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Determination of U(IV) and U(VI) by ion chromatography–inductively coupled plasma mass spectrometry and its application to kinetic studies

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Abstract

The ions normally formed by actinides in aqueous solutions by the oxidation states III–VI are M^{3+} , M^{4+} , MO_2^+ and MO_2^{2+} , respectively. Oxidation state representatives such as Am^{3+} , Th^{4+} , NpO_2^+ and UO_2^{2+} , which resist oxidation state changes, were used to investigate a method to separate the oxidised species (MO_2^+ and MO_2^{2+}) from the reduced species (M^{3+} and M^{4+}). With this method the hexavalent state of uranium could be separated from the tetravalent state of uranium in aqueous media in less than 8 min. Uranium concentrations down to 10^{-9} M could be analysed without changing the redox composition during the separation. The oxidation kinetics of the tetravalent uranium for different hydrochloric acid concentrations was investigated. The measurements showed good agreement with values found in the literature, although the uranium concentrations were one million times lower. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Kinetic studies; Actinides; Uranium

1. Introduction

Direct disposal of spent nuclear fuel in deep geological formations is being considered by several countries [1]. The resistance to radionuclide release to the biosphere is primarily dependent on the chemical stability of the UO_2 spent fuel matrix [2,3]. The near field performance assessment requires information about the spent fuel matrix dissolution under both reducing and oxidising conditions [4,5]. These chemical conditions are usually described by the Eh (equilibrium reduction potential with respect to a normal hydrogen electrode), which often is

measured with an inert electrode such as Pt [6–8]. Because of the low concentrations of dissolved actinides and fission products in leaching solutions, the measured potential need not correspond to a single redox process but is a composite of two or more processes. The measured mixed potentials are often not in accordance with simple Nernst theory and cannot be quantitatively interpreted. Several authors have pointed out that the most serious problem in applying the concept of Eh to natural low-temperature aqueous systems is the lack of internal equilibrium. Hence, the Eh values computed from analytical data on the various redox couples do not agree with one another [9–12]. We believe that the redox conditions in leaching solutions could be better described by measuring certain redox sensitive species. For spent fuel dissolution processes under reducing and oxidising conditions it is advisable to

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start a speciation analysis of the uranium in the oxidation state IV and VI.

Ion chromatography with inductively coupled plasma (ICP)-MS detection has been used to analyse very low concentrations of actinides and lanthanides [13–17]. An ion chromatography method to separate the oxidised actinide species (MO_2^+ and MO_2^{2+}) from the reduced species (M^{3+} and M^{4+}) is investigated in this work. In aqueous solutions uranium is only stable in the hexavalent and tetravalent oxidation state [18]. The hexavalent state of uranium is separated from the tetravalent state of uranium in aqueous media at concentrations as low as 10^{-9} M without changing the composition of the redox species. A major problem for speciation analyses is the handling of samples without changing the redox composition. Some results of the oxidation kinetics of the tetravalent uranium for different aqueous solutions are presented.

2. Experimental

2.1. Instrumentation

The ion chromatography system consisted of two Dionex (Sunnyvale, CA, USA) advanced gradient pumps (AGP-1 and AGP-2) and two Dionex BF-4 four-way valves (injection valve and post-column valve). The sample loop (0.5 ml) was a 72 cm long Tefzel tube (1.57 mm O.D. \times 0.94 mm I.D.). A Dionex cation chromatography column (IonPac CG10, 50 \times 4 mm) was used for all separations. The column is packed with a latex based cation-exchange resin carrying negatively charged sulfonate functional groups [19]. In order to pump high acid concentrations, acid resistant pistons had to be used. All the chromatography items except the gradient pumps and the eluent bottles were installed in a glove box connected with a ventilation system for radiological use.

A VG Elemental (Winsford, UK) Plasmaquad PQ2+ inductively coupled plasma mass spectrometer had been installed in the same glove box. Two independent cooling systems were used, one for the interface (18°C) and the other for the spray chamber (2°C). Platinum sample cones were used in order to analyse high acid concentrations. The response was

about 50 000–100 000 counts/s for a 1 ng/ml In-115 standard. Data were acquired in the time resolved analysis mode of the software version Plasmaquad 4.3. The signals for the masses 232, 237, 238, 239 and 241 were measured in the peak jumping mode with three points per mass and a dwell time of 81.9 ms per point. This resulted in one mass spectrum every 1.28 s. The chromatograms were integrated with the software Mass Lynx version 2.0.

HPLC was run with a flow-rate of 1 ml/min and could directly be coupled to the ICP-MS system. Fig. 1 shows a schematic view of the coupled system. The separation procedure is divided in two different valve positions.

2.1.1. Load

A Gilson (Gilson Medical Electronics, Villiers-le-Bel, France) Minipuls peristaltic pump was used to suck and push the sample through the sample loop of the injector valve into the waste container. The peristaltic pump was also used to suck solution from the spray chamber of the ICP-MS system to the waste. The pump operated at 35 revolutions/min. To fill the loop, the peristaltic pump was equipped with a poly (vinyl chloride) (PVC) tube with an inner diameter of 0.5 mm and a resulting flow-rate of 0.84 ml/min. For the outlet of the spray chamber the peristaltic pump was equipped with a PVC tube with an inner diameter of 0.9 mm and a resulting flow-rate of 2.9 ml/min. Eluent was pumped with the AGP1 through the column at a flow-rate of 1 ml/min and passed through the post-column valve into the waste container. With the AGP2, 1% nitric acid was pumped in the flow mode through the post-column valve into the spray chamber of the ICP-MS system.

