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^{63}Ni and ^{57}Co Uptake and Selectivity of Tin Antimonates of Different Structure

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ABSTRACT

Tin antimonates of different structure were studied for their uptake of cobalt and nickel in the decontamination of floor drain water and neutral bond water simulates. Selectivity was observed to vary with the structures and degree of crystallinity of the tin antimonates (pyrochlore, rutile, and mixed metal oxides). High selectivity for cobalt of exchangers with pyrochlore structure is attributed to ion exchange of cobalt inside the tunnel structure; nickel uptake evidently took place mostly at the outer surfaces of the materials. Electrostatic forces governed the ion exchange of rutile tin antimonates owing to their particle hydrate structure. With some limitation, the prediction of nickel uptake on tin antimonates from the more abundant cobalt uptake data is considered possible.

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INTRODUCTION

Tin antimonates have attracted considerable interest for their potential as ion exchangers in radioactive waste management. They possess suitable physical characteristics, good resistance to decomposition at elevated temperatures and under ionizing radiation, and chemically, they are stable and withstand strongly acidic media.[1,2] Ion exchange properties of tin-substituted antimony oxides and antimony-substituted tin oxides, usually called tin antimonates, have been investigated as ion exchangers for the major fission nuclides, such as ^{137}Cs and ^{90}Sr and some actinides. In these experiments, tin antimonates showed good ion exchange properties, i.e., high selectivity for certain elements and relatively high ion exchange capacities (2 mEq g^{-1} for Na^+).[1,3–6]

Although a considerable number of articles dealing with the ion exchange properties of tin antimonates have been published, some important elements have gone almost unnoticed. Separation of activation/corrosion elements (e.g., Co, Ni, Fe) originating from nuclear power plants has attracted only minor attention in ion exchange research. Although some research dealing with cobalt uptake on tin antimonates have appeared, studies on nickel uptake are almost nonexistent. Only a few distribution coefficients have been reported for nickel, and merely as the supplementary results of larger studies.[3,7] Yet ^{63}Ni, with its reasonably long half-life (100 a), will be the major source of radioactivity from activation/corrosion products about 20 years after shut down of nuclear reactors. The question of activation/corrosion products is becoming more relevant now that many nuclear reactors are approaching their licensed operation time and decommissioning will occur in the near future. The decommissioning will produce large volumes of acidic waste solution that has to be dealt with evermore-stricter waste effluent regulations. Comparison of the data reported for nickel and cobalt uptake on tin antimonates suggests that cobalt uptake data, which is more abundant, might be used to predict nickel uptake. Besides their similarities in nickel and cobalt uptake, tin antimonates also exhibit considerable differences, in selectivity, for example.[8,9]

In this study, we investigate the reasons for the marked difference in the selectivity coefficients of cobalt and nickel and consider whether cobalt uptake data could be used for estimating nickel uptake. In addition, the possibility of using tin antimonates for decontaminating floor drain water from activation/corrosion products and the possibility of separating cobalt and nickel from each other were of interest. Also of interest was the influence of pH and the chemical form of the exchanger on the metal uptake.

EXPERIMENTAL

Ion Exchange Materials

All the ion exchangers (tin antimonates) were synthesized from acidic metal chloride ($SnCl_4$ and $SbCl_5$) solution by precipitation with NH_3 solution. The synthesized materials could be divided into three categories on the basis of their structures: materials with pyrochlore structure, materials with rutile structure, and materials including both structures. The rutile structure was identified as a solid solution of tin dioxide (space group $P4_2/mnm$), with antimony substitution up to ~35%. The structure of the pyrochlore materials was similar to the structure of antimony pentoxide (space group Fd3m), with tin substitution up to ~35%. Materials with from 35% to 65% Sn/Sb substitutions were more or less amorphous in structure, since they were mixtures of the two metal oxides at various metal substitution levels (Table 1 and Figure 1). Low crystallinity was favored because of the possible positive effect of this on the ion exchange properties of the materials.[3] The synthesis and structures of the materials are described elsewhere.[8]

For some of the batch experiments the exchanger was changed into calcium form before the experiment by sequential shaking in 0.1-M $Ca(NO_3)_2$ solution and decanting. The pH of the solution was adjusted to ~7 with 0.01-M $Ca(OH)_2$ and shaking was continued till the equilibrium pH was neutral. Otherwise, the materials were used in their original form without further pretreatment after synthesis.[8]

Characterization

The ion exchangers were characterized by x-ray methods. Powder x-ray diffraction (XRD) patterns were collected with a Phillips PW 1710 powder

Table 1. Sample codes, Sn content as metal percentage and structure of the studied tin antimonates.

