Sorption of radiocobalt and its EDTA complex on titanium antimonates

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ABSTRACT

The sorption properties of two synthesized titanium antimonate materials were tested in simulated nuclear power plant decontamination solutions. The aim was the removal of radiocobalt in the presence of complexing agents such as EDTA and oxalate. The first titanium antimonate material had a mixture of pyrochlore and rutile structures and it showed good tolerance for the complexing agents. 91% of cobalt was removed in the presence of EDTA, and oxalic acid had only a minor effect on the sorption. The other material, which had a mopungite structure, tolerated EDTA well (97% removal of cobalt) but its sorption properties for cobalt were restricted to a specific amount of oxalic acid. The sorption efficiency of cobalt increased for both materials when the cobalt concentration was less than 1 μM, indicating that the synthesized materials are efficient sorbents, especially for trace amounts of radiocobalt.

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1. Introduction

Nuclear power plants (NPP) produce large volumes of liquid radioactive waste. Solidification and volume reduction of these liquid wastes is required to decrease aquatic radioactivity emissions and to obtain suitable waste forms for final disposal. Recently, selective inorganic ion exchange and sorbent materials have gained ground in the treatment of liquid waste, replacing older technologies based on evaporation and organic ion exchange resins [1–3]. Good radiation stability of inorganic materials is an obvious advantage over the organic resins but the major advantage comes from the high selectivity obtainable with the inorganic materials. Liquid wastes from NPPs contain high amounts of inactive metal ions such as K$^+$, Na$^+$ and Ca$^+$ and quite often only trace concentrations of radionuclides. Thus a high selectivity is required to efficiently remove the radionuclides and to obtain a minimal amount of waste (i.e. spent sorbent) for final disposal.

Due to its relatively long half-life (5.2 a) and high gamma decay energy, ^{60}Co is one of the most problematic waste nuclides in NPP liquid waste streams. Many of these waste streams contain complexing agents such as EDTA and oxalate originating from cleaning solutions used to remove the radioactive metals, such as ^{60}Co, from contaminated structures and equipment of NPPs. The 1:1 metal complexes of EDTA are stable over a broad pH range, which leads to a marked increase in the mobilization of toxic metal ions [4,5]. Removing metals from the EDTA complexes is difficult due to their high stability constants. Cation exchange resins have normally been used for the removal of ^{60}Co, and in addition recent studies have proven the efficiency of highly selective inorganic ion exchangers for its removal [2,3,6,7]. However, as soon as ^{60}Co is complexed, for example with EDTA, both methods turn out to be quite inefficient.

The aim of this study was to find an efficient titanium antimonate material for the removal of ^{60}Co and its EDTA complex from solution. Oxalic acid and NaNO$_3$ were added to some of the solutions in order to simulate the liquid waste from NPPs. The sorption ability of the titanium antimonate materials was also examined over a wide pH range of the solution. Two different titanium antimonate materials, TiSbA and TiSbB, were tested in this study. The final synthesis pH values for the TiSb materials were 2 and 11, respectively. According to the X-ray diffraction patterns TiSbA was a mixture of pyrochlore and rutile TiO$_2$ structures while the TiSbB structure was similar to that of mopungite. The energy-dispersive X-ray spectra revealed that titanium was present in both of the TiSb materials.

Titanium antimonate materials have been developed previously for e.g., the removal of ionic ^{60}Co from acidic solutions [2] and from acidic solutions in the presence of calcium [3]. An extensive study of titanium antimonates has been made by Abe et al. [8]. In addition, studies [9,10] have shown that the acidity of the titanium antimonate material can be increased by enhancing the amount of pentavalent antimony in the material. This improves the sorption properties of the material at low pH values. The sorption properties are also affected by the synthesis conditions of hydrous metal oxides since the composition and crystallinity are dependent on the synthesis.