2.1.2. Inject

After 1 min the injection valve was changed to the inject position. At the same time the post-column valve position was changed from position “waste” to “ICP-MS”. The sample solution was then sucked and pushed with the peristaltic pump into the waste container. The AGP1 pumps the sample solution in the loop through the chromatographic column and the post-column valve into the spray chamber of the ICP-MS. The AGP2 pumps 1% nitric acid through the post-column valve into the waste container.

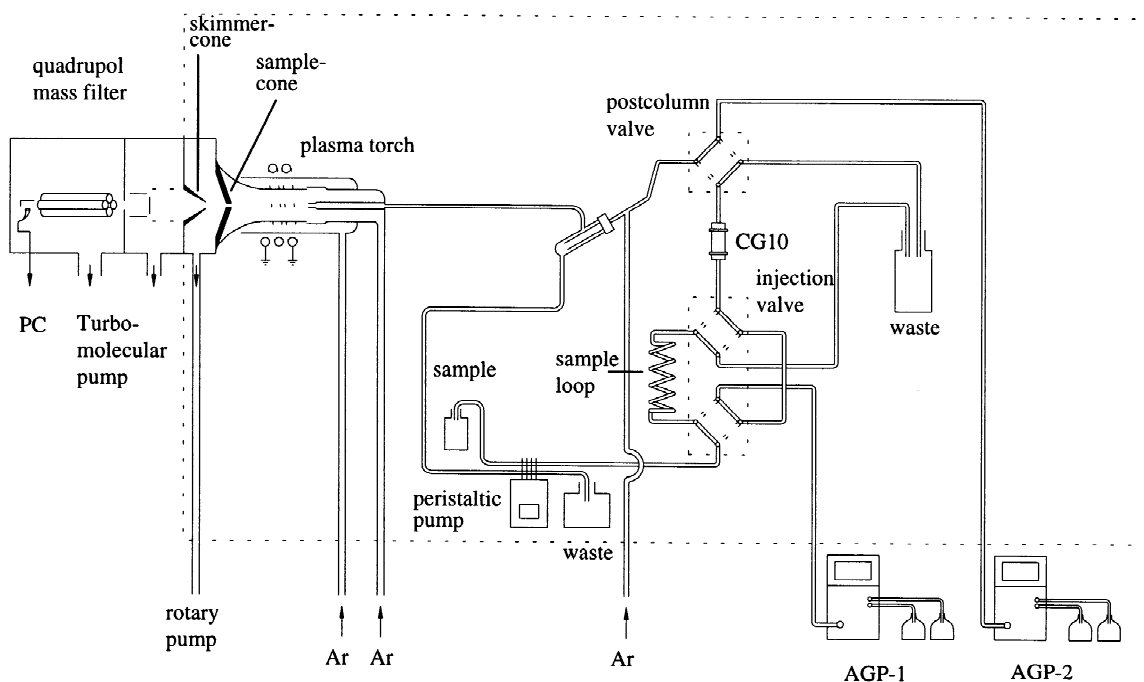


Fig. 1. Schematic view of the coupling of the HPLC and ICP-MS systems. In order to analyse highly toxic radioactive samples, all parts of the system which have contact with the samples are installed in a glove box.

2.2. Reagents, standards and samples

Distilled, deionized water from a Milli-Q system (Millipore, Milford, MA, USA), 65% nitric acid, 30% HCl (analysed: 31.6%) and 96% H₂SO₄ (analysed: 96.4%) all suprapure from Merck (Darmstadt, Germany) and 48% hydrofluoric acid pro analysis from Merck were used. Spex 10 µg/ml multielement standard solutions (SPEX Industries, Edison, NJ, USA) and a uranium 1000 ppm solution from Merck were used as stock solutions. To reduce UO₂²⁺ hydrogen gas (Fogon 5% H₂–95% Ar and 100% H₂) from AGA (AGA Special Gas, Lidingö, Sweden) was used.

Furthermore, our own actinide standard solutions were used. The Th, Am and Np standard solutions were mono isotopic and contained Th-232, Am-241 and Np-237. The Pu standard solution contained the isotopes: Pu-238 (0.13 mol%), Pu-239 (84.03 mol%), Pu-240 (14.45 mol%), Pu-241 (0.73 mol%) and Pu-242 (0.34 mol%). A uranium standard was made by dissolving uranium oxide. The UO₂ had an isotopic composition of 1.1 mol% U-235 and 98.9%

U-238. A 1.29-g aliquot of UO₂ was added to 80.1 g of 16.5% HCl (4.9 M). Three drops of HF were added and heated to 96°C. As after 2 weeks not all the UO₂ was dissolved, three drops of HF were added again. After 17 days all the UO₂ was dissolved. The solution with the dissolved UO₂ weighed 76.1 g. The uranium concentration was hereby 14.9 mg/g. Then, 2.2 ml of this solution was added to 122.3 g of 1 M HCl. This resulted in a 284 µg/g uranium solution in 1.06 M HCl. This solution was used as stock solution for further dilutions.

