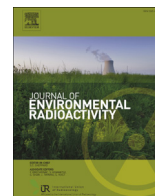




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journal homepage: www.elsevier.com/locate/jenvradRemoval of ^{210}Po from aqueous media and its thermodynamics and kineticsS. Akyil Erenturk ^{a,*}, A. Kilincarslan Kaygun ^b^a Istanbul Technical University, Energy Institute, 34469 Maslak-Istanbul, Turkey^b Cekmece Nuclear Research and Training Center, Yarimbuzurg Mah. Nukleer Arastirma Merkezi Yolu, 34303 Kucukcekmece-Istanbul, Turkey

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ABSTRACT

In this study, the composite adsorbent as granule was prepared by mixing of polyacrylonitrile (PAN) and a natural zeolite (clinoptilolite) in specific conditions. The prepared composite adsorbent was used for investigating the adsorption behaviour of ^{210}Po . Adsorption of ^{210}Po was studied in a column system. The effective parameters such as initial activity concentration of ^{210}Po , pH of the aqueous solution, contact time and temperature of solution for adsorption behaviour of ^{210}Po were studied. Adsorption yield of ^{210}Po on composite adsorbent from aqueous solution in optimum conditions were determined as $75.00 \pm 0.15\%$. The adsorption equilibrium data was examined using various well-known isotherm models such as Freundlich, Langmuir, Dubinin and Radushkevich and Tempkin, and it was observed that the experimental equilibrium data well fitted and found to be in good agreement with the Tempkin model. Adsorption thermodynamics and kinetics of the polonium were studied. It was found that the processes for ^{210}Po were exothermic and spontaneous. The kinetic data conformed better to the pseudo-second order equation.

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1. Introduction

Preconcentration and separation processes based on adsorption processes are important in nuclear technology, industry, medicine and daily life. One of the most effective methods for treatment and disposal of radioactive wastes has been based on adsorbents and ion-exchangers (Meloan, 1999; Humeinici et al., 2004; Nash and Lumetta, 2011).

Many organic and inorganic adsorbents are used in treatment of radioactive and industrial wastes. The composite adsorbents are very special class of adsorbents, developed to improve the processes of separation, concentration or removal of metal ions from aqueous solutions. They have attracted attention as adsorbent materials in recent years (Sebesta et al., 1993; Sebesta and John, 1995; Narbutt et al., 1994; Tranter et al., 2002).

The desirable characteristics of high exchange capacity and favourable selectivity for some radionuclides have made certain zeolites quite useful for the treatment of radioactive wastes (Akyil and Eral, 2005; Kilincarslan and Akyil, 2005; Kilincarslan Kaygun and Akyil, 2007). It is well known that natural zeolites are

aluminasilicates of alkaline and alkaline earth cations with high chemical and radiation stability. Natural and synthetic zeolites have been used for treatment of radioactive waste solution, medium level liquid waste (MLLW) and high level liquid waste (HLLW) resulting from nuclear fuel cycle (Harjula and Lehto, 1986; Borai et al., 2009), for recovery and removal of radionuclides from aqueous solutions and wastes (El Afifi et al., 2016; Munthalia et al., 2015).

Polonium is one of the most dangerous of elements because of its intense radioactivity (Benedik et al., 2009; Skwarzec and Fabisiak, 2007). There is little available information on removal of polonium from aqueous media and its thermodynamic parameters and kinetics (Bagnall, 1973). This study was performed to understand the adsorption behaviour of ^{210}Po and thermodynamic parameters and kinetics of adsorption processes.

The sorption kinetics and thermodynamics in liquid waste or water treatment are very important since it provides valuable insights into the reaction pathways and mechanism of adsorption process.

The purpose of this work is to investigate the ability of the composite adsorbent prepared by mixing of polyacrylonitrile (PAN) and clinoptilolite to remove Po from aqueous solutions by testing the influence of specific process parameters. The applications of the

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isotherm models such as Langmuir, Freundlich, Dubinin–Radushkevich (D–R) and Temkin isotherms were studied to evaluated adsorption characteristics of ^{210}Po onto composite adsorbent. The thermodynamic parameters such as free energy (ΔG°), enthalpy of adsorption (ΔH°) and entropy (ΔS°) were calculated. Furthermore, adsorption kinetics as well as the diffusion parameters for the adsorption of ^{210}Po onto the composite adsorbent were studied.

The sorption study of Po-210 and thermodynamical parameters (activation enthalpy, Gibbs free energy constant and change in entropy constant) reported in this article are useful to understand the sorption of Po onto natural particles in aquatic system and predict the fate and transport of polonium in aqueous environment.

2. Material and methods

Polonium-210 was supplied from Baird Atomic Incorporate. All the reagents used in this work were of analytical reagent grade and were obtained from chemical commercial suppliers.

Natural zeolite clinoptilolite from Manisa-Gordes was obtained from Pamukkale University, Faculty of Science and Literature, Department of Chemistry, Denizli-Turkey. According to the XRD patterns of zeolite, it was obvious that the major phase in zeolite was clinoptilolite mineral (up to 70%) with plagioclase, mica, quartz, smectite, K-feldspar and chlorite (Kilincarslan and Akyil, 2005). The natural zeolite was ground and sieved, and was used as size of –200 mesh for the adsorption experiments.