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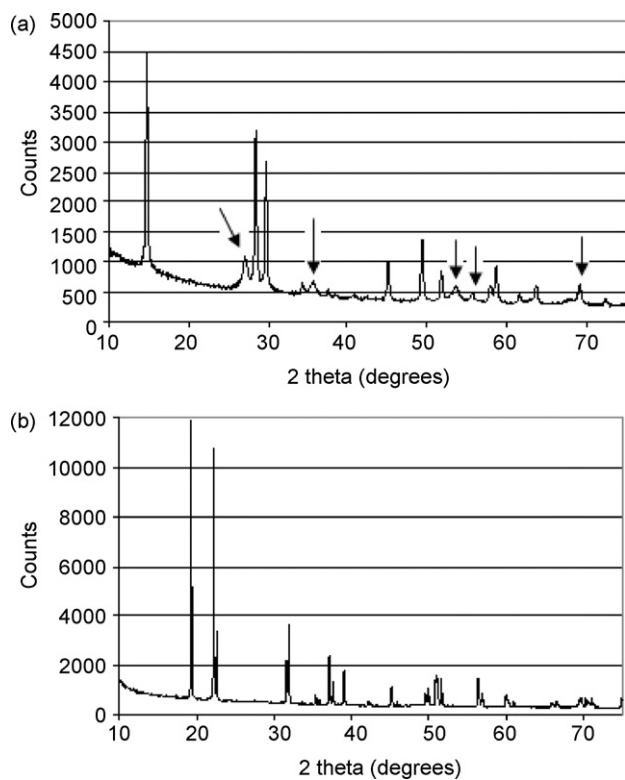*E-mail address:* leena.k.malinen@helsinki.fi (L.K. Malinen).

Fig. 1. Powder XRD pattern of (a) TiSbA, arrows are showing the peaks which belong to rutile TiO_2 structure and (b) TiSbB.

2. Experimental

2.1. Reagents

Titanium tetrachloride 99% (Riedel-de Haën), antimony pentachloride 98% (Fluka) and analytical grade $CoCl_2 \cdot H_2O$ (Riedel-de Haën), Na_2EDTA (Merck), $NaNO_3$ (Merck) and oxalic acid (Merck) were used without further purification. The radioactive tracer, ^{57}Co, used in the sorption experiments was obtained from Eckert & Ziegler Isotope products. Solutions were prepared from water purified by a Milli-Q water purification system (18 MΩ cm).

2.2. Synthesis of materials

Titanium antimonates were synthesized by modification [2] of a known method [8,11]. First, a mixture of titanium tetrachloride and antimony pentachloride was prehydrolyzed in 4 ml of distilled water with the initial molar ratio Ti/Sb being 1.5 for TiSbA sorbent material. Then the amount of water was increased to 500 ml. The white precipitate was allowed to stand in the mother liquor at 60 °C overnight, centrifuged and washed with distilled water until the pH of the wash solution was about 2. Then the precipitate was dried at 110 °C. The sorbent material TiSbB was made by prehydrolyzing titanium tetrachloride and antimony pentachloride (Ti/Sb, 1.5) in 10 ml of distilled water and adding 240 ml of 6 M NaOH. The precipitate was allowed to stand in the mother liquor overnight at room temperature, and then it was centrifuged and washed with distilled water until the pH of the wash solution was about 11.5. The precipitate was dried at 110 °C.

2.3. Characterization

Crystal structures of TiSbA and TiSbB (Fig. 1) were determined from X-ray diffraction (XRD) patterns recorded with a Panalytical

Table 1
The elements identified in the titanium antimonates (TiSbA and TiSbB) and an estimation of their content (%) from EDX spectra.

Material	Element	%
TiSbA	O	73
	Cl	3
	Ti	12
	Sb	12
TiSbB	O	71
	Na	10
	Ti	5
	Sb	14

X'pert Pro MPD X-ray diffraction system. The generator was operated at 40 kV and 40 mA. An X'pert High Score Plus software was used for the identification of TiSb structures. TiSbA was found to be a mixture of pyrochlore (cubic, space group Fd_3m) and rutile TiO_2 (tetragonal, space group $P4_2/mnm$) structures while the TiSbB structure resembled that of mopungite (tetragonal, space group $P4_2/m$).