3. Results and discussion

3.1. Separation method

The ions normally formed by actinides in aqueous solutions by the oxidation states III–VI are M³⁺, M⁴⁺, MO₂⁺ and MO₂²⁺, respectively. Oxidation state representatives such as Am³⁺, Th⁴⁺, NpO₂⁺ and UO₂²⁺, which resist oxidation state changes, were used to evaluate a separation methods for all four

oxidation states [15]. The separation methods were further developed to separate the oxidised from the reduced actinide species. Considering the isotopic compositions of the actinide standards, the masses 232, 237, 238, 239 and 241 can be assigned to the actinides Th, Np, U, Pu and Am, respectively. Ions in the oxidation state IV often elute as wide peaks. By applying only the Dionex CG 10 guard column these ions could be eluted with H_2SO_4 concentrations as low as 0.5 M without getting wide peaks. Fig. 2 shows the elution time for the different ionic species. Np(V) is not retained on the column and elutes after 1.7 min for all H_2SO_4 concentrations. With 0.1 M H_2SO_4 , U(VI) elutes just after Np(V), whereas Th(IV) and Am(III) would hardly move down the column. With 0.5 M H_2SO_4 , all the actinide species are eluted in less than 3 min.

Table 1 shows the elution parameters to separate the oxidised species, NpO_2^+ and UO_2^{2+} , from the reduced species, Th^{4+} and Am^{3+} . The oxidised species were eluted with 0.1 M H_2SO_4 and the reduced species with 0.5 M H_2SO_4 . The column was

cleaned for 11 min with 1 M H_2SO_4 . Fig. 3 shows the chromatogram of 0.5 ml of 0.1 M HNO_3 containing 5 ppb of Pu, Am, Np, Th and 7.5 ppb of U. The oxidised actinide species are completely separated from the reduced species. Th(IV) elutes before Am(III) due to complex building with HSO_4^- . With the same chromatographic conditions uranium solutions containing the species U^{4+} and UO_2^{2+} were separated. Fig. 4 shows the separation of three 10-ppb uranium solutions in different media. The retention time of U(IV) does not depend on the medium and is in accordance with the retention time of Th(IV). The retention time of the species UO_2^{2+} is slightly dependent on the sample medium. For a uranium solution in 1 M HCl, U(VI) elutes as a wide peak. Most of the U(VI) is not retained at all and elutes at the same time as Np(V). Another part of the U(VI) elutes at the same time as U(VI) in 1% HNO_3 . In [15] the reason for this splitting was thought to be that the two species, UO_2^+ and UO_2^{2+} , were coexisting in the solution. This is probably not true. It is more likely that a high chloride con-

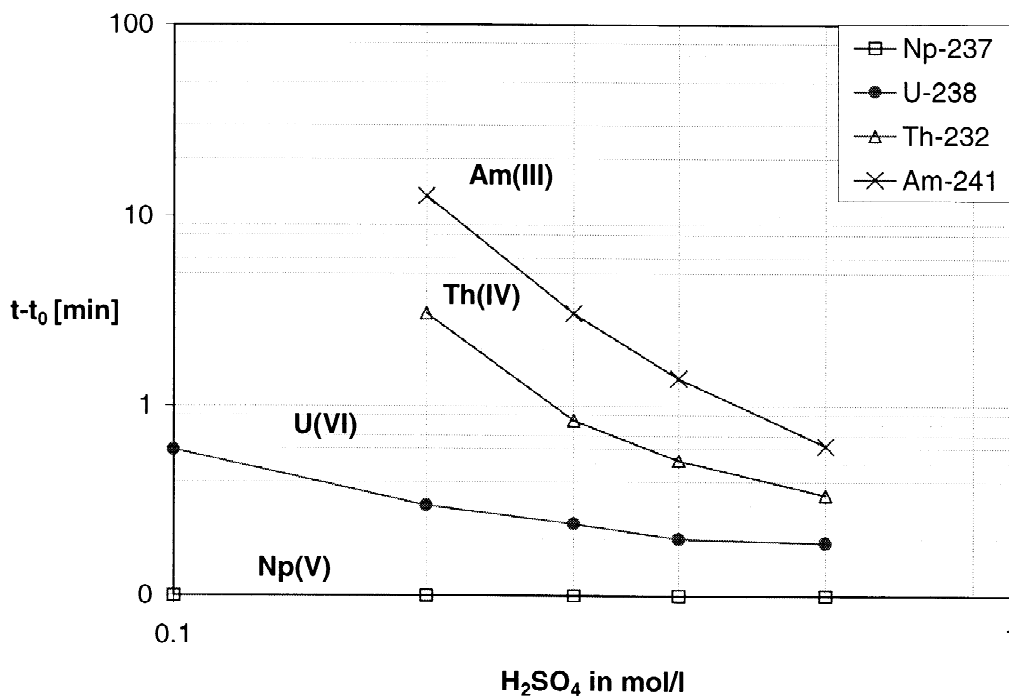


Fig. 2. Adjusted elution times for actinides in different oxidation states. Samples of 0.5 ml of 1% HNO_3 containing 5 ppb of Am, Np, Th and 7.5 ppb of U were eluted isocratically with different H_2SO_4 concentrations.

Table 1

Chromatographic conditions to separate the oxidised actinide species from the reduced actinide species

AGP1 time (min)	mQ-H ₂ O (%)	1 M H ₂ SO ₄ (%)	Injection valve position	Post column valve position
Initial	90	10	Load	Waste
0.0	90	10	Load	Waste
1.0	90	10	Inject	ICP-MS
3.0	90	10	Inject	ICP-MS
3.1	50	50	Inject	ICP-MS
9.0	50	50	Inject	ICP-MS
9.1		100	Load	Waste
20.0		100	Load	Waste
20.1	90	10	Load	Waste

centration in the sample medium is enough to elute part of the UO_2^{2+} faster than expected with a 0.1 M H_2SO_4 eluent. However, the U(VI) species are completely separated from the U(IV) species. Any shifting in the redox state composition during the separation would lead to bridged peak shapes. No bridged peaks between the two oxidation forms were found and thus no U^{4+} was oxidised to UO_2^{2+} on the column.

