reliability of the collected data, the χ^2 test is used to measure deviations between the observed and expected K-40 activity concentration captured by the bentonite clays and passed through it. No significant difference is observed between observed and expected data at 5% significant level. The results indicate that the distribution coefficient can be used in predicting the amount of an ion exchanger media needed to remove potassium-40 with a certain decontamination factor.

Key words: Distribution Coefficient, Radioactive Pollution, K-40, batch system, Bentonite Clays.

تقييم معامل التوزيع لإزالة التلوث الإشعاعي من مياه المخلفات باستخدام ترب البنتونايت

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الخلاصة:

استخدمت تقنية التبادل ألايوني بأسلوب الخلط لإزالة البوتاسيوم-٤٠ (المشع) من المحاليل الملوثة المحضرة مختبرياً بأذابة كمية معينة من كلوريد البوتاسيوم في حجم معين من الماء المقطر . تم خلط كمية موزونة من مادة المبادل ألايوني (أطيان البنتونايت) مع المحلول المشع المقاس و المعد مسبقا والحاوي على البوتاسيوم-٤٠ في وعاء خلط مناسب . تم قياس تركيز البوتاسيوم-٤٠ في النماذج الداخلة والخارجة باستخدام تقنية تحليل أطياف جاما، يتألف الجهاز من كاشف جرمانيوم عالي النقاوة بكفاءة ٤٠% مرتبط مع محلل حاسوبي شخصي متعدد القنوات. استخدمت عدة تجارب مختبريه في تقدير أكثر من معامل توزيع كمية المبادل ألايوني المطلوبة ومعدل التبادلات. وجد أن معامل التوزيع لأطيان البنتونايت المستخدمة لإزالة البوتاسيوم-٤٠ يساوي 0.104815 لتر/غرام وبمعامل ارتباط بين البيانات ألمقاسه والمتوقعة بحدود ٩٦%. استخدم اختبار مربع كاي لقياس معدل الانحرافات مابين تراكيز البوتاسيوم-٤٠ المقتنصة بواسطة مادة المبادل ألايوني المقاسة والمقدرة بواسطة قانون معامل التوزيع، أشارت النتائج إلى أمكانية استخدام معامل التوزيع في تقدير كمية المبادل ألايوني المطلوبة لإزالة البوتاسيوم-٤٠ وبمعدل إزالة معين.

Introduction

Potassium is a chemical element is commonly found in nature. It is present in soil and drinking water. It is also an essential element for the growth of plants and beings. The concentration of potassium is important in determining the quality of soil in applications such as greenhouses and agriculture. Potassium salts are also a common component of fertilizers (HANNA, 2002). Of the three naturally occurring potassium isotopes, only ^{40}K is unstable, occurs to an extent of 0.017% in natural potassium, imparting a specific activity of approximately 800 pCi/g (31KBq/Kg). Because of its relative abundance and its energetic β emission (1.3 MeV), the radioactivity of ^{40}K is easily the predominant radioactive component in normal foods and human tissues. Radionuclides that occur naturally in soil are incorporated metabolically into plants and ultimately find their way into the bodies of animals and human (Eisenbud, 1973).

Annual limit on intake (ALI) of radioactive potassium-40 by oral ingestion pathway is 300 μCi (US. Nuclear Regulatory Commiss 2003).

Ion exchange is used in removing potassium-40 from radioactive liquid solution, The first application of ion exchange resins for the removal of radioactivity was developed for the separation of the fission products including the rare earth (Nachod, 1956). Ion exchange is a process in which mobile ions from an external solution are exchanged for ions that are electro statically bound to be the functional group contained within a solid matrix (IAEA, 2002). The ion exchange technology is applied in batch operation, which involves vessels in which the exchanger particles are mixed with the liquid stream, followed by settlers in which the two streams are separated to be send later to other stages (Nachod, 1956).

A measured quantity of the ion exchange medium is mixed with the liquid waste in a suitable container. The amount of media required and the rate of exchange can be determined by laboratory tests or by calculation using the following equation (IAEA, 2002):

$$K_d = (DF - 1) * \frac{V}{m} \quad (1)$$

where:

K_d is the measured distribution coefficient,

DF is the required decontamination factor, V is the volume of the liquid to be purified,

m is the amount of the ion exchange medium needed to reach the required decontamination factor.

