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Chemical behavior of cobalt and cesium in the presence of inorganic components of a semiarid soil using water of nuclear purity

M. Jiménez-Reyes, M. Solache-Ríos*

Departamento de Química, Instituto Nacional de Investigaciones Nucleares, Carretera México-Toluca s/n, La Marquesa, Ocoyoacac, Edo. de México C. P. 52750, Mexico

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ABSTRACT

The inorganic components of soil from a semiarid region were separated and characterized. Kinetics of adsorption of cesium and cobalt ions by the inorganic components of the soils behave according to the kinetic model of pseudo-second order, indicating that the adsorption process is chemisorption. Equilibria were quickly reached in ca. 1 min, $98.6 \pm 0.5\%$ of cesium and $96 \pm 1\%$ of cobalt were retained in the solid from solutions. The sorption isotherm data, q_e vs. C_e , were best adjusted to the Langmuir model. The adsorption capacities for cobalt and cesium are similar from pH 4 to 8 and at higher pH cobalt precipitates as hydroxide. Ionic strength plays an important role in adsorption; even low concentrations of sodium (<0.1 M) virtually avoid the adsorption for cesium and cobalt. The thermodynamic parameters indicated that the adsorption processes were exothermic for both cesium and cobalt by the inorganic materials of soil.

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1. Introduction

Environmental contamination caused by radionuclides is a serious problem worldwide (Gavrilescu et al., 2009), and they exist either naturally or artificially. Sediments at nuclear facilities have been analyzed and different isotopes have been found like fission-produced ^{137}Cs , ^{90}Sr and ^{134}Cs ; activation-produced ^{60}Co , ^{58}Co , ^{54}Mn , and ^{65}Zn , and the transuranium isotopes ^{239}Pu and ^{241}Am . These radionuclides were attributed mostly to discharges at the monitored facilities. Some ^{137}Cs , ^{90}Sr , and ^{239}Pu are fallout from nuclear tests in the atmosphere. Naturally occurring radionuclides uranium plus progeny, thorium plus progeny, and ^{40}K also have been detected (Rosson et al., 2001). Radionuclides in the environment can, therefore, eventually be passed on to human beings through food chains, and so many represent an environmental threat to the health of local population (Howard

et al., 1991). Remediation of radionuclide-contaminated soil has become increasingly important (Zhu and Shaw, 2000) and the geochemical behavior of radionuclides is interesting from the point of view of chemical industry wastes and the security of long-term deposits of radioactive waste (Sun et al., 2014). Removal of the contaminated surface soil or immobilization of radionuclides in soils by applying mineral and chemical amendments are physically difficult and not likely cost-effective in practicality (Zhu and Shaw, 2000). The interaction of radionuclides behavior in soil is of particular importance to apply the most efficient methods for decontamination (Cremers et al., 1988).

Inorganic components of soil include sand, clay, and limestone. The organic components are fulvic acids (FA), soluble in solutions of pH = 1; humic acids, which are the most abundant and soluble in solutions of pH = 10; and humin (insoluble in both pH 1 or 10 solutions), which may remain with the

* Corresponding author. Tel.: +52 5553297200x12280; fax: +52 5553297301.

E-mail address: marcos.solache@inin.gob.mx (M. Solache-Ríos).<http://dx.doi.org/10.1016/j.psep.2016.04.003>

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inorganic components when the separation is based on alternating the pH of solutions. Water from the cooling system of a nuclear reactor is exhaustively purified and virtually free of solids and ions, and then its ionic strength is near to zero.

Therefore, this work was performed using water from the cooling system of a nuclear reactor in order to examine the retention behavior mechanisms of cesium and cobalt, dissolved in high purity water, by the inorganic components of a semiarid soil. The purposes were to determine: (1) the kinetic and isotherms, (2) the ionic strength effect, and (3) the thermodynamic parameters of both systems.

2. Materials and methods

2.1. Inorganic components of soils

Samples of a soil from a semiarid zone were used for the present study. The details of separation of the inorganic components and their characterization were previously reported (Jiménez-Reyes and Solache-Ríos, 2014). This material contained only ca. 1% of organic material and the main inorganic components were two feldspars (disordered albite and calcium ordered albite), quartz, and montmorillonite. In addition to the elements of these minerals (Na, Mg, Al, Si, and Ca), Fe (3.4 ± 3.3%), Ti and Mn (ca. 0.2% each) were quantified. The size particles were: 62–177 μm.

Determination of the zero charge point (ZCP) of the inorganic material of soil (IM) was carried out with 0.1 M NaClO₄ solutions whose pH values were previously adjusted between 1 and 10, with intervals of 1 unit by adding 0.1 M HCl or NaOH solutions. After 30 min of contact, the systems were centrifuged and decanted and pH was then measured in the final liquid phases with an equipment pH STAT Controller MeterLab PHM 290.