A 10-ppb U(IV) solution in 0.1 M HCl was oxidised in an air atmosphere. Samples were withdrawn periodically and directly analysed. Fig. 5 shows the proceeding of the oxidation reaction. At the beginning, the solution contained 91% U(IV) and

at the time when the last sample was withdrawn the solution contained 14% U(IV). De Beer [20] separated the uranium species with anion chromatography and found bridged peaks especially for low concentrations [2 ppm U(IV)]. With the separation method described here, U(VI) is completely separated from U(IV) for all compositions. No bridged peaks were found for U(IV) concentrations down to 1 ppb.

3.2. Calibration

The sensitivity for the uranium species UO_2^{2+} and U^{4+} was determined with a one point calibration for

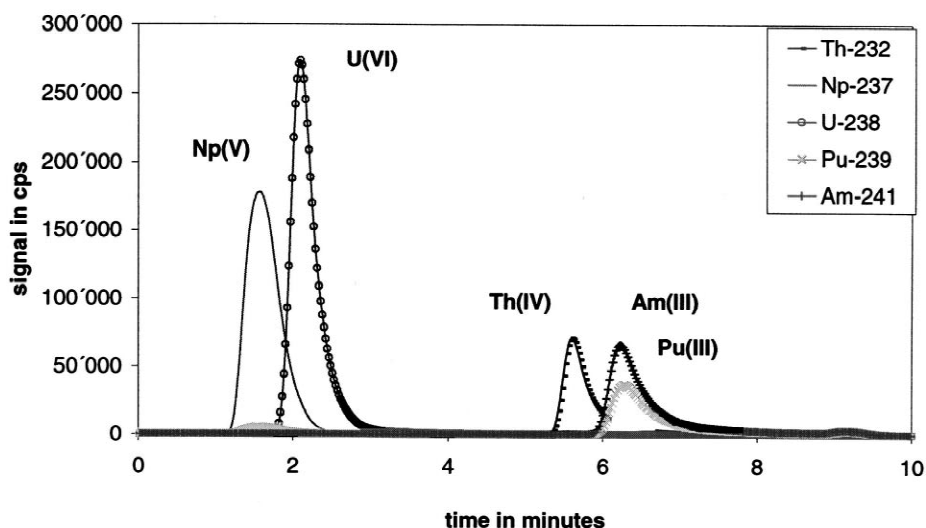


Fig. 3. The separation of 0.5 ml of 1% HNO_3 containing 5 ppb of Am, Np, Th and 7.5 ppb of U with the chromatographic conditions from Table 1. The oxidised actinide species NpO_2^{2+} and UO_2^{2+} are completely separated from the reduced actinides species Th^{4+} and Am^{3+} .

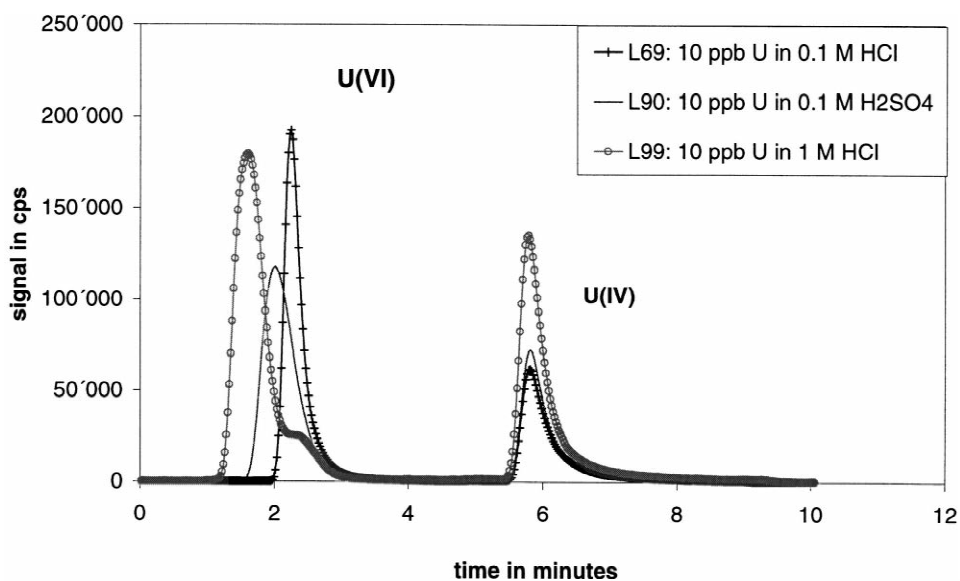


Fig. 4. Samples of 0.5 ml of 10-ppb U in three different media were separated with the chromatographic conditions from Table 1. The solutions contained the species UO_2^{2+} and U^{4+} . The oxidised uranium species UO_2^{2+} are completely separated from the reduced uranium species U^{4+} for all three media. The retention time of U(VI) is slightly dependent on the medium.