Sample code; Sn content (%)	Structure	Sample code; Sn content (%)	Structure
SnSb(P_1), Sn 0	Pyrochlore	SnSb(A_4), Sn 46.4	Amorphous
SnSb(P_2), Sn 21	Pyrochlore	SnSb(A_5), Sn 63.1	Amorphous
SnSb(A_1), Sn 34.1	Amorphous	SnSb(R_1), Sn 67	Rutile
SnSb(P_3), Sn 38.5	Pyrochlore	SnSb(R_2), Sn 83.1	Rutile
SnSb(A_2), Sn 42.1	Amorphous	SnSb(R_3), Sn 93.1	Rutile
SnSb(A_3), Sn 45.8	Amorphous	SnSb(R_4), Sn 100	Rutile

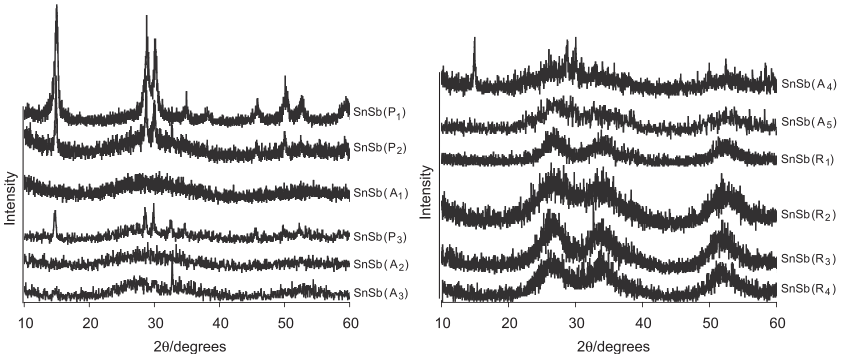

Figure 1. XRD patterns of tin antimonates used in the study.

diffractometer (operating at 30 kV and 50 mA) with Cu K_α (1.54 Å) radiation. Elemental analysis of the metals was carried out by x-ray fluorescence (XRF) technique using an Amptek 7mm^2 Si(Li) XR-100CR detector and 500 mCi ^{241}Am excitation source. Solid samples were used for the XRF measurement to avoid errors due to dissolution of metal oxides of low solubility.

Ion Exchange Studies

The ion exchange properties of the tin antimonates were studied by measuring the distribution coefficients (K_d) for cobalt and nickel in batch experiments and the breakthrough levels of the radionuclides in dynamic column experiments.

Batch Experiments

The K_d values were used to represent the selectivity of the material for metal (Co and Ni) ions. The selectivity determines how efficiently a certain element is separated from other elements. In applications such as radioactive waste treatment, these other elements are usually present in vast concentrations and they compete for the ion exchange sites of the exchanger. Because of this, the K_d values, i.e., the selectivity of the exchanger for radionuclides, must be high for meaningful separation to occur. In batch experiments, 25 mg of finely ground (sieved between 74 and 149 μm) ion exchange material was placed in a polyethylene vial with 10 mL of test solution. Samples were equilibrated for 3 days in a constant rotary mixer (50 rpm), during which time, the solid/solution system reached equilibrium. The solid phase was then separated by centrifuging

(3000 *g*) the vials for 10 minutes and 5-mL subsamples were pipetted and filtered through a 0.2-μm filter (I.C. Arcodisc, Gellman Sciences) for measurements of cobalt concentration. Subsamples of 2 mL were taken for measurements of nickel concentration. The equilibrium pH was measured in the remaining solution.

The distribution coefficient tells how the element of interest is distributed between the initial sample solution and the solid material after a certain equilibration time (3 d). It was calculated as follows:

$$K_d = \frac{(A_i - A_{eq})}{(A_{eq})} * \frac{V}{m} \tag{1}$$

where A_i = initial metal concentration of the solution, A_{eq} = metal concentration of the solution at equilibrium, V = volume of the solution, m = mass of the solid material. In the case of radioactive tracers, the activity of the tracer can be used instead of element concentrations. Here, gamma-emitting ^{57}Co and beta-emitting ^{63}Ni were used as radioactive tracers. An automatic gamma counter (Wallac, 1480 Wizard 3") and a liquid scintillation counter (Wallac 1217 BackBeta) were employed for the radioactivity measurements.