For liquid volume of 1 liter, equation (1) is rearranged:

$$\mathbf{DF - 1 = K_d * m} \quad (2)$$

The slope of the line of linear relationship between K-40 decontamination factor minus one (**DF-1**) and amount of clays used (**m**)(equation (2)) represents the distribution coefficient, and can be determined from the formula (Volk, 1969):

$$b = \frac{\sum x * y}{\sum x^2} = \frac{\sum m * (D-1)}{\sum m^2} = K_d \qquad (3)$$

Test for Goodness of Test:

Data observed and expected can be tested by statistical methods for deviations from some hypothesized values. χ^2 is a measure of this deviation (Volk, 1969):

$$\chi^2 = \sum \frac{(deviation)^2}{\exp ectation} = \sum \frac{(O-E)^2}{E} \qquad \ldots (4)$$

Where **O** =observed frequency (or concentrations that indicated by measuring device), **E**=expected frequency (or concentrations indicated by the distribution coefficient formula).

Experimental Work:

1 Preparation of Samples

The radioactive solution containing potassium-40 is prepared in laboratory by mixing known amount of potassium chloride (KCl) to known volume of distilled water and use it as wastewater , the gamma-ray activity concentration of natural potassium in the influent samples is measured. Certain amount of an ion exchange media ((0.5-4)gm bentonite clay) is added to the sample, the mixture is stirred for several minutes (15 min.) by magnetic stirrer, **Fig.(1)**. The gamma-ray activity concentration of the effluent sample is measured by the gamma-ray spectroscopy technique after treatment.

2 Samples Counting:

A gamma- ray spectrometric system, **Figure (2)** composed of a pure germanium detector with 40% efficiency (Tennelec, USA), coupled to an 8192 channel personal computer analyzer (Tandy, USA) is used in measuring K-40 activity concentration in the influent and effluent samples. Energy and efficiency calibrations are carried out with a standard Eu-152 source (IAEA, Vienna). The computer code GDR-4 was used to determine the activity concentration of potassium-40 in the samples (Marouf, 2000). Every sample is counted for 3600 s.

Results and Discussion:

Radioactive potassium is used as a tracer in predicting radioactivity removal efficiency in batch tests. A synthetic radioactive wastewater is prepared by mixing measured amount of KCl with distilled water. The untreated water is contacted with the bentonite particles at different dosages in a suitable mixing vessel for sufficient period of time (15 min) to allow ion exchange process to occur between K-40 ions in the synthetic radionuclide wastewater and the H^+ and Na^+ ions in the structure of bentonote clays. After completion of mixing time, the suspended matters are allowed to precipitate in order to separate the exchanged K-40 ions from the effluent samples. Quantitative analysis by the gamma ray spectroscopy system indicates the K-40 activity concentration in the effluent samples. The quantitative analysis of the radioactive samples, both before and after contacting with a measured amount of bentonite clays, is given in **Table (1)**.

The distribution coefficient is estimated by equation (2) to be the slope of the line of linear relationship (correlation line) between DF-1 and dosage (**m**), **Figure (3)**. The assessed value is 0.104815 l/g. Comparison between observed and predicted activity of K-40 captured within the bentonite clays as observed by measuring instrument and indicated by the distribution coefficient formula is shown in **Figure (4)**, with a correlation coefficient between observed and predicted values of 96%.

The χ^2 test is used to measure deviations between the observed and expected K-40 activity concentration captured by the bentonite dosage when mix with it. The influent K-40 activity concentration is divided into two groups: captured and passed activity, the observed concentration is that measured by the gamma-ray spectroscopy system, and the expected is that indicated by Eq.(1). The estimated χ^2 in **Table (1)** for all the run experiments are found to be less than the tabulated χ^2 value (3.841) for 5% significance level and one degrees of freedom.

Conclusions:

(1) The use of a cation-ion exchange bentonite clays in a batch reactor was found to be an suitable technique for K-40 removal from polluted water.

(2) To protect the public health from radiation hazards caused by contamination of water supplies by K-40, the concentration of K-40 in tap water (C_{in}) must be reduced to the maximum permissible concentration (MPC). The amount of bentonite dosage (mg/day) needed to reduce concentration of K-40 in water to the (MPC) of K-40 in water can be well approximated by:

$$m = \left[\frac{C_{in}}{MPC} - 1 \right] \frac{V}{0.104815} \quad (5)$$

where V is the untreated water flow rate (l /day).