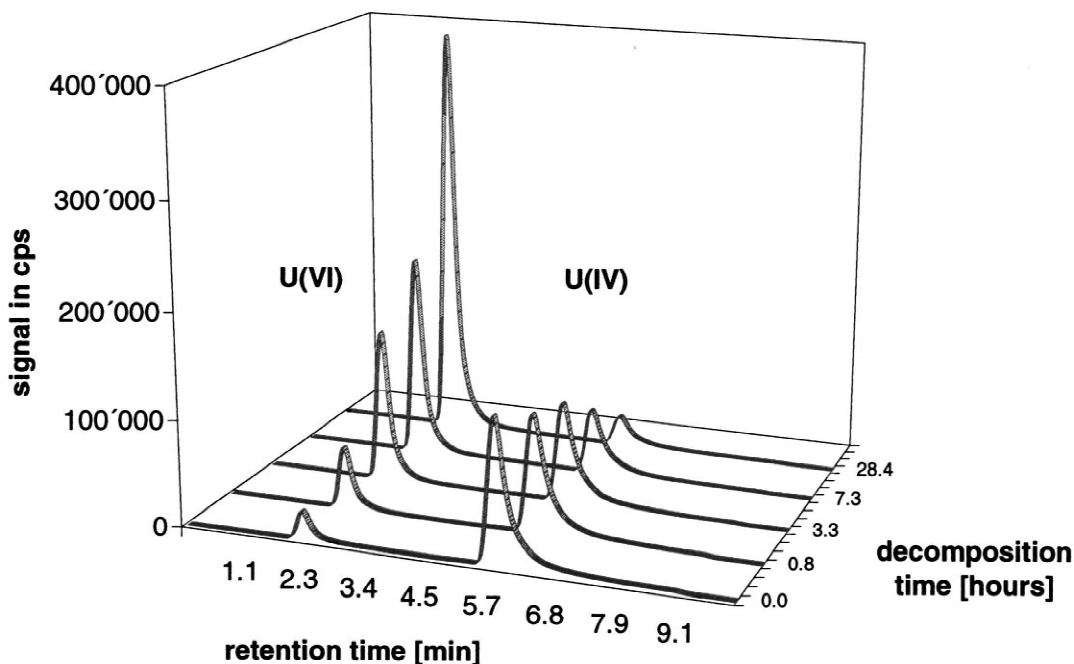


Fig. 5. A 10-ppb U(IV) solution in 0.1 M HCl was oxidised in an air atmosphere. Samples were withdrawn periodically and directly analysed. U(VI) is completely separated from U(IV) for all compositions.

each species. The 284- $\mu\text{g/g}$ uranium standard solution was diluted with 1 M HCl to a concentration of 1 $\mu\text{g/g}$ U and reduced with hydrogen and a Pd coated Pt-foil. This stock solution was used to make the following two calibration solutions.

1. Calibration solution for U(IV): A 0.1049-g aliquot of 1 $\mu\text{g/g}$ U was added to 9.9738 g of 1 M HCl. With a specific mass of 1.02 g/ml for the solution this results in a 10.6 ng/ml uranium solution. The solution was directly analysed. The integrated peak area for U(VI) and U(IV) was 82 995 and 5 791 870 cps, respectively. Without any sensitivity corrections a composition of 98.6% U(IV) was obtained. The solution contains hereby 10.5 ng/ml U(IV) and the sensitivity is 551 607 cps per ng/ml of U(IV).
2. Calibration solution for U(VI): A 0.202-g aliquot of 1 $\mu\text{g/g}$ U was added to 19.84 g of 0.01 M NaCl solution. With a specific mass of 1 g/ml for the solution this results in a 10.1 ng/ml uranium solution. This solution was oxidised in air to 100% U(VI) and analysed. The integrated peak area for U(VI) was 8 893 853 cps. The sensitivity for U(VI) is hereby 880 580 cps per ng/ml of U(VI).

Without any sensitivity corrections a sensitivity factor [sensitivity for U(IV)/sensitivity for U(VI)] of 0.63 was obtained. The composition of the calibration solution for U(IV) was recalculated by applying this sensitivity factor. A composition of 99.1% U(IV) was found. The difference was small and the same sensitivity factor was obtained by recalculation.

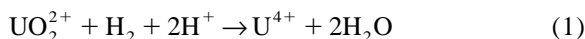
To check the linearity of the calibration, a mixed solution was made out of the two calibration solutions. The composition for the mixed solution was 65.9% U(IV). A chromatographic determination led to a composition of 66.3% U(IV) and agreed perfectly with the theoretical composition. As U(IV) solutions are not stable in air a one point calibration will probably lead to the best results. Thus, for all determinations the sensitivity factor of 0.63 was used.

3.3. Application: kinetic studies

3.3.1. Reduction of UO_2^{2+} to U^{4+} with H_2

Two UO_2^{2+} solutions were reduced with hydrogen

on a Pt-foil with a surface of 4 cm^2 . Both experiments were made at room temperature in glass vessels. To have an estimation about the rate of the reduction, samples were withdrawn periodically and diluted with 1 M HCl to 10 ppb U solutions. These solutions were measured right after the dilution. In the first experiment 5% H_2 –95% Ar was bubbled over a Pd coated Pt-foil in a 1 M HCl solution containing 1 ppm uranium. In the second experiment 100% H_2 was bubbled over a Pt-foil in a 1 M HCl containing 10 ppm uranium. The overall reduction reaction can be written as follows:



The proceeding of the reduction reaction is shown in Fig. 6. The rate of the reduction of U(VI) was found to show a first order dependence on the concentration of U(VI) according to:

$$d[\text{U(VI)}]/dt = -k_{\text{red}}[\text{U(VI)}] \quad (2)$$

The experimental points for the reduction with 100% H_2 on a Pt-foil were fitted by the method of least squares for an equation form $y = ce^{-bx}$, where y = percentage of U(VI), $b = k_{\text{red}}$ and x = time in h. The equation $y = 0.73e^{-0.0046x}$ was found. The half-life time for the reduction of U(VI) is hereby 151 h. The reduction with the Pd coated Pt-foil between points 4 and 5 was disturbed by O_2 diffusing in the glass vessel and causing an oxidation of U^{4+} . By using a higher gas flow, about the same reduction rate as for the Pt-foil was found.

