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Transport and leaching of technetium and uranium from spent UO_2 fuel in compacted bentonite clay

H. Ramebäck[a,*], Y. Albinsson[a], M. Skålberg[a], U.B. Eklund[b], L. Kjellberg[b], L. Werme[c]

[a] *Department of Nuclear Chemistry, Chalmers University of Technology, SE-412 96 Göteborg, Sweden*
[b] *Studsvik Nuclear AB, Hot Cell Laboratory, SE-611 82 Nyköping, Sweden*
[c] *Swedish Nuclear Fuel and Waste Management Co., SKB, P.O. Box 5864, SE-102 40 Stockholm, Sweden*

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Abstract

The transport properties of Tc and U in compacted bentonite clay and the leaching behaviour of these elements from spent nuclear fuel in the same system were investigated. Pieces of spent UO_2 fuel were embedded in bentonite clay ($\rho_d = 2100$ kg/m^3). A low saline synthetic groundwater was used as the aqueous phase. After certain experimental times, the bentonite clay was cut into 0.1 mm thick slices, which were analysed for their content of Tc and U. Measurements were made using inductively coupled plasma mass spectrometry. Tc analysis comprised chemical separation. The analysis of U was done by means of detecting ^{236}U, since the natural content of U in bentonite clay made it impossible to distinguish between U originating from the fuel and the clay. The influence of different additives mixed into the clay was studied. The results showed an influence on both transport and leaching behaviour when metallic Fe was mixed into the clay. This indicates that Tc and U are reduced to their lower oxidation states as a result of this additive. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

The behaviour of spent fuel in the near field of a geological repository is important for the safety, since the fuel itself is the first barrier to the release of radionuclides. The constituents of the irradiated fuel can be present at different locations: fuel–clad gap, grain boundaries and the fuel matrix [1]. Examples of radionuclides found at the fuel–clad gap are ^{129}I and ^{137}Cs. ^{99}Tc is to a small extent segregated to the grain boundaries, frequently in the form of metallic inclusions, together with molybdenum, ruthenium, rhodium and palladium [2]. These metallic inclusions (often referred to as ε-phase particles) are also found within the UO_2 grains [3]. The actinides and the lanthanides are expected to be in solid solution with the fuel matrix [2]. As a consequence of this, different mechanisms will govern the release rates when the fuel is in contact with water. The release of the radionuclides present at the fuel–clad gap is a fast process that occurs within a few days after first contact with water. The amount released has been shown to be comparable to the fission gas release during reactor operation [4]. If there is specific release rate from the grain boundaries, it is a somewhat slower process, although believed to be much faster than the release from the fuel matrix [5]. The release from the fuel matrix will require dissolution or alteration of the UO_2 itself [5]. It is often assumed that the Sr release rate is an indicator for the matrix dissolution, and following that assumption, the release rate would be about 0.01% of the inventory per year under oxidising conditions [6]. While the release rate of a radionuclide to a first approximation is governed by their accessibility for water, solubility limitations may also be important [6,7].

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[*] Corresponding author. Tel.: +46-31 772 2802; fax: +46-31 772 2931.

E-mail address: ramebeck@nc.chalmers.se (H. Ramebäck).

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Bentonite clay has been proposed as a buffer and backfill material for the waste storage. The clay will reduce the water inflow to the fuel canisters. The main transport mechanism for dissolved radionuclides will be by diffusion through the clay.

Technetium may, depending on the redox conditions, be present as different species. Under environmental conditions, technetium can exist in three different oxidation states: Tc(VII), Tc(IV), and Tc(0) [8,9]. TcO_4^- is the stable species in oxidising aqueous environments while, in reducing waters, TcO_2 is the solid phase and $TcO(OH)_2$ the soluble species [8]. Tc(VII) has both a higher solubility and a faster mobility than Tc(IV). Uranium will be present under reducing conditions as U(IV), which has a lower solubility than the species present at oxidising conditions (U(VI)) [10]. U(IV) is also expected to have a lower mobility than U(VI).

Apparent diffusivities (D_a) of technetium have been reported earlier [11,12]. Sato et al. [13] evaluated D_a for different clay densities. The apparent diffusivity of uranium has been reported by Albinsson et al. [11] and by Idemitsu et al. [14].

