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Development of a radiochemical separation for selenium with the aim of measuring its isotope 79 in low and intermediate nuclear wastes by ICP-MS

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Abstract

Selenium (Se) 79 is a beta emitter produced from ^{235}U fission thus occurring as one of the fission products found in nuclear reactors. Due to its long half life (about 10^5 years), ^{79}Se is one of the radionuclides of interest for the performance of assessment studies of waste storage or disposal. Thus, the National Radioactive Waste Management Agency (Andra, France) requests its monitoring in wastes packages before their disposal in specific sites.

Measurement of ^{79}Se is difficult owing to its trace level concentration and its low activity in nuclear wastes. A radiochemical procedure has to be carried out in order to separate selenium from the matrix and to concentrate it before the measurement with a mass spectrometric or a nuclear technique. The beginning of the development is presented in this paper. The optimised protocol firstly developed in view of an ICP-MS measurement, includes five steps based on microwave digestion, evaporation and separations on ion exchange resins. It was tested first on synthetic solutions and was optimised in order to be applicable to a large number of sample types. The recoveries of the whole procedure were evaluated using natural ^{82}Se or the gamma emitter ^{75}Se as a radioactive spiker. Then, the protocol was applied to two solid samples spiked with natural selenium, a glass microfiber filter and an ion exchange resin, and two liquid samples spiked with ^{75}Se, a synthetic solution and an effluent. The yields obtained for both samples ranged from 70 up to 80%.

Keywords: Selenium; Intermediate low level waste; Radiochemistry

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1. Introduction

The control of long half life radionuclides in waste packages is necessary to insure the respect of waste acceptance criteria that are fixed in order to avoid any potential impact of the radio contaminants on the environment of the repository site. Selenium (Se) 79 is a beta emitter produced by the fission of ^{235}U thus occurring as one of the fission products found in nuclear reactors and the value of its long half life is not precisely established up to now (about 10^5 years) [1–3]. It is one of the radionuclides of interest for the performance of assessment studies because of its potential migration ability to the environment by leaching/dissolution and then transport by flowing groundwater [4–6]. In France, the National Radioactive Waste Management Agency (Andra) is in charge of the management of nuclear waste repositories. Waste producers have to respect packaging specifications in view of waste acceptance criteria. The highest global activity for the low level wastes (LLW) and intermediate level wastes (ILW) packages is 10^5 and 10^6 Bq g^{-1}, respectively. These wastes come from various origins (e.g. nuclear industry, research labs, hospitals) and are made up of very heterogeneous materials such as filters, gloves, water treatment resins or wood for example.

^{79}Se has never been quantified in low and intermediate level wastes. Only few papers deal with this radionuclide measurement in high level wastes (solution of fission products) resulting from nuclear fuel reprocessing [2,7–10]. In these studies, the analytical procedure includes: (i) a preliminary separation

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allowing to isolate selenium by complexation followed by a liquid–liquid extraction [11,12], a precipitation [8,9] or an ion exchange method [2,7] and (ii) the measurement of selenium by a nuclear technique (liquid scintillation) [7,9] or a mass spectrometric technique (ICP-MS or SMA) [2,10]. ^{79}Se activity in fission product solution has already been evaluated by liquid scintillation measurement but the values obtained were very variable [7,13] (from 6 up to 2300 Bq ml^{-1} according to the sample and the study), certainly because of the potential contribution of other β emitters remaining in the measured solution. Moreover, Comte et al. [2] have determined a concentration of 0.43 mg l^{-1} by ICP-MS measurement. Whatever the HLW sample measured, ^{79}Se is present at trace level in these solutions.

As a consequence, in case of LLW and ILW, ^{79}Se concentration could be lower than these values (about 10^{-3} Bq g^{-1} according to calculation codes from Andra [14]). Therefore, a sensitive and accurate analytical method has to be developed in order to allow the determination of this radionuclide at μg l^{-1} level in the largest variety of matrices as possible. Even if the ICP-MS measurement presents problems because of selenium high ionisation potential and isobaric interferences on all the natural isotopes [15,16], this analytical technique has been chosen for its high selectivity and sensitivity. In case of ^{79}Se measurement by mass spectrometry, many elements or molecular species arising from the sample matrix (^{79}Br$^+$, ^{158}Gd^{2+}, ^{63}Cu^{16}O$^+$) and from recombination in argon plasma (^{39}K^{40}Ar$^+$, ^{38}Ar^{40}ArH$^+$), can induce interferences [2]. Therefore, to allow the measurement of ^{79}Se in complex matrices, a radiochemical procedure has to be carried out firstly in order to decrease sample global activity, considering that ICP-MS is not situated in a glove box and cannot accept radioactive samples (for a measurement without radioprotection precautions), then to preconcentrate selenium and finally to eliminate potential interferences, particularly ^{79}Br$^+$.