An Oxford INCA 350 microanalysis system connected to a Hitachi S-4800 field emission scanning electron microscope (FESEM) was used to measure the EDX (energy-dispersive X-ray spectroscopy) spectra in order to identify the elements present in TiSbA and TiSbB after synthesis (Table 1). Titanium was present in both of the materials even though its structural phase could not be seen in the XRD pattern of TiSbB (Fig. 1). Since EDX measurements were performed from a powder-like sample, the values obtained should be considered estimates. The specific surface area and porosity of TiSbA and TiSbB was measured with a Quantachrome Autosorb-1 surface area and pore size analyzer (Table 2).

2.4. Experiments

The sorption properties of the synthesized titanium antimonates were studied in room temperature using ^{57}Co-traced (^{57}Co concentration $<3 \times 10^{-14}$ M) test solutions of 0.01 M $NaNO_3$ or Milli-Q water with Co^{2+} and EDTA concentrations (equimolar) varying between 0.2 μM and 200 μM. In addition, test series were made where $NaNO_3$ concentration varied between 0.1 mM and 100 mM or oxalic acid concentration varied between 1 mM and 100 mM while Co–EDTA concentration was 10 μM. Constant rotary mixing (50 rpm) was used to equilibrate 20 mg of synthesized powder-like sorbent with 10 ml of test solution for 1 or 4 days. Longer mixing times (4 days) were used in the experiments where acid or base was added to the mixture of sorbent material and solution. The objective of the longer mixing time was to assure that equilibrium between solid and solution was achieved and this probably increased the distribution coefficients to some extend. Nonetheless, when the results are compared, the effect of the mixing time seems minimal. Test solution pH was adjusted with varying amounts of HNO_3 or NaOH as necessary. The sorbent was removed from the solution by centrifugation (Sepatech Megafuge 1.0, Heraeus) for 10 min at 4000 rpm (3000 G). The solution was filtered with a 0.2 μm Acrodisc filter (PALL Life Sciences) and its equilibrium pH was measured with an Orion 3 star pH meter with a BNC waterproof electrode. Aliquots (5 ml) of solution before and after the experiments were counted for ^{57}Co with an automatic gamma

Table 2
Specific surface area and total pore volume of the titanium antimonates (TiSbA and TiSbB).

Sorption material	Specific surface area ($m^2 g^{-1}$)	Total pore volume ($cm^3 g^{-1}$)
TiSbA	74	0.200
TiSbB	1	0.011

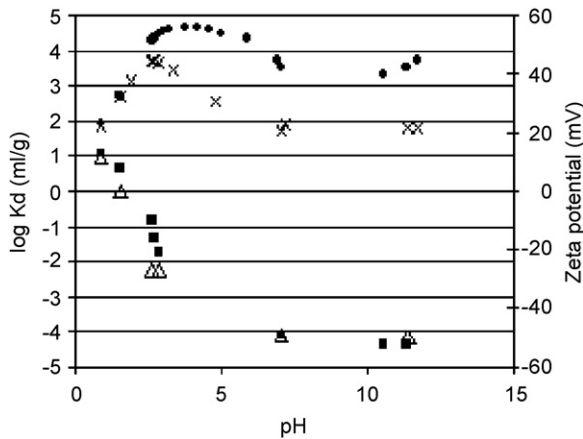**Fig. 2.** ^{57}Co distribution coefficient (K_D/ml g^{-1}) in 0.01 M NaNO$_3$ solution containing either 10 μM Co^{2+} (●) or 10 μM Co–EDTA (×) and estimates of zeta potential values of the former (■) and the latter (△) as a function of pH for TiSbA.

Fig. 3. ^{57}Co distribution coefficient (K_D/ml g^{-1}) in 0.01 M NaNO$_3$ solution containing either 10 μM Co^{2+} (●) or 10 μM Co–EDTA (×) and estimates of zeta potential values of the former (■) and the latter (△) as a function of pH for TiSbB.

counter (Wallac 1480 Wizard^{TM} 3). The zeta potential of the solution with suspended sorbent materials was measured with a Malvern Zetasizer Nano ZS which was designed to measure 5 nm to 10 μm particles. Therefore, the sample solution was filtered with a 10 μm Acrodisc PSF filter (PALL Life Sciences) before zeta potential measurements.