Column Experiments

In the column experiments, a minicolumn (BioRad, Bio-Scale MT2 with radius of 3.5 mm and 28-μm bed support) was packed with 0.4 mL (0.5 g) of SnSb(A_3) exchanger (capacity of ~2.1 mEq g^{-1}). The exchanger was preconditioned by pumping ~4000 bed volumes (BV) of inactive test solution through the column before adding the radioactive tracers to the solution. Sample SnSb(A_3) was chosen for the experiment in view of its good tolerance for calcium ions and acidic conditions, as determined in earlier experiments.[8,9] The pumping rate was set to ~30 BV per hour, and a fraction collector collected the effluent with a time interval of 60 minutes. The pumping rate of 30 BV h^{-1} can be considered high since ion exchange reactions in this type of inorganic exchanger are known to be relatively slow.[10] Metal (^{57}Co and ^{63}Ni) concentrations of the effluent fractions were measured in the same way as in batch experiments. The pH values of the fractions were also measured.

Test Solutions

The solutions for batch experiments were chosen so that ion exchange data from acidic (0.1-M HNO_3) and from competing ion [0.01-M $Ca(NO_3)_2$] conditions could be obtained. In view of the binary ion exchange system

($2H^+ \rightarrow M^{2+}$) and almost constant equilibrium pH of the batch experiments in nitric acid solution, nitric acid solution was the preferred media for comparative experiments with the different materials. A 0.01-M $Ca(NO_3)_2$ solution was chosen as crude simulate for floor drain water of nuclear power plants because Ca^{2+} is known strongly to interfere with radionuclide uptake from the most typical ions (Na^+, K^+, Mg^{2+}).[11,12]

The distribution of the tracer metals was studied one at a time in batch experiments but in column experiment both tracers were present and measured from the same experiment.

The solution in the column experiment was neutral bond water simulate, where in addition to calcium (1.5 ppm) also sodium (100 ppm) and magnesium (0.7 ppm) were present. This solution was used to simulate the storage water of spend nuclear fuel that is constantly purified by normal ion exchange resins. The pH of the simulate (the feed solution) was lowered during the column experiment through the addition of small amounts of nitric acid of appropriate concentration to achieve decreases in effluent pH.

The concentrations of the tracer metals (^{57}Co and ^{63}Ni, carrier free) varied slightly between the experiments but typically were in the order of 10^{-13} to 10^{-15} M, and the speciation of the metals was calculated by HydraQL[13] to be almost entirely ionic (M^{2+}) in all test solutions.

RESULTS AND DISCUSSION

The selectivity of tin antimonates has been found to be good for nickel and excellent for cobalt ions.[8,9] Plotting the distribution coefficients (K_d) obtained from the 0.1-M HNO_3 solution in the same graph (Figure 2) showed that the selectivities for the two metals change in a similar manner as a function of Sn content. Although the K_d values of Co and Ni are at different levels, the values change in almost identical fashion for the rutile-structured materials and in closely similar fashion for the pyrochlore-structured materials. The relation of Co and Ni K_d values of the same tin antimonate changed most with mixed metal oxide materials, particularly with materials of amorphous structure.

Effect of Tunnel Structure on Selectivity of Pyrochlore Materials

Physical properties of the exchanging ions (Table 2)[14] and steric hindrance of the exchangers were considered as the most probable reasons for the differences in metal selectivity. Progressive Sn/Sb substitution resulted in an increase in the lattice parameter (a), and in the K_d values of

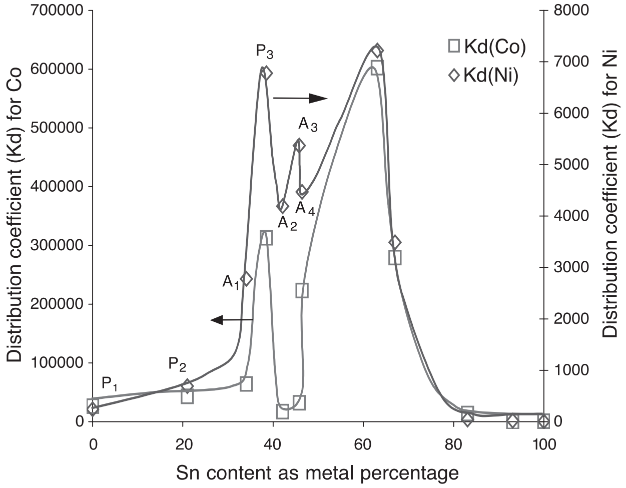

Figure 2. Distribution coefficients (mL g^{-1}) for ^{57}Co (□) and ^{63}Ni (◇) in 0.1-M HNO_3 solution as a function of Sn content (in metal percentage).