The final goal was to propose an universal radiochemical protocol for the determination of ^{79}Se at trace level in all types of ILW and LLW samples in view of LSC or ICP-MS measurement. This paper only presents the first part of the initiated study. As no ^{79}Se source is commercially available, the development was performed with a natural selenium standard and the validation was achieved with ^{75}Se, used as a radioactive spiker. The different goals of the separation are the following ones:

- find a mineralization protocol adapted to the largest number of samples as possible;
- test selenium recovery after an evaporation step in order to make a preconcentration and to allow the medium to be changed if necessary between two steps;
- eliminate radionuclides potentially occurring at high level activity such as ^{90}Sr, ^{137}Cs or ^{60}Co in order to decrease the global activity of the sample (decontamination step);
- reduce the salinity to allow the introduction of the sample in the ICP-MS plasma torch;
- eliminate species which can induce interferences during ICP-MS measurement such as ^{79}Br, ^{39}K^{40}Ar$^+$ or ^{63}Cu^{16}O$^+$.

The manipulation simplicity, the reduction of steps number and the adaptability to a large part of low level waste matrices were taken into account for the optimization experiments. The developed method includes a mineralization, a five steps separation, based on precipitation reaction and ion exchange, and finally the measurement by ICP-MS. The entire process was then applied to two representative waste samples spiked with natural selenium (a glass–fiber filter and an ion exchange resin).

2. Experimental

All the concentrations reported in this paper are expressed as the mass of selenium per unit of mass or volume. Activity concentrations are expressed as Bq g^{-1}.

2.1. Reagents and standards

All dilutions were made using deionised water (18 MΩ) obtained from a Milli-Q system (Millipore system).

Different grades of nitric acid (69%, m/m) were used. Analytical grade purchased from Prolabo was chosen for conditioning and washing steps of ion exchange resins. Ultrex grade from Baker was used for microwave digestion and elution steps on ion exchange resins.

Hydrofluoric acid (40%, m/m, for trace analysis) and sodium hydroxide (50%, m/mm) were purchased from Merck.

All compounds used to prepare standard solutions were initially dissolved with deionised water to form stock solutions containing 1000 μg(Se) ml^{-1}. Serial dilutions were performed to obtain the analytical concentrations required. All solutions analysed were prepared in 2% nitric acid. Selenate [Se(VI)], as sodium selenate (99.80%) and selenite [Se(IV)] as sodium selenite (99.75%) were obtained from Merck.

All the nitrate salts used to make the synthetic solutions were analytical reagent grade (Merck, Prolabo).

^{75}Se(IV) spiker was used to determine separation/extraction yields after the different steps of the separation. The liquid standard solution was obtained from Cerca. The initial activity was (74 ± 11) kBq ml^{-1}. Dilutions were performed to obtain an activity close to 75 Bq for gamma spectrometry measurement.

2.2. Ion exchange resins

Two types of ion exchange resins were used:

- a strongly basic anion exchanger AG® 1-X4 (50–100 mesh)
- a strongly acid cation exchanger AG® 50-X8 (100–200 mesh).

All ion exchange resins and eco-columns were provided by Bio-Rad. Elution flow could be regulated using a pinch clip. The resin was prepared in milli-Q water, poured into the column and washed with 200 ml of HNO_3 3 mol l^{-1} in order to elute impurities. Then, the resin was conditioned with five bed volumes of eluent and a blank sample was systematically performed.

2.3. Samples

Two matrices representative of ILW samples were chosen to test the separation protocol:

- glass microfiber filters from Whatman (diameter = 2.5 cm)
- an anion exchange resin AG® 1-X4 similar to that described in Section 2.2.

Five hundred milligrams of each material was used for the mineralization. They were both spiked with 500 ng of natural sodium selenite.

An effluent obtained from a nuclear liquid waste treatment plant was also used to validate the separation procedure. Its solid content was estimated to 3.78 g l^{-1}.