This work presents the results concerning the leaching of technetium and uranium from spent UO_2 fuel in contact with compacted bentonite clay. The transport of technetium is discussed, whereas the transport of uranium was presented earlier [15]. Results concerning the transport and leaching of ^{137}Cs [16] and ^{90}Sr [17] have also been reported earlier.

2. Experimental

A combined fuel leaching and radionuclide diffusion experiment was started in 1985 [18]. Spent UO_2 fuel (42 MWd/kg U) from rod No. 418-A6 of the boiling water reactor (BWR) Oskarshamn 1 was cut into 4.8 mm thick segments (diameter 12.25 mm including cladding) in a hot cell at Studsvik Nuclear Fuel Laboratory. These fuel specimens were placed between two cylinders of compacted bentonite clay ($\rho = 2100$ kg/m^3) in diffusion cells. The bentonite cylinders had a diameter of 25 mm and a length of 25 mm. The diffusion cells were made from stainless steel. There was a lid at each end in which a filter was placed to allow for water penetration through the bentonite clay to fuel specimen. After loading, each cell was placed in a sealed stainless steel vessel. These vessels contained a low saline synthetic groundwater [19] (see Table 1) which had been equilibrated with bentonite clay. The actual pore-water chemistry in compacted bentonite was largely unknown, or at least much debated at the time when the experiments were started. (Recently, more reliable data on the pore-water chemistry have been available [20].) A diffusion cell placed in a stainless steel vessel is shown in Fig. 1. After loading all cells into the vessels, they were transported using a transfer box out of the hot cell into the Fuel Laboratory's loading area where they were placed behind a lead shield at the ambient temperature in the area (about 25°C). Ten such experiments were started.

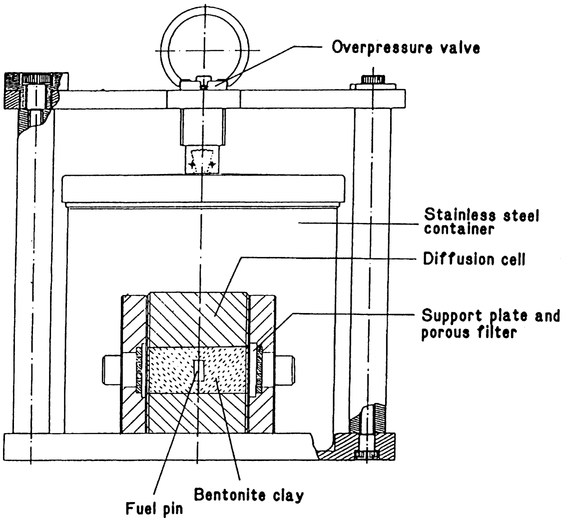

Fig. 1. Schematic view of the diffusion cell.

The clay used in the experiments was a commercial bentonite, Volclay MX-80. The composition and mineral content of the clay are given in Tables 2 and 3, respectively. The clay fraction (montmorillonite) of the bentonite was fine grained typically 2 μm or smaller, while the contaminating minerals as well as the additives (see below) had a size of 60 μm (or larger).

In six of the diffusion cells, additives were homogeneously mixed into the clay. These additives were metallic iron, metallic copper and the Fe(II) mineral vivianite ($Fe_3(PO_4)_2 \cdot 8H_2O$). Cells without additives in the bentonite clay were also set up.

An apparatus for automatic slicing of the clay after the experiment has been developed for use in a remotely controlled hot cell. After appropriate time (see Table 4)

Table 1
Composition (mg/dm^3) of the low saline groundwater [19] [a]

Species	HCO_3^-	SiO_2	SO_4^{2-}	Cl^-	Ca^{2+}	Mg^{2+}	K^+	Na^+
Concentration	123	12	9.6	70	18	4.3	3.9	65

[a] pH: 8.0–8.2, Ionic strength: 0.0085.

Table 2
Typical composition of bentonite clay (%) for constituents exceeding 1%

SiO_2	61–64
Al_2O_3	20–21
CaO	1.2–1.4
Fe_2O_3	3.8–3.9
MgO	2.4–3.7
Na_2O	2.1–2.4
LOI[a]	5.2–6.3

[a] Loss of ignition = carbonates, sulphides, sulphates and crystal bound water.