cubic, pyrochlore-structured materials (Table 3, Figure 3). Higher K_d values were associated with easier access of exchanging ions to inner ion exchange sites (inside tunnels and cavities of pyrochlore materials) of the materials. The K_d values rose sharply with the increase in lattice parameter from 10.34 Å to 10.39 Å [from exchanger SnSb(P_2) to (P_3)]: the K_d values were over 9.6 times as high for nickel and 7.7 for cobalt. This level of increase was almost the same as observed in earlier experiments (see Figure 2), where the increase in K_d values from SnSb(P_2) to (P_3) was 9.8 times for nickel and 7.3 for cobalt. The exchanger SnSb(A_1), which lies in between the two mentioned exchangers [SnSb(P_2) and (P_3), see Figure 2], exhibited 4.0 times as great in nickel uptake but only 1.5 as great in cobalt uptake in K_d values from SnSb(P_2) to (A_1). The smaller increase in K_d values of cobalt compared to nickel may be associated with the amorphous structure

Table 2. Radii of hydrated ions and the molar Gibbs energies of hydration of ions.

Ion	Radii of hydrated ion (Å)	Δ_{hyd} G/kJ mol^{-1}
Ca^{2+}	2.71	−1505
Ni^{2+}	3.02	−1980
Co^{2+}	2.95	−1915

Table 3. Most intensive XRD reflections and mean lattice constants (a) of the pyrochlore-structured materials, and K_d values (ml g^{-1}) in 0.1-M HNO_3 solution for ^{57}Co and ^{63}Ni.

Sample	*hkl* (111), *d*/Å	*hkl* (311), d/Å	*hkl* (222), *d*/Å	Mean lattice constant (a)	K_d value (Co/Ni)
SnSb(P_1)	5.893	3.110	2.972	10.272	19,300/850
SnSb(P_2)	5.968	3.122	2.983	10.342	20,800/1200
SnSb(P_3)	6.015	3.131	2.991	10.386	160,400/11,500
SnSb(P_2) Na-form	5.974	3.115	2.985	10.335	—
SnSb(P_2) Ca-form	5.928	3.097	2.964	10.269	—

of SnSb(A_1). In the case the of the pyrochlore-structured materials, most of the cobalt uptake is believed to take place in tunnels and cavities,[10,15] while most of the nickel uptake is proposed to take place at the outer surfaces of the material owing to steric hindrance of the larger hydratation shell of nickel ion (see Table 2). Since the SnSb(A_1) exchanger probably consists of small pyrochlore crystals with low ordering, producing an amorphous XRD pattern, the pore structure of the material must be disordered, restricting entrance and exchange of the ions deep inside the

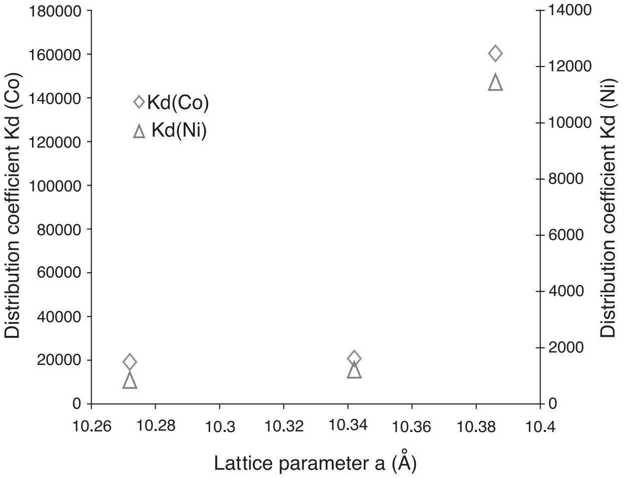

Figure 3. Distribution coefficients (mL g^{-1}) of ^{57}Co and ^{63}Ni in 0.1-M HNO_3 solution as a function of lattice parameter of tin antimonates with pyrochlore structure.