3.3.2. Oxidation of U^{4+} to UO_2^{2+} with O_2

A major problem for speciation analyses is the handling of samples without changing the redox composition. In order to estimate the oxidation rate for U^{4+} solutions which are in contact with air, a series of 10 ppb uranium solutions were prepared by appropriate dilution of the reduced 1 ppm uranium stock solution. The experiments were made in glass vessels at room temperature. About 200 ml of 10 ppb U(IV) solutions were prepared in 0.01 M HCl, 0.1 M HCl, 0.3 M HCl, 1.0 M HCl and 0.1 M H_2SO_4 . The vessels were open to the air atmosphere. The oxygen partial pressure was thus about 0.2 bar. The dissolved oxygen concentration was assumed to be

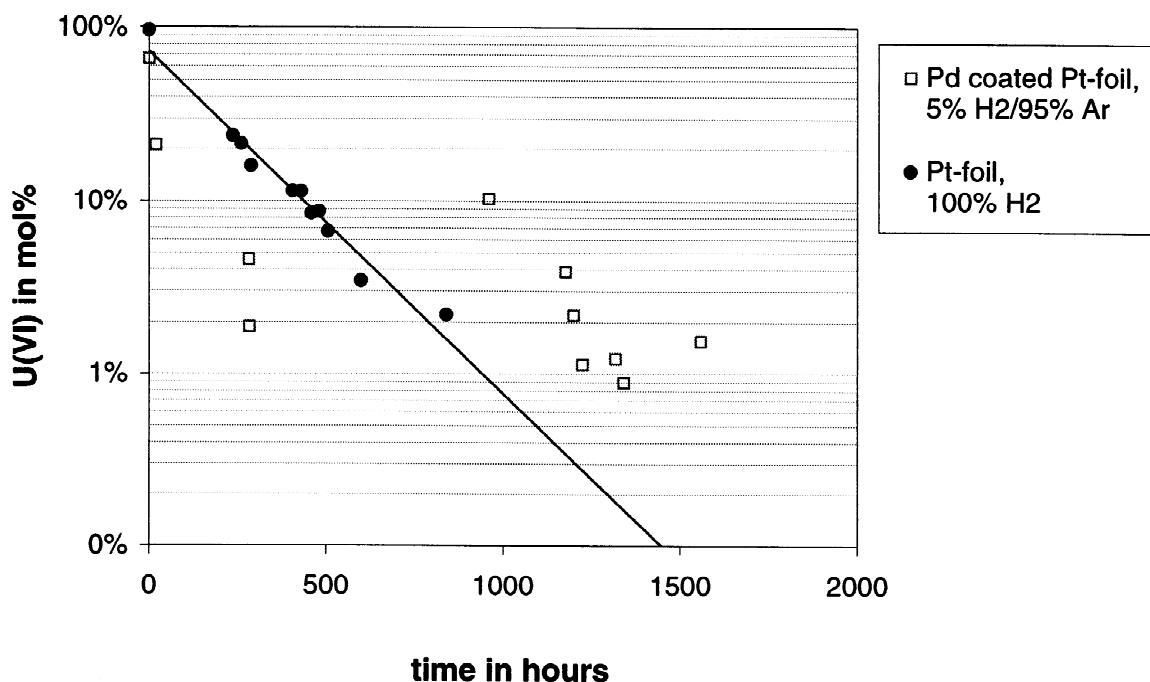


Fig. 6. Two UO_2^{2+} solutions in 1 M HCl were reduced with hydrogen on a Pt-foil. A 1 ppm UO_2^{2+} solution was reduced by bubbling 5% H_2 -95% Ar over a Pd coated Pt-foil and a 10 ppm UO_2^{2+} solution by bubbling 100% H_2 over a Pt-foil.

equal to the oxygen concentration in pure water and was expected to be about $2.5 \cdot 10^{-4}$ M. For a 10 ppb U^{4+} solution ($4 \cdot 10^{-8}$ M) the consumption of oxygen for the complete oxidation of U^{4+} to UO_2^{2+} is expected to be much smaller than the dissolved oxygen concentration. The concentration of dissolved oxygen can therefore assumed to be constant during the oxidation reaction. The hydrogen ion concentration was >0.01 M in all experiments and can also assumed to be constant as the hydrogen ion production for the complete oxidation of a 10 ppb U^{4+} solution is much lower. Samples were withdrawn periodically and analysed directly to determine the extent of the oxidation reaction. The overall oxidation reaction can be written as follows:



Fig. 7 shows the U(IV) percentage in mol% vs. time. The rate of the oxidation of U(IV) showed a first order dependence on the concentration of U(IV) according to:

$$d[\text{U(IV)}]/dt = -k_{\text{ox}}[\text{U(IV)}] \quad (4)$$

The experimental points were fitted by the method of least squares for an equation form $y = c e^{-bx}$, where y = percentage of U(IV), $b = k_{\text{ox}}$ and x = time in h. Table 2 shows the equations, the apparent first order constants and the half-life times ($t_{1/2} = \ln 2 / k_{\text{ox}}$) for the different solutions. The reaction rate was found to be independent of the U(IV) concentration in the range $4 \cdot 10^{-8}$ – $4 \cdot 10^{-11}$ M for all HCl solutions. In contrast to this, previously in the literature [21,22] some departures from this rate law for HClO_4 concentrations of >0.2 and <0.07 M after the reaction had proceeded about halfway to completion had been found.