Table 3
Mineral composition of bentonite clay (%) [31]

Montmorillonite	75
Quartz	15
Mica	<1
Feldspars	5–8
Carbonates	1.4
Kaolinites	<1
Pyrite	0.3
Other minerals	2
Organic carbon	0.4

the diffusion cells were opened, the support plate and filters were removed and the cell was placed in a holder where a motor-driven plunger pressed out the clay through a punching cylinder with a sharp edge and a diameter of 20 mm. By this operation the surface layer of the clay which had been in contact with the metal surface of the diffusion cell was removed. The clay was pressed out step by step in lengths of 0.1 mm. Then an angled knife cut slices of the clay and transferred them into small plastic sample-storage boxes. The samples were transported to Chalmers University of Technology and stored under radiation shielding awaiting analysis. The activity of each sample was sufficiently low to allow handling in a specially shielded fume hood. These samples were analysed for their content of different radionuclides.

2.1. Analytical procedures

2.1.1. ^{99}Tc

A separation procedure was applied to the analysis of ^{99}Tc since ^{99}Ru (and ^{98}MoH) interferes with the detection using inductively coupled plasma mass spectrometry (ICP-MS). After leaching the clay samples with a mixture of 2 M sulphuric acid/10 mM sodium bromate at 60°C for 10 h, solid residues were removed by centrifugation. Technetium was extracted from the acid solution as pertechnetate with 50 mM Alamine-336 dissolved in chloroform. Technetium was thereafter back-extracted into 1 M nitric acid. The radiochemical procedure for ^{99}Tc is presented in detail elsewhere [21].

2.1.2. Uranium

The bentonite clay samples were treated with 1 M nitric acid at 65°C for 12 h in order to leach the actinides from the dried clay samples. Natural bentonite contains 10–15 ppm of uranium as impurities in the minerals. The aggressive leaching process used released sufficient amounts of this uranium to override the contribution from the fuel. As a consequence, ^{236}U had to be used to determine the amount of uranium originating from the fuel and the clay. ICP-MS was used for the measurements.

2.2. Evaluation of apparent diffusivity

Fick's second law of diffusion can described diffusion in a homogenous and porous medium. In one dimension, it can be written as

$$\frac{\partial c}{\partial t} = D_a \frac{\partial^2 c}{\partial x^2}, \tag{1}$$

Table 4
Short summary of the experiment of leaching and diffusion of radionuclides from spent fuel into compacted bentonite

Date (Year-month-day)	Time (days) from experiment start in 85-03-18	Diffusion medium	Amount of U (g)
85-06-27	101	Bentonite	2.7
85-10-01	197	Bentonite	3.0
86-04-08	386	Bentonite	2.8
		Bentonite + 0.5% Cu	3.2
		Bentonite + 0.5% Fe	3.0
		Bentonite + 1% Vivianite[a]	3.1
91-04-09	2213	Bentonite	3.2
		Bentonite + 0.5% Cu	3.2
		Bentonite + 0.5% Fe	3.2
		Bentonite + 1% Vivianite[a]	3.1

[a] $Fe_3(PO_4)_2 \cdot 8H_2O$.

where c is the concentration, t the diffusion time, x the space co-ordinate and D_a the apparent diffusivity, which in this case is assumed to be independent of the space co-ordinate. The apparent diffusivities were calculated from the least square fit of $\ln c$ versus x^2. The slope, k, will then be equal [22] to

$$k = -\frac{1}{4D_a t} \tag{2}$$

and the apparent diffusivity may be calculated. However, Eq. (2) is valid for the evaluation of apparent diffusivities when a constant amount of substance is diffusing. If instead the source behaves as a constant concentration source, the solution to the diffusion (Eq. (1)) is

$$\frac{c}{c_0} = \operatorname{erfc}\left[\frac{x}{2\sqrt{D_a t}}\right]. \tag{3}$$

2.3. Reporting units

Because of the difficulty in defining the surface area of highly cracked spent fuel, some experimental results are expressed as fractions of the initial inventories of each radionuclide. Also, since what was actually measured is the resultant of such processes as selective leaching, dissolution and precipitation and adsorption, the neutral term Fraction of Inventory on Clay and in Aqueous Phase (FICAP) is used here.

3. Results and discussion

3.1. Diffusion

3.1.1. Technetium

Fig. 2 shows the concentration profile for technetium after 3 months of diffusion when no additive was mixed into the clay. From this concentration profile, an apparent diffusivity of 9.9×10^{-13} m^2/s was evaluated. However, after one year of diffusion, a factor of 3 lower apparent diffusivity was found. After six years of diffusion, an even lower value for D_a was found (6.1×10^{-14} m^2/s). The decrease in D_a after longer diffusion times differed by a factor almost corresponding to the ratio between the experiment times.