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material. A similar difference in K_d values between nickel and cobalt can be seen for the exchangers SnSb(A_2), (A_3), and (A_4). Here, the SnSb(A_4) has traces of pyrochlore structure in the XRD pattern, i.e., more ordering of crystals, producing more tunneled structure and higher cobalt uptake.

Changes in uptakes of cobalt and nickel as a function of Sn content followed an almost identical pattern for the exchangers with rutile structure [(SnSb(R_1) to (R_4), see Figure 2]. This similarity in metal uptake is due to the particle hydrate structure of the rutile-exchangers,[15] where the ion exchange is expected to take place at the material surfaces. This reduces the differences in selectivity originating from the size of the exchanging cation and emphasizes the effect of electrostatic attraction. The Pauling electronegativity is 1.8 for both metals.

Influence of pH and Calcium on K_d Values

The pH dependence of nickel uptake is of importance in planning the decontamination of floor drain waters since the pH can easily be altered. Due to promising ion exchange performance in acidic condition,[9] original and calcium-form SnSb(P_2) exchangers were tested for their Ni uptake in solutions of different pH, but, unexpectedly, marked differences were not seen between the two exchanger forms. The uptake of nickel was only slightly lower in the Ca-form SnSb(P_2) exchanger than in the original form

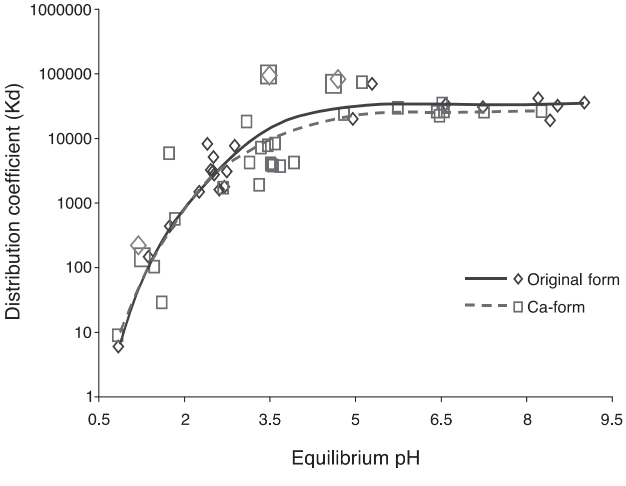

Figure 4. Distribution coefficients of original (◇) and calcium form (□) SnSb(P_2) for ^{63}Ni in 0.01-M Ca(NO_3)$_2$ solution (mL g^{-1}), larger symbols represent K_d values of ^{57}Co.

$SnSb(P_2)$ exchanger (Figure 4). This may have to do with the batch conditions, where the solution contained calcium ions [0.01M $Ca(NO_3)_2$]. In these conditions, as much as half of the original form exchanger could have converted to calcium form. Such change is known to decrease the lattice parameter of materials with pyrochlore structure, and decrease in the lattice parameter of calcium form $SnSb(P_2)$ was indeed observed (see Table 3). Possibly, the outer edge crystals of the original $SnSb(P_2)$ were changed into calcium form, decreasing the size of the opening of the tunnel structure and resulting in lower K_d values for nickel. The same trend was observed with cobalt uptake, which was tested at three different pH values, where the K_d values for the two $SnSb(P_2)$ forms differed only slightly.

Column Experiments

The column experiment demonstrated that $SnSb(A_3)$ was well suited for dynamic column use; both metals (^{57}Co and ^{63}Ni) were efficiently removed from the solution (Figure 5). Clear steps in breakthrough (BT) levels, for nickel particularly, were observed and associated with proton concentration of the eluent. Preconditioning of the exchanger with the inactive simulate most likely changed the exchanger partly into calcium form (the selectivity series for tin antimonates has been reported as

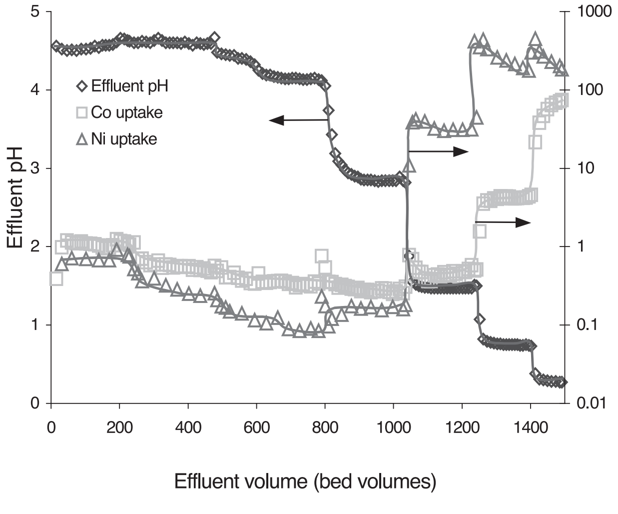

Figure 5. Breakthrough curves of ^{57}Co (□) and ^{63}Ni (△) for $SnSb(P_2)$ in neutral bond water simulate.