The distribution coefficient (K_D) illustrates the distribution of solute between the solution and the solid material at equilibrium, i.e.

$$K_D = \frac{C_s}{C_l} \qquad (1)$$

where C_s (mol/kg) and C_l (mol/l) are the solute concentrations in the solid and solution, respectively. Thus K_D can be used to estimate the processing capacity (l/kg) of the sorbent material under the prevailing conditions. It was calculated for ^{57}Co using Eq. (2):

$$K_D = \frac{(A_0 - A_{eq})}{A_{eq}} \times \frac{V}{m} \qquad (2)$$

where A_0 is the initial count rate of ^{57}Co in the solution, A_{eq} is the count rate of ^{57}Co at equilibrium and V/m is the solution volume to sorbent mass ratio (batch factor, ml g^{-1}). The errors of K_D values represent the difference between the replicate samples.

3. Results and discussion

3.1. Sorption experiments in 0.01 M NaNO$_3$ solution

3.1.1. Sorption of ionic cobalt

The TiSb materials synthesized showed divergence in their behavior in the sorption experiments. The sorption maximum of ionic cobalt (10 μM) from the 0.01 M NaNO$_3$ solution for TiSbA was located at pH 4 (Fig. 2) and for TiSbB at pH 7 (Fig. 3). The corresponding K_D values were 47,000 ± 500 ml/g (98.9% removal of cobalt) and 38,000 ± 700 ml/g (98.7% removal of cobalt), respectively. There was also a peak at pH 2 for TiSbB but its K_D value (17,400 ± 1000 ml/g) was noticeably smaller than the K_D maximum. Over 95% removal of cobalt has also been achieved by TiSb materials synthesized by Tsuji et al. [12] and Karhu et al. [2] doing sorption experiments in nitric acid solutions.

Based on earlier observations [9,10] that increasing the Sb(V) content (decreasing the Ti content) in the titanium antimonates increases metal ion uptake in acidic solutions, it could have been expected that TiSbB would have performed at the same level as or even better than TiSbA. As this was not the case, it was concluded that synthesis pH and the specific surface area (74 m^2 g^{-1}
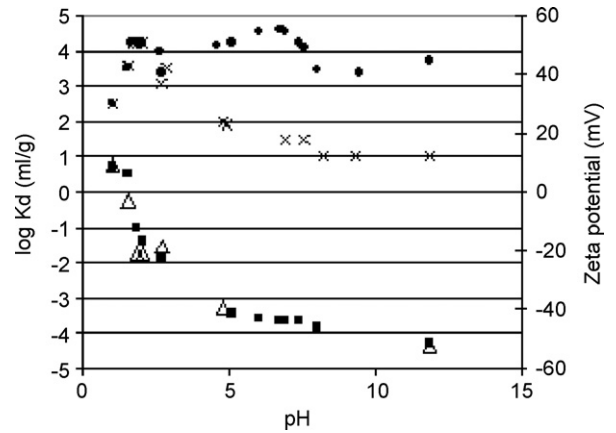
and 1 m^2 g^{-1} for TiSbA and TiSbB, respectively) have more effect on the sorption properties than the pentavalent antimony content. The results also showed that the synthesis pH affected the pH range of the maximum sorption area of the sorbent. Acidic synthesis conditions led to better sorption properties at low pH values, whereas basic synthesis conditions led to a sorption maximum at higher pH.

The equilibrium chemical speciation estimates for cobalt in solution over pH range of the experiments were calculated using the program Visual Minteq version 2.51 [13]. The speciation distribution for ionic cobalt shows that it remains in a free ionic form up to pH 8, above which the hydrolyzed species Co(OH)$^+$, Co(OH)$_2$ (aq) and Co(OH)$_2$ (s) start to form. At this pH, the sorption properties for both TiSb materials deteriorated significantly.