Fig. 3 shows the concentration profiles after three months, one year and six years of diffusion time. The data points are fairly well assembled as can be seen in the figure, showing that virtually no migration of Tc has taken place after the initial three months. This can be explained by an initial diffusion of technetium oxidised to pertechnetate, which, as the experiment proceeded, was reduced to Tc(IV). Tc(IV) is much less soluble than Tc(VII) and will, therefore, be retained in the system.

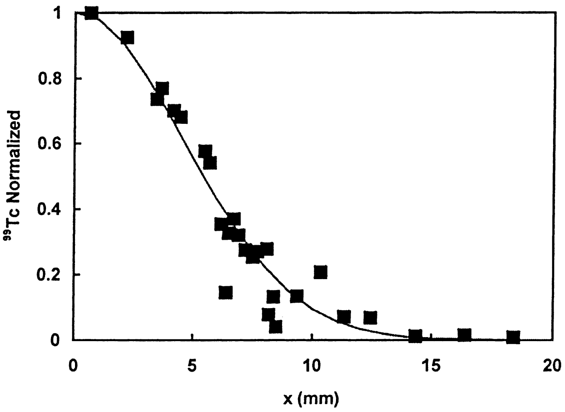

Fig. 2. Concentration profile of technetium. The contact time was 101 days and the temperature ambient hot cell temperature (25°C). The solid line represents the corresponding diffusion equation (Eq. (2)), with an evaluated D_a of 9.9×10^{-13} m^2/s.

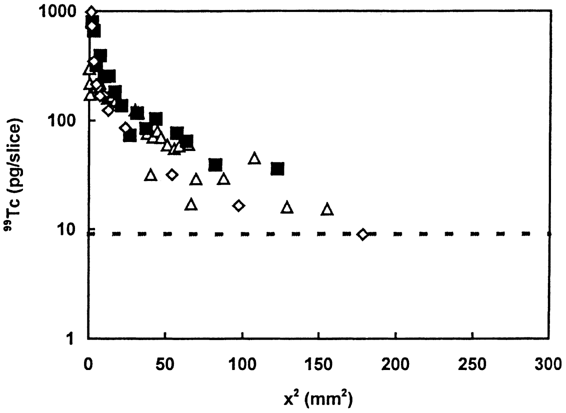

Fig. 3. Concentration profiles of technetium after different diffusion times at ambient hot cell temperature (25°C). Triangles: 3 months; squares: 1 yr; diamonds: 6 yr.

As can be seen in Table 4, bentonite contains as an auxiliary mineral 0.3% pyrite, FeS_2, but also a small amount of Fe(II) as a substitute element in the clay structure itself. Pyrite is known to oxidise quite rapidly in oxygen-containing water [23]. The amount of pyrite in the diffusion cells is about ten times more than the amount of oxygen entrapped in the closed cells, i.e. more than enough to consume all oxygen in the system. The actual rate of oxygen consumption in compacted bentonite is not accurately known, but experiments by Melamed and Pitkänen indicate that it takes place at a time scale of months [24]. Thus, the Fe(II) minerals in the bentonite is expected to consume the oxygen in the system, but also have the potential for reducing oxidised species of radionuclides released from the fuel. The reductive capacity of the bentonite is also confirmed by

Snellman et al. [25], who have shown that bentonite clay will buffer the Eh to −0.2 to −0.3 V.

When metallic iron is added to the bentonite, the reducing conditions are expected to be established more rapidly. This was also confirmed by the observation that in the cell where metallic iron was present no transport of the released technetium could be observed. The FICAP value for cell A presented in Fig. 4 is an estimated maximum value for the release of Tc in this cell.

After the initial release of technetium in the experiment, no further oxidation of technetium to pertechnetate takes place. A similar observation has been made by Forsyth and Werme [26] when corroding spent fuel in synthetic groundwater under oxidising and anoxic conditions. Under oxidising conditions, the release rates for Tc are within a fairly large spread independent of time resulting in a continuous increase in the solution concentration of Tc. This is consistent with the release of Tc as pertechnetate, which has a very high solubility under these conditions. Under anoxic conditions, the average Tc concentration was found to be 6×10^{-9} mol/dm^3. This is to be compared with the TcO_2 solubility of 3×10^{-8} mol/dm^3. Apparently, the oxidising effects of radiolysis has in these experiments been insufficient to oxidise the technetium in the fuel to Tc(VII).