Good agreement is noticed between observed and predicted amount of exchange media dosage, with a correlation coefficient of 96%.

References

1- HANNA Instruments General Catalog, HI, Italy, 2002.
2- Eisenbud, Merril, "Environmental Radioactivity", Harcourt Brace Jovanovich Publishers, Academic Press, New York and London, 1973.
3- US. Nuclear Regulatory Commission, "Potassium-40", Private Policy, Site Disclaimer, Last revised Tuesday, August 05, 2003 (Internet).
4- Nachod, F.C., Jack Schubert, "Ion Exchange Technology", Academic Press INC Publishers, New York, 1956.
5- IAEA, "Application of Ion Exchange Process for the Treatment of Radioactive Waste and Management of spent Ion Exchangers", Technical Reports Series No.408, IAEA, Vienna, 2002.
6- Volk, W., "Applied Statistics For Engineers", Mc-Graw Hill Book Co., 2nd Edition, 1969.
7- Marouf, B.A., "Environmental Impact of Depleted Uranium (DU) contamination in Iraq", Conference on the effects of the use of depleted uranium weaponry on human and environment in Iraq, Baghdad, 5-6 November, 2000.

Table (1): Activity concentration of K-40 before and after the treatment by addition of selected bentonite dosages:

Clays Dosage (gm)	Concentration (Bq/l)		Captured Concentration (Bq/l)		Passed Concentration (Bq/l)		χ^2
	Input	Output	Observed	Expected	Observed	Expected	
0.5	52.7	47.1	5.6	2.624	47.1	50.075	3.55
1	52.7	45.8	6.9	4.999	45.8	47.700	0.79
2	52.7	44.4	8.3	9.132	44.4	43.567	0.09
2.5	52.7	42.6	10.1	10.492	42.6	41.757	0.081
3	52.7	39.4	13.3	12.607	39.4	40.092	0.050
4	52.7	37.4	15.3	15.567	37.4	37.132	0.0065