3.1.2. Sorption of Co–EDTA

When cobalt was complexed with EDTA, a drop in the K_D values could be seen compared to those obtained for ionic cobalt (Figs. 2 and 3). For TiSbA the coefficient maximum for ^{57}Co was at pH 2.6, where the K_D value was 5400 ± 400 ml/g (91% removal of cobalt). Based on calculations [13], at pH 2.6 the following Co–EDTA species are present in the solution (the percentage value of the total): CoH$_2$EDTA (4%), Co^{2+} (10%), CoEDTA^{2-} (29%) and CoHEDTA$^-$ (57%) (Fig. 4). As the removal of total cobalt was 91%, TiSbA either took up the various Co–EDTA species or removed Co^{2+} ions from the EDTA complexes (decomplexation). The latter possibility was estimated from a rough calculation of cobalt distribution between
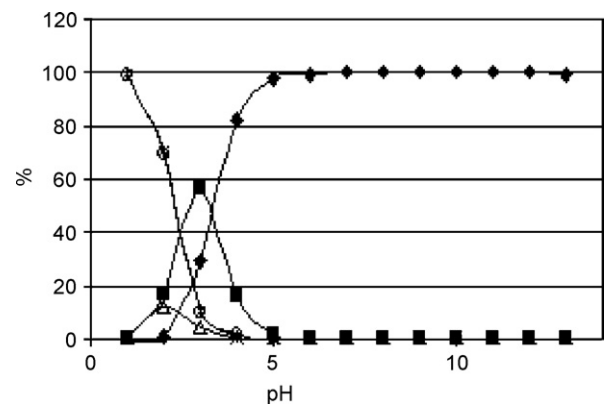

Fig. 4. Equilibrium estimates in solution over the tested pH range for Co–EDTA (10 μM) complex, performed using the chemical speciation program Visual Minteq, version 2.51 [13]. CoEDTA^{2-} (♦), CoHEDTA$^-$ (■), CoH$_2$EDTA (△), Co^{2+} (□) and H$_n$EDTA (*).

the EDTA complex and TiSbA, ie.

$$\frac{[\text{CoEDTA}^{2-}]}{C_S} = \frac{K_f}{K_D}[\text{EDTA}^{4-}] \qquad (3)$$

where $[\text{CoEDTA}^{2-}]$ is the concentration of the CoEDTA^{2-} complex, C_S is the concentration of sorbed Co^{2+} on TiSbA (see Eq. (1)), K_f is the formation constant of CoEDTA^{2-}, K_D is the distribution coefficient of Co^{2+} sorption on TiSbA and $[\text{EDTA}^{4-}]$ is the concentration of EDTA^{4-}. At pH 2.6 the concentration of EDTA^{4-} is in order 2×10^{-18} M, based on calculations from Visual Minteq [13], while K_D was 5400 ± 400 ml/g. Formation constant K_f is 2.8×10^{16} [14]. Inserting these values in Eq. (3) shows that the ratio of Eq. (3) is approximately 1/100,000 which indicates that practically all cobalt is sorbed as Co^{2+} by TiSbA at pH 2.6.

TiSbB has its sorption maximum for Co–EDTA at pH 2, where the K_D value was $17,900 \pm 1200$ ml/g (97% removal of cobalt), which is the same as for ionic cobalt solution. The species present in the solution at that pH are: CoH_2EDTA (12%), Co^{2+} (69%), CoEDTA^{2-} (1%) and CoHEDTA^{-} (17%) [13]. The estimation about the possible decomplexation of Co–EDTA was calculated for TiSbB using Eq. (3). At pH 2 the concentration of EDTA^{4-} is in order 7×10^{-20} [13], while K_D was 17900 ± 1200 ml/g. Inserting these values in Eq. (3) shows that the ratio of Eq. (3) is approximately 1/9,000,000 which clearly indicates that all cobalt is sorbed as Co^{2+} by TiSbB at pH 2.