A compilation of experimental apparent diffusivities of technetium can be seen in Fig. 5. The apparent diffusivities of technetium found in this work seem thus to be in the reducing condition range. Another conclusion is that the apparent diffusivities found in this work are probably overestimated, as the pre-treatment of the spent fuel pieces in air probably, including cutting the fuel specimens, caused an oxidation of technetium to Tc(VII). The diffusion profiles in Fig. 2 may, therefore, be the results of an initial diffusion of pertechnetate that was available when the experiments started.

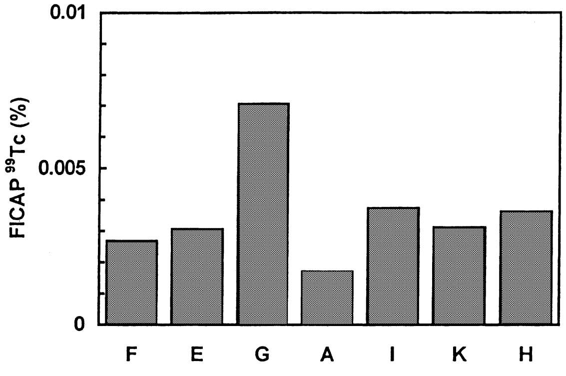

Fig. 4. Released fraction of ^{99}Tc from spent UO_2 fuel. Leaching time: F: 101 days; E: 197 days; G, A, I: 386 days; K, H: 2200 days. Additives: A: metallic iron; I, K: vivianite; F, E, G, H: no additives.

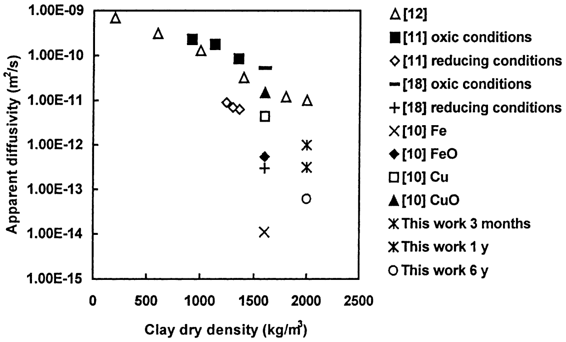

Fig. 5. Compilation of apparent diffusivities of technetium from data in the literature and the D_a found in this work.

3.1.2. Uranium

The transport properties of uranium in compacted bentonite clay were presented earlier by Ramebäck et al. [15] and will, therefore, only be summarised here. When no additives were mixed into the clay, an apparent diffusivity of 1.9×10^{-13} m^2/s was evaluated after three months of diffusion. Similar D_a data were obtained for 6.5 months and after one year. However, after six years of diffusion, the diffusivity was 1 order of magnitude lower. When metallic iron was mixed into the clay, an almost 2 orders of magnitude lower apparent diffusivity was obtained. This was interpreted as a consequence of the reducing conditions being established more rapidly by the addition of the metallic iron.

3.2. Leaching

Fig. 4 shows the FICAP of technetium. It can be seen that, when metallic iron was mixed into the clay, a decrease in FICAP with approximately 50% was noted. This is consistent with the reducing conditions being established more rapidly in the diffusion cell as a consequence of the addition of iron. With the exception of cell G, the release fractions shown in Fig. 4 are about 0.003% to compare with 0.001% measured in Studsvik's Series 11 during the first 7 days of leaching under anoxic conditions [27]. For oxidising conditions, the corresponding releases were about 0.007%. However, after this initial contact the release rates of technetium under anoxic conditions dropped almost 2 orders of magnitude, while they remained constant under oxidising conditions. This supports the observation made from calculations of the apparent diffusivity: the amount of technetium that has diffused into the bentonite clay was probably oxidised before the experiment started or was oxidised by the residual oxygen in the bentonite.