The composite adsorbents were prepared from natural zeolites as an active component and polyacrylonitrile (PAN) as a binding polymer. The composite adsorbent were prepared in a flask with reflux as a reactor. PAN was solved in 11.86 g of n-dimethylformamide (DMF) at 70 °C for 1 h. Very fine colloidal particles of the natural zeolite clinoptilolite were stirred with the solution of PAN at 1:1 ratio at 70 °C for 1 h to form homogeneous solution. The mixture was fed into the nozzle to obtain the spherical composite beads. Ultrapure distilled water (Millipore) was used as a gelation agent. The obtained composite beads were washed repeatedly by ultrapure distilled water to remove the solvent and then dried at 60 °C for 2 h. Then, the spherical composite beads were sieved and fractionated according to the particle size. The dried composite adsorbent was stored in wide mouth plastic bottle for further use. It was assumed that the composite beads have homogeneous distribution of the inorganic particles in their matrix structure (Akyil and Eral, 2005; Kilincarslan Kaygun and Akyil, 2007).

For the column experiments, a polyethylene column of 7.5 cm in length and 1.8 cm in diameter was used and 0.2 g composite adsorbent beads was packed in the column. The flow rate of the solution (10 mL) was adjusted at 2.5 mL min^{−1}. The effluent collected was counted using ZnS(Ag) scintillation alpha counter (Eberline, SAC-4 model). The detection efficiency of the alpha counting systems were determined as 40% by counting certified U₃O₈ reference material (IAEA-S13). Experimental scheme was given in Fig. 2. The results are the average of at least duplicate independent measurements. Standard deviation never exceeded 5% for each replicate.

Adsorption yield (%) was calculated by using the following expression:

$$\text{Adsorption yield (\%)} = \left[\frac{(A_i - A_f)}{A_i} \right] \times 100 \quad (1)$$

where A_i is the activity concentration of the initial solution (Bq mL^{−1}), A_f is the activity concentration of the solution in equilibration (Bq mL^{−1}).

The adsorption data was subjected to four different adsorption isotherms, namely Langmuir, Freundlich, Dubinin–Radushkevich and Temkin to explain the adsorption equilibrium data. The adsorption isotherms were obtained by analysing solutions in contact with composite adsorbent before and after equilibrium and plotted in terms of the equivalent fraction of polonium ions in the composite phase against the equivalent fraction in the initial polonium solution at the ambient temperature. Linear regression is frequently used to determine the best-fitting isotherm, and the applicability of isotherm equations is compared by judging the correlation coefficients.

Thermodynamic parameters were calculated to assess the spontaneity of the sorption process. The influence of temperature variation from 293 K to 323 K was examined on the sorption of polonium ions of fixed concentration 0.47 Bq mL^{−1} onto composite adsorbent. The thermodynamic parameters such as Gibbs free energy change (ΔG°), heat of sorption (ΔH°) and entropy change (ΔS°) for sorption of polonium ions on composite adsorbent were calculated for the system.

Kinetic studies were carried out in order to investigate the relationship between contact time from 1 min to 180 min and polonium adsorption at 30 °C. The experimental data obtained for adsorption of polonium ions onto composite adsorbent were modelled using various kinetic models including the pseudo first-order, the pseudo second-order and Elovich models.

3. Results and discussion

3.1. Effectual parameters on ^{210}Po sorption onto composite adsorbent

The effects of various parameters such as initial activity concentration of polonium, pH of the adsorptive solution, contact time and temperature were investigated.

3.1.1. Effect of initial concentration of ^{210}Po

The initial concentration of metal has a crucial impact on the adsorption capacity of adsorbent. For isotherm studies, the relative polonium sorption on the composite adsorbent as a function of initial polonium activity concentration was studied from 0.16 to 1.40 Bq mL^{−1} at 30 °C. Fig. 3 shows the effect of initial polonium activity concentration on adsorption. ^{210}Po adsorption increased with increasing the initial activity concentration ranging from 0.16 to 1.40 Bq mL^{−1}. It was observed that the adsorbent attained equilibrium at 0.47 Bq mL^{−1} as 66.8% sorption. On the basis of these results, 0.47 Bq mL^{−1} of ^{210}Po for composite adsorbent was used for all further studies. Amount of adsorbed polonium at equilibrium was found as 14.0×10^{-5} ng g^{−1}.

3.1.2. Effect of pH of the adsorptive solution

Earlier studies on metal sorption have shown that the pH value of the solution has been identified as important controlling parameters for the sorption of radionuclides on the adsorbents, and influences the metal speciation and surface metal binding sites because of changing the adsorption chemistry of adsorbent–adsorbate. The effect of pH on the adsorption capacity of composite adsorbent was investigated using solution of 0.47 Bq mL^{−1} ^{210}Po for a pH range from 2 to 9, while keeping all other parameter constants. The pH of the solution was adjusted with 1 M NaOH and 1 M HNO₃. Fig. 4 shows the influence of pH on the adsorption of ^{210}Po on the composite adsorbent. It reveals that the highest adsorption levels (66.7%) for composite adsorbent between pH 4.0 ± 0.1 and 6.0 ± 0.1 which indicate that a high affinity for polonium ions is predominant in this region. At initial pH lower than 4.0, as a result of protonation, the functional groups on the

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