3.1.3. Zeta potential measurements

Even though the zeta potential values can not be accurately calculated from electrophoretic mobility when the material is irregularly shaped, porous and has a large size distribution (<10 μm), a rough estimation of the zeta potential values was made. Zeta potential is a value for the electrochemical double layer (mV) which exists at phase boundaries between solids and electrolyte solutions. It was altered by changing the solution pH with either HNO_3 or NaOH. EDTA in the solution altered the zeta potential compared to the solutions including only cobalt even though there was a great surplus of Na^+- and $\text{NO}_3{}^-$-ions (0.01 M) in the solution. This was clearly seen at low pH values (Figs. 2 and 3). The zeta potential values in the presence of EDTA were more negative than the values when only ionic cobalt was present. This might have been a result of the sorption of the anionic EDTA species on the materials, as accumulation of such species would have further lowered the potential. Zeta potential values were very similar for both of the sorption materials and it was seen that the sorption maxima were located in areas where the zeta potential was close to −30 mV (usually particles are considered to be unstable when the zeta potential is between −30 mV and +30 mV).

3.2. The effect of NaNO$_3$ concentration on the sorption of Co–EDTA

The effect of the NaNO_3 concentration on the sorption properties of the TiSb materials was studied in the presence of Co–EDTA. Sorption tests were made without any pH alteration, allowing the mixture of the solution and sorbent to settle to its natural equilibrium. The K_D values were below 4600 ml/g and 1000 ml/g for TiSbA and TiSbB respectively, throughout the experiment. The equilibrium pH for TiSbA during the test series was approximately 3 and for TiSbB it was approximately 8, which explains the low K_D values, especially for TiSbB. When the concentration of NaNO_3 increased in the solution, the distribution coefficient of TiSbA decreased (Fig. 5). At pH 3 there is still some free ionic cobalt present in the solution and the decrease in K_D is probably due to the competing effect of the increased Na^+ concentration. An opposite effect can be seen with TiSbB (Fig. 5), for which the distribution coefficient slightly increases when the concentration of NaNO_3 increases. At pH 8, there
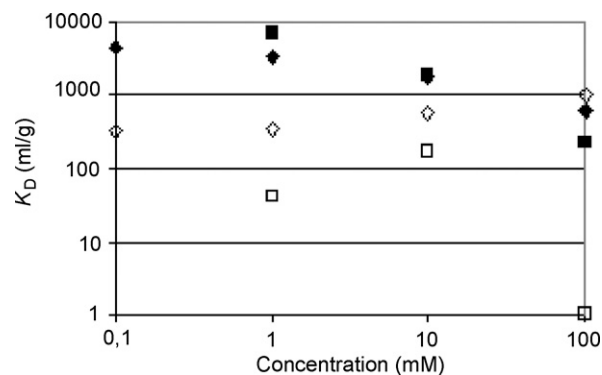

Fig. 5. ^{57}Co distribution coefficient (K_D/ml g^{-1}) in various NaNO_3 solutions with 10 μM Co–EDTA concentration for TiSbA (♦) and TiSbB (◊) or in various oxalic acid solutions with 10 μM Co–EDTA concentration for TiSbA (■) and TiSbB (□).

is no ionic Co^{2+} in solution and uptake might be due to CoEDTA^{2-} sorption. The increasing Na^+ concentration is likely to decrease the negative surface charge and thus facilitate the sorption of the negatively charged species.