2.2. Aqueous solutions

All solutions were prepared with water from the cooling system of a nuclear reactor. This water was characterized (Solache-Ríos et al., 2015) and the analysis indicated that it is of high purity (pH = 5.83, hardness: 1.99 mg/L, Cl⁻ = 11.7 mg/L) and the ionic strength is ca. 3.3 × 10⁻⁴ M. Initial pH values of both cesium or cobalt solutions were 5.9 ± 0.1.

Solutions of cesium and cobalt were labeled with the isotopes ⁶⁰Co (t_{1/2} = 5.3 years) or ¹³⁴Cs (t_{1/2} = 2.1 years). The isotopes were obtained by irradiating 1 mg/mL of cobalt (as nitrate salt) or cesium (as sulfate salt) solutions (Merck KGaA, Germany), in the nuclear reactor TRIGA MARK III of the Nuclear Center of Mexico, with a neutron flux of 9.9 × 10¹² cm⁻² s⁻¹ for 4 min. These isotopes were identified by their gamma radiation spectra (detector GeH coupled to a multichannel analyzer).

2.3. Sorption: kinetics and isotherms

Kinetic experiments: IM (0.05 g) were shaken with solutions of cesium or cobalt (5 mL) labeled with the radioactive isotopes from 1 min to 4 h. After centrifuging at 6000 rpm for 5 min, aliquots were taken for the measurements of radioactivity, which were performed with a gamma radiation detector of NaI(Tl) coupled to a monochannel analyzer. The reference was a solution containing the same amounts of ⁶⁰Co or ¹³⁴Cs used in each experiment, whose radioactivity was measured in the same experimental conditions. All experiments were done by

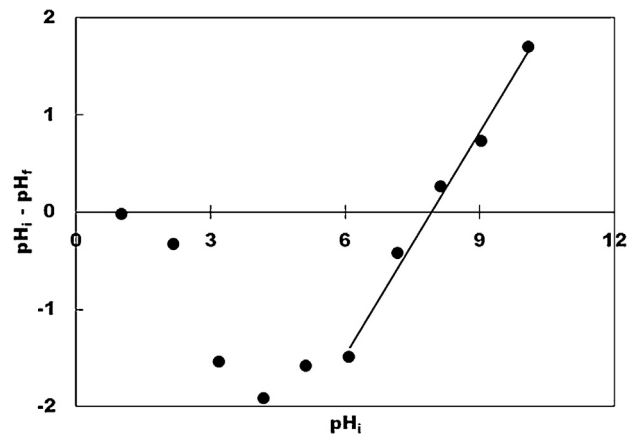


Fig. 1 – Determination of ZCP. The equation of the linear portion is: $(\text{pH}_{\text{initial}} - \text{pH}_{\text{final}}) = 0.77(\text{pH}_{\text{initial}}) - 6.1$ ($R^2 = 0.99$).

duplicate. Equilibrium pH values were 7.4 ± 0.5 and 7.1 ± 0.1 for cesium and cobalt, respectively.

Isotherm experiments: 0.05 g of material and 5 mL of solution were shaken for 30 min and the [Cs⁺]_{initial} or [Co²⁺]_{initial} ranges were between 19.6 and 90.9 mg/L. Adsorption isotherms were obtained at 291, 303 and 313 K in order to determine the thermodynamic parameters.

The sorption capacities of the inorganic materials for cesium or cobalt ions for a specific contact time (q_t) or at the equilibrium (q_e) were calculated as follows:

$$q_t \text{ or } q_e = \frac{m(M)_I - m(M)_F}{m_{\text{IM}}} = \frac{m(M)_{\text{IM}}}{m_{\text{IM}}} \quad (1)$$

where $m(M)_I$ and $m(M)_F$ are the initial and final mass (mg), of Cs⁺ or Co²⁺ in solution, respectively; $m(M)_{\text{IM}}$ is the mass (mg) of Cs⁺ or Co²⁺ adsorbed by IM; and m_{IM} the mass of the IM in grams (0.05 g). Adsorbed mass of Cs⁺ or Co²⁺ was calculated considering the known initial mass and the radioactivity measured for each experiment and the reference; loss of radioactivity on the polyethylene tubes was not observed.

2.4. Effect of pH

In order to check the effect of pH on cesium and cobalt uptake by IM, the experiments were carried out with 5 mL of a 19.6 mg/L solution of cesium or cobalt and 0.05 g of adsorbent. The initial pH values of solutions (2, 4, 6, 8 and 10) were adjusted by adding HCl or NaOH solutions and the mixtures were shaken until the equilibrium was reached.

2.5. Effect of ionic strength

IM (0.05 g) were shaken with 5 mL of sodium chloride solutions (0.035, 0.07 or 0.1 M) containing 74.1, 82.6 or 90.9 mg/L of cesium or cobalt.

3. Results and discussion

3.1. ZCP determination

The pH_{eq} values for the sets of experiments with 0.1 M NaClO₄, showed changes of pH (Fig. 1). The zero charge point for IM was determined when the pH_{initial} - pH_{final} = 0 and it was 7.9. Different pH_{ZCP} values ranged from 4.7 to 6.3 have been reported

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