For uranium, the release fractions are generally in the range 0.004–0.006%, see Fig. 6. Cell G had a considerably higher release fraction (about 0.0016%). The decrease in FICAP when metallic iron was mixed into the

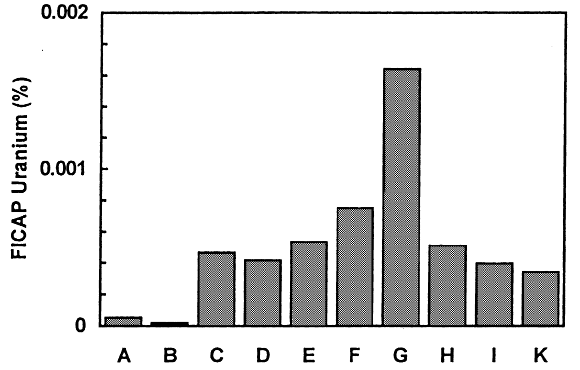

Fig. 6. FICAP of uranium from spent UO_2 fuel. Leaching time: F: 101 days; E: 197 days; G, A, I, C: 386 days; B, D, K, H: 2213 days. Additives: A, B: metallic iron; I, K: vivianite; C and D: metallic copper; E, F, G and H: no additives.

bentonite clay was much more pronounced than for technetium. Cells A and B had release fractions of 0.00006% and 0.00002%, respectively. As for technetium, this can be explained by more rapidly achieved reducing conditions by the addition of metallic iron, which is more reactive than the Fe(II) containing minerals in the clay. Leach tests in Studsvik's Series 11, performed under anoxic conditions, showed total uranium releases of about 0.0002% during the first 7 days and cumulative releases of 0.0004% after the first 8 contacts with anoxic water (4 yr cumulative time) [26]. The ninth contact showed an increased release, which may be due to loss of anoxic conditions during the test. For oxidising conditions, the uranium release during the first 7 days was about 0.002% and remained at that level for each contact period.

For the first three periods of 3, 6.5 and 12 months, respectively, comparable apparent diffusivities are calculated for uranium (1.9×10^{-13}, 2.2×10^{-13}, 1.8×10^{-13} m^2/s, respectively) [15]. This indicates that the diffusion of the mobilised uranium continued for a longer time than what was the case for technetium. Both uranium and technetium are redox sensitive. In aerated aqueous solutions at neutral at mildly alkaline pH values, the stable oxidation states are U(VI) and Tc(VII). The MX-80 bentonite pore water is expected to have pH values in the range 8.5–9.5 and bicarbonate concentrations of some several millimoles per litre [28]. Under these conditions, the stable solution species are TcO_4^- and $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)_3^{4-}$ for technetium and uranium. Under oxygen-free conditions, the corresponding solutions species are $TcO(OH)_2$ and $U(OH)_4$, respectively. The reduction of Tc(VII) to Tc(IV) as a homogenous redox process in a natural system is very slow, if it proceeds at all. Surface mediated electron transfer reactions will be the most effective pathway for the reduction [29,30]. The diffusion data for Tc and U indicate that the rate of reduction of $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)_3^{4-}$ is slower than the rate of reduction of TcO_4^-. This is not unexpected from structural chemistry considerations, but it will have to be studied further before any definite conclusions can be drawn. Such studies are in progress.

After six years a lower apparent diffusivity was found (1.6×10^{-14} m^2/s). It is possible that this difference in apparent diffusivity is caused by difference in the availability of mobilised (oxidised) uranium at the beginning of the experiment and the time required to consume the residual oxygen in the diffusion cells. It should also be pointed out that cell G had in comparison with the other cells a higher release fraction for both uranium and technetium. This was not observed for Sr which on the other hand had at least ten times higher release fractions than uranium with a continuous increase with time [17]. This is commonly observed in fuel corrosion experiments at Studsvik's Fuel Laboratory (see e.g., [26,27]). The reasons for this has not been fully clarified, but has been discussed in terms of either precipitation of secondary U(VI) phase on the fuel itself or segregated strontium phase in the fuel. No conclusive evidence has been presented in our opinion for either case.

4. Conclusions

Mixing metallic iron into bentonite clay lowered the mobility of uranium by about 2 orders of magnitude as compared to the case with no additive and the release by about 1 order of magnitude.

Metallic iron was shown to have an influence on the transport and leaching behaviour of technetium as well. The release was about 2 to 4 times lower. However, it was not possible to evaluate an apparent diffusivity when iron was mixed into the clay, and it may be questioned whether an apparent diffusivity can be evaluated. In pure bentonite, it was shown that the concentration profiles were very similar using different diffusion times. This is consistent with a reduction of Tc(VII) to Tc(IV) in the bentonite clay. The data for uranium also indicate a reduction of U(VI) to U(IV), but the reduction kinetics seem to be slower than for Tc. Further research will be required to confirm this observation.

Acknowledgements

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