3.3. The effect of oxalic acid concentration on the sorption of Co–EDTA

The effect of oxalic acid on the sorption of Co–EDTA without any additional pH changes was also studied. The initial pH values for the solutions differed due to the oxalic acid concentration. Increasing the concentration of oxalic acid had a minor effect on the equilibrium pH of TiSbA, since TiSbA is an acidic material. The pH value decreased from 2.4 to 1.4 when the concentration of oxalic acid increased from 1 mM to 100 mM. When the oxalic acid concentration was 10 mM (equilibrium pH 2), the K_D value for TiSbA was 1900 ± 100 ml/g (79% sorption of cobalt) (Fig. 5). This was about the same value as obtained for Co–EDTA in NaNO_3 solution (K_D 1400 ± 50 ml/g at pH 1.9). For TiSbB, the pH values changed from 6.4 to 1.5 when the oxalic acid concentration increased from 1 mM to 100 mM. The maximum K_D for TiSbB could be seen at an oxalic acid concentration of 10 mM, where the pH was 2.7 (Fig. 5). The K_D value at this point was 170 ± 10 ml/g (25% sorption of cobalt) which was noticeably smaller than the K_D value (3600 ± 500 ml/g) obtained for Co–EDTA in NaNO_3 solution at the corresponding pH without oxalic acid. The dramatic effect of oxalic acid on the sorption ability of TiSbB could be explained by the formation of Co–oxalate complex which prevented the removal of cobalt.
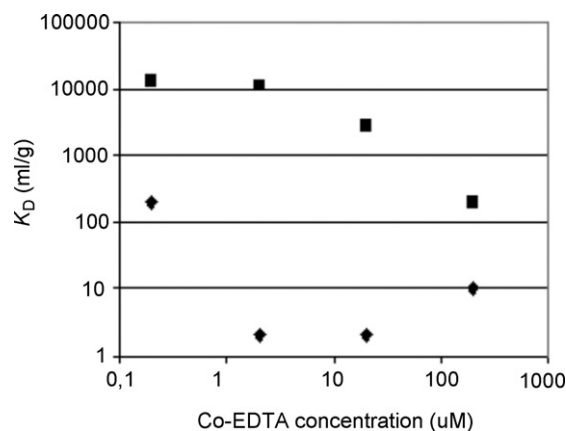

Fig. 6. ^{57}Co distribution coefficient (K_D/ml g^{-1}) in various Co–EDTA solutions for TiSbA (■) and TiSbB (♦) using equimolar Co–EDTA concentrations.

3.4. The effect of Co–EDTA concentration on the sorption of Co–EDTA

The equilibrium pH for TiSbA was approximately 3 and for TiSbB approximately 10, when the effect of Co–EDTA concentration on the sorption properties was tested without NaNO$_3$ background and pH alterations (Fig. 6). When the concentration of CoEDTA was 20 μM, the distribution coefficient was 2900 $\pm$ 15 ml/g for TiSbA and less than 10 ml/g for TiSbB. It can be seen that increasing the concentration of CoEDTA to 200 μM decreases the sorption properties of TiSbA but has a favorable effect on the sorption properties of TiSbB. In addition, the sorption efficiency of both materials can be seen to improve with low concentrations (<1 μM) of Co–EDTA, indicating that the materials are efficient sorbents for the trace amounts of cobalt.

4. Conclusions

Promising results were obtained with the two synthesized titanium antimonate (TiSb) materials for cobalt removal from various solutions simulating nuclear decontamination solutions. Sorption levels of cobalt were more than 98% in the presence of only ionic Co^{2+}, and over 90% in the presence of Co–EDTA complex for both materials. Indications were seen that TiSb materials were able to extract cobalt from the EDTA complexes.

It was shown that an increase in NaNO$_3$ concentration lowered the sorption ability of the material with the mixture of pyrochlore and rutile structures (TiSbA) but increased the distribution coefficient for the material with a mopungite structure (TiSbB). The equilibrium pH for TiSbA was about 3 throughout the experiment and it is probable that Na$^+$-ions compete with Co^{2+}-ions in acidic conditions. On the other hand, Na$^+$-ions might decrease the negative surface charge at pH 8, which was the equilibrium pH for TiSbB throughout the experiment. The decrease in the negative surface charge probably led to better sorption of the negatively charged species of Co–EDTA, since at pH 8 practically all cobalt is bound by the EDTA complexes. At low Co–EDTA concentrations (<1 μM), both materials showed efficient cobalt uptake. This indicated the feasibility of using the synthesized materials for the sorption of trace amounts of cobalt in decontamination solutions. Since the sorption of trace amounts of radionuclides is difficult, this result was very promising, and the experiments should be continued with very low concentrations (<1 μM) of cobalt and other common radionuclides in the decontamination solutions.

A common decontamination chemical, oxalic acid, had only minor effects on the sorption properties of TiSbA but strongly decreased the distribution coefficient of TiSbB. It was assumed that the formation of Co–oxalate complex prevented the sorption of cobalt for TiSbB. The excellent tolerance for oxalic acid of TiSbA was explained by the acidic (pH 2) synthesis conditions of the material.

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