$Co^{2+} > Ni^{2+} > Sr^{2+} > Ca^{2+} > Mg^{2+} > Cs^{+} > K^{+} > Na^{+}$).[17] With increased proton concentration of the eluent, calcium would elute from the exchanger, leaving more of the ion exchange sites available for cobalt and nickel exchange resulting in their increased uptake. As can be seen in Figure 5, decreased BT levels were observed for both metals at the beginning of the experiment, and the decreases continued till the effluent pH fell below 4.1 in the case of nickel and below 2.8 in the case of cobalt. Differences in the elution of nickel and cobalt are clearly evident in Figure 5, where nickel breakthrough occurs at higher effluent pH. The preference in the elution can be explained by the enthalpy of the ion exchange reaction on crystalline antimony pentoxide (ΔH° of $M^{2+} \rightarrow H^{+}$ is 9.0, 79.5, and 159.1 kJ eq^{-1} for Ca, Ni, and Co, respectively).[10] The marked differences in the elution of the metals means that nickel and cobalt might themselves be separated through variation in acid concentration of the eluent.

The clear deviation of some of the experiment points from the general trend is due to stopping the pump to adjust the eluent pH and to the slow recovery of the column.

The preference of nickel and cobalt for different ion exchange sites (at the surface and inside tunnels) can also be seen in dynamic column experiment. The uptake of nickel was greater than uptake of cobalt in the dynamic experiment (lower breakthrough level in Figure 5), although the K_d values from batch experiments indicated otherwise (see Table 3, Figure 2). This preference of uptake for different sites most likely originates from kinetic limitations of the dynamic experiment, which favors ion exchange at the surface rather than inside the tunnels of the material, i.e., more favorable for nickel uptake than cobalt uptake.

CONCLUSION

Tin antimonates showed good cobalt and nickel uptake properties both in batch and in dynamic column experiments, and the application of tin antimonates for the decontamination of floor drain and neutral bond waters is concluded to be a promising possibility.

The effects of increasing Sn content on distribution coefficients for cobalt and nickel were almost identical for materials with rutile structure and mostly similar for materials with pyrochlore structure. The findings suggest that ordering of the crystals has strong influence on the K_d values of tin antimonates with pyrochlore structure. Cobalt uptake was diminished in the low crystallinity exchangers with distorted pore structure, since the majority of the ion exchange sites are located inside the tunnels and cavities of materials with pyrochlore structure. Most of the nickel uptake is proposed to take place at surfaces of the materials, so crystal ordering has less effect on nickel selectivity.

The kinetics of the ion exchange reaction had a strong influence on nickel and cobalt uptake in the dynamic column experiment. High selectivity of pyrochlore-structured materials in the batch experiments for cobalt was attributed with ion exchange in tunnels inside the material. The higher breakthrough level of cobalt than of nickel in the column experiment can, thus, be explained by restricted flow of ions inside the tunnels of the exchanger material, resulting in a lower ion exchange rate for cobalt than for nickel, the uptake of which occurs mostly on the surface of the exchanger.

The selectivity of pyrochlore $SnSb(P_2)$ most likely changed during the batch experiment due to a change in lattice constant caused by the experimental conditions. The original form $SnSb(P_2)$ exchanger may have been partially converted into calcium form by the experimental solution because only small differences in K_d values between the calcium and the original form $SnSb(P_2)$ exchanger were found. Support for this hypothesis could be sought through use of more powerful structure characterization techniques, such as extended x-ray absorption fine structure (EXAFS).

Data on the uptake of cobalt on tin antimonates can be used in some degree for the prediction of nickel uptake, even though the uptake level of nickel is much lower. The structure and crystallinity of the tin antimonate also need to be known because of the strong effect of these factors on the metal uptake properties.

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