The apparent first order reaction constant for the oxidation of U(IV) was found to depend on the oxygen and hydrogen ion concentration. The kinetics of the oxidation reaction in perchloric acid could approximately be described by Eq. (5) for a fairly wide range of conditions, where k is the oxidation reaction constant [21]:

$$k_{\text{ox}} = k \cdot \frac{[\text{O}_2]}{[\text{H}^+]} \quad (5)$$

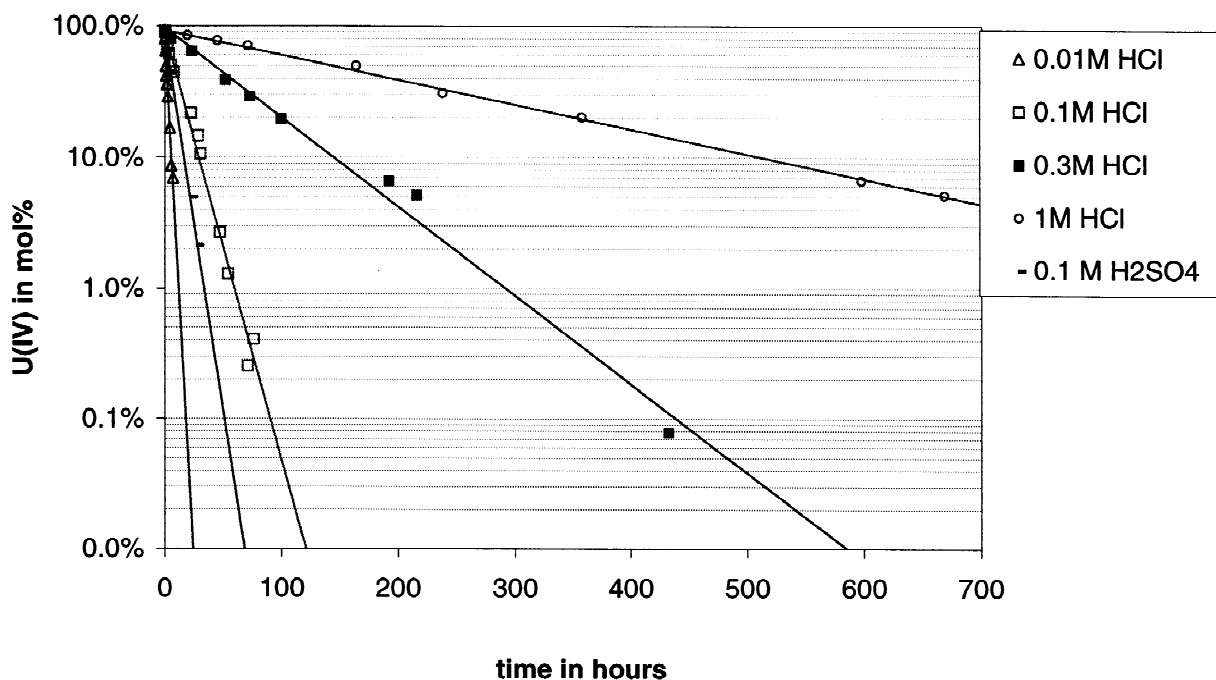


Fig. 7. The oxidation of 10 ppb U(IV) solutions in an air atmosphere. In all media the percentage of U(IV) in the logarithmic mode decreased linearly with time.

The half-life time for the oxidation of U(IV) is thus approximately proportional to the hydrogen ion concentration according to Eq. (6):

$$t_{1/2} = \frac{\ln 2}{k_{\text{ox}}} = \frac{\ln 2}{k \cdot [\text{O}_2]} \cdot [\text{H}^+] \quad (6)$$

In Fig. 8 the half-life times for the oxidation of U(IV) in 1.0, 0.3, 0.1 and 0.01 M HCl are plotted vs. the HCl concentration. The dissolved oxygen concentration was assumed to be 2.5×10^{-4} M. The four

experimental points were fitted by the method of least squares for a polynomial function through the origin. No attempt was made to investigate the exact influence of the hydrogen ion concentration on the oxidation reaction constant. The intention was rather to show the strong influence of the HCl concentration on the oxidation reaction rate. The diagram can be used to estimate the reaction rate for high HCl concentrations which are used to store U(IV) solutions. For a more accurate kinetic study more care must be taken to have well controlled experimental conditions (more accurate acid concentration, constant temperature and constant oxygen concentration).