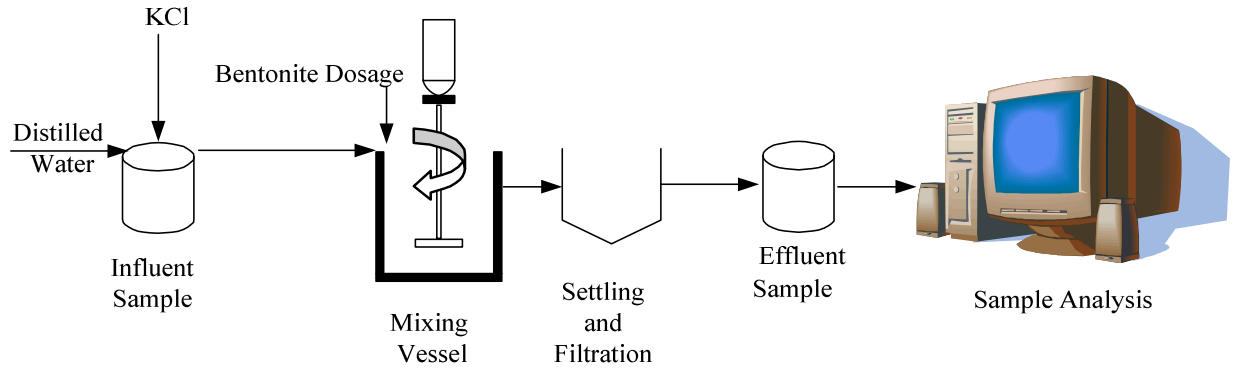

Figure (1): Schematic the batch operation of ion exchange technology

Figure (2): A gamma- ray spectrometry system

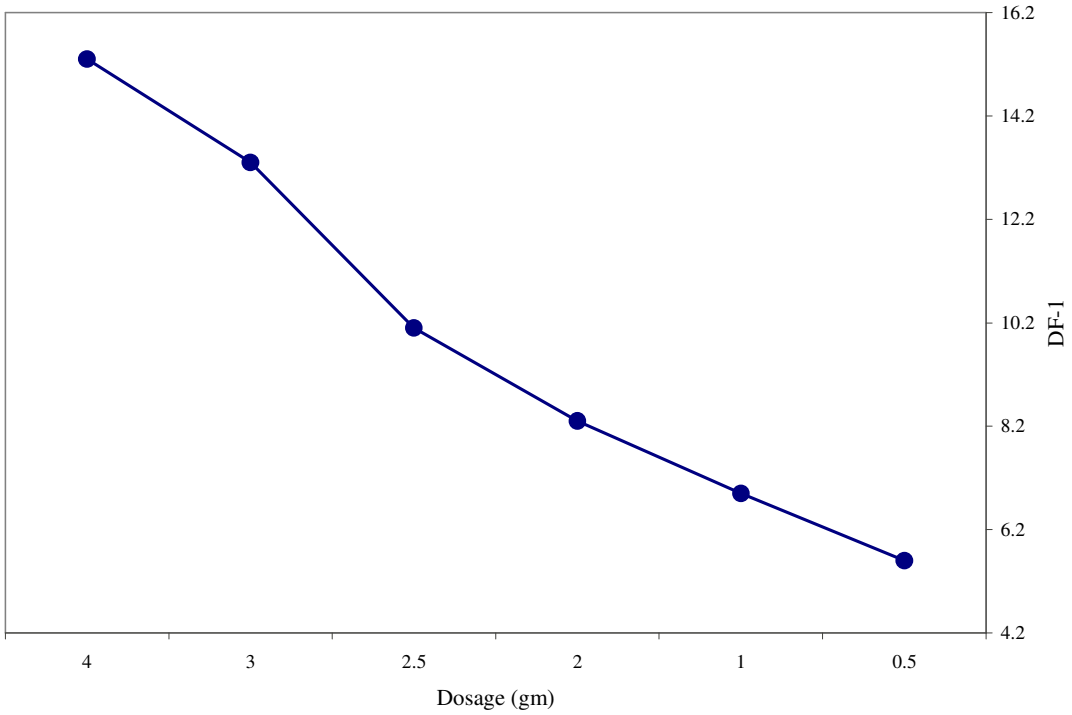

Figure (3): Computation of K_d by correlation line method

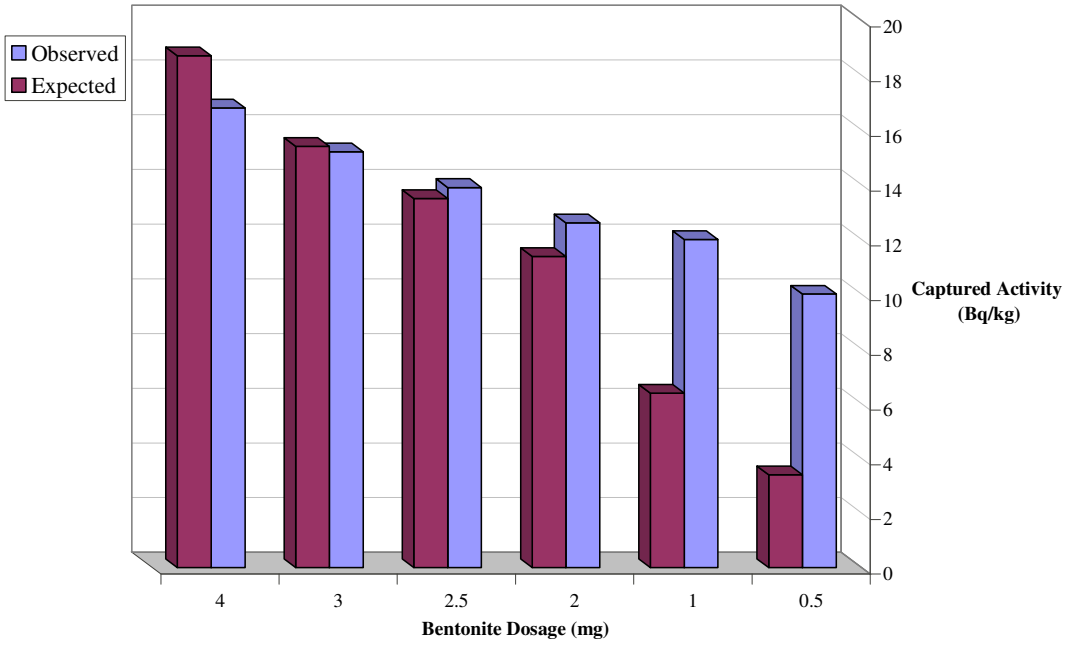

Figure (4): Comparison between observed and predicted amount of bentonite dosage required to achieve a certain decontamination factor.