In Table 3 the oxidation reaction constants found in this work for HCl solutions are compared with values found by other authors for HClO_4 . Halpern et al. [21] found an inhibiting effect of Cl^- on the oxidation reaction. Addition of only 0.001 mol/l of NaCl decreased the rate to about half its original value. Above 0.01 mol/l of NaCl, the inhibiting effect decreased the rate to about one-fifth of the uninhibited rate. As all solutions in our work contain

Table 2

The fitted equations, the apparent oxidation reaction constants and the half-life times for the oxidation of U(IV) with O_2 in different media^a

Media	Equation	k_{ox} (h^{-1})	$t_{1/2}$ (h)
1.0 M HCl	$y = 0.92e^{-0.0043x}$	0.0043	161.2
0.3 M HCl	$y = 0.96e^{-0.016x}$	0.016	44.1
0.1 M HCl	$y = 0.88e^{-0.075x}$	0.076	9.2
0.01 M HCl	$y = 0.72e^{-0.367x}$	0.367	1.9
0.1 M H_2SO_4	$y = 0.83e^{-0.132x}$	0.133	5.2

^a The experimental points in Fig. 7 were fitted by the method of least squares.

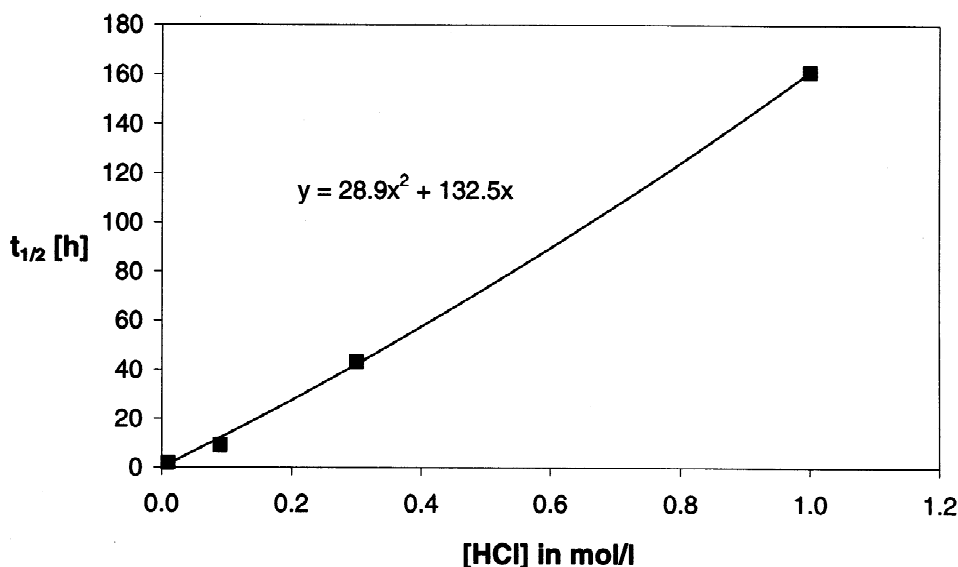


Fig. 8. The half-life times for the oxidation of U(IV) in 1.0, 0.3, 0.1 and 0.01 M HCl are plotted vs. the HCl concentration. The four experimental points were fitted by the method of least squares for a polynomial function through the origin.

more than 0.01 M NaCl, this has to be considered when comparing the oxidation rates with rates found in perchloric acid. In our work, taking in account a decreasing of the rates to one-fifth of the uninhibited

Table 3

The oxidation reaction constants are compared with values from other authors^a

Experimental conditions	k (s ⁻¹)	Ref.
0.01 M U(IV) $T=30^{\circ}\text{C}$, $p(\text{O}_2)=0.2$ bar in 0.08 M HClO ₄	$1.6 \cdot 10^{-2}$	[21]
0.01 M U(IV), $I=1$ M $T=25^{\circ}\text{C}$, $p(\text{O}_2)=1$ bar in 0.3 M HClO ₄	$2.4 \cdot 10^{-2}$	[22]
4×10^{-8} M U(IV) room temperature, $p(\text{O}_2)=0.2$ bar		
in 0.01 M HCl	$4.1 \cdot 10^{-3}$	This work
in 0.1 M HCl	$7.6 \cdot 10^{-3}$	This work
in 0.3 M HCl	$5.3 \cdot 10^{-3}$	This work
in 1.0 M HCl	$4.8 \cdot 10^{-3}$	This work

^a In order to compare the oxidation reaction constants found in this work for HCl solutions with values found for HClO₄ solutions, the inhibiting effect of Cl⁻ has to be considered. The oxidation rate constants in this work have to be multiplied by five, as the Cl⁻ ion concentrations decrease the rate to about one-fifth.

rate, the rates for the oxidation in hydrochloric acid were found to agree quite well with the rates found in perchloric acids [21,22], although in our study the experiments were made at a uranium concentration which was lower by a magnitude of one million. Thus the reaction rates found in our work do not point at a uranium concentration dependency. Hussonnois et al. [23] found the reaction rate to be dependent on the uranium concentration. A decrease of the reaction rate to 10% was found for a uranium concentration of 10^{-8} M compared to a uranium concentration of 10^{-5} M. In contrast to this, the authors in Ref. [21] did not find any dependence on the uranium concentration in the range 0.05–0.15 M.

4. Conclusions

It was shown that the hexavalent state of uranium can be separated from the tetravalent state of uranium in aqueous media in less than 8 min. Uranium concentrations down to 10^{-9} M could be analysed without changing the redox composition during the separation. The oxidation kinetics of the tetravalent uranium for different hydrochloric acid concentrations were determined. The measurements

showed good agreement with values found in the literature, although the uranium concentrations in this work were one million times lower.

The method shall be applied to measure redox compositions of uranium in leach solutions. Furthermore, the separation method shall be developed to perform speciation analysis of other actinides.

Acknowledgements

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