2.4. Microwave digestion

The sample dissolution was carried out in a closed high-pressure microwave oven from CEM (Mars 5). 0.5 g of sample spiked with 500 ng of Se(IV) or Se(VI) were put in a Teflon vessel vial with 10 ml of HNO_3 (65%, ultrex grade) and eventually 5 ml of HF (in case of matrices containing silicium compounds). A radioactive spiker (^{75}Se) could also be eventually added. According to the manufacturer recommendations, the digestion was performed during 30 min, at 190 °C and 600 W. Then, the containers were cooled during one night before opening. The mineralized sample was removed, transferred to a 25 ml volumetric flask. The volume was adjusted with deionised water.

2.5. Analytical techniques

2.5.1. Q-ICP-MS

The instrument is a Perkin-Elmer Sciex Elan 6000 equipped with a cross-flow nebuliser and a Scott-type spray chamber. The device is not included in a glove box and is devoted to the measurement of samples containing a very low global activity. The monitored isotopes were: ^{76}Se, ^{77}Se, ^{78}Se, ^{82}Se, ^{79}Br and ^{81}Br. Operating parameters including plasma power (from 1050 up to 1300 W), nebulisation gas flow rate (from 0.8 up to 1.2 ml min^{-1}) and acquisition parameters (dwell time from 50 up to 200 ms) were optimised by injection of a 1 μg(Se) l^{-1} solution and the measurement of signal/noise ratio. Higher signal intensity was observed for all isotopes with an increase of plasma power, up to 1300 W. After optimisation, the operating conditions were as follow: Ar plasma gas flow rate: 15 l min^{-1}, Ar nebuliser gas flow rate: 0.9 ml min^{-1}, plasma power: 1300 W, dwell time: 100 ms, peak hopping acquisition mode. The *m/z* 82 is the less interfered peak and was used for quantification. A detection limit of 0.15 μg(Se) l^{-1} was obtained in these conditions (calculated according to IUPAC recommendations).

2.5.2. HG-QFAAS

Hydride generation hyphenated to atomic absorption spectrophotometry (FIAS 4110 system, Perkin-Elmer) was used for complementary measurements of total selenium and also for the verification of selenium oxidation state. Indeed, when using $NaBH_4$ as reducing agent, Se(IV) can form volatile hydrides. The carrier solution was HCl 10%(v/v). The atomizer was a flame-heated quartz T-shaped tube. A hollow-cathode lamp (Perkin-Elmer) was used as a light source and the wavelength for Se was 196.0 nm. The detection limit obtained in these conditions was 17 ng(Se) l^{-1}.

2.5.3. Ionic chromatography–electrochemical detector

Measurement of major cations (K^+, Na^+, Ca^{2+}, Mg^{2+}) and anions (Cl^-, $NO_3{}^-$, $SO_4{}^{2-}$) at mg l^{-1} levels was performed with a DIONEX DX600 system, equipped with a GP40 pump and a ED40 conductimeter. The anion exchange column (Dionex AS 17) and the cation exchange column (Dionex CS12A) were purchased from DIONEX. Cations separation was performed using methanesulfonic acid (MSA, 18 mM) as the mobile phase at 1 ml min^{-1} flow rate in isocratic mode. Anions separation was carried out using KOH at 1 ml min^{-1} flow rate as the mobile phase and a linear elution gradient (from 1 up to 20 mM). Acquisition times were 35 and 20 min, respectively for cations and anions separation.

2.5.4. Gamma spectrometry

Measurements of ^{75}Se at 265 keV (59.1% I.E) were performed with a n-type Be detector from ORTEC (GMX-15185). The detector relative efficiency was 17% and the peak resolution at 265 keV was 1 keV.

3. Results and discussion

3.1. Mineralization

A microwave digestion in nitric acid medium has already been described in the literature for selenium mineralization in different matrices such as human plasma [17], coal, fly ashes, sorbents [18] or food samples [19]. This digestion, suitable for a large variety of samples, was chosen for waste applications. The mineralization had to be tested on a representative complex sample, so a glass–fiber filter (rich-silicium matrix) was chosen. The protocol described in Section 2.4. was applied but the sample was spiked with higher selenium concentration (5 μg of Se(IV) or Se(VI)) in order to allow a sample dilution for a direct measurement by ICP-MS or HG-QFAAS. The first tests showed that the sample dissolution was incomplete. Then, a reagent mixture (2/3 HNO_3, 1/3 HF) was used and allowed the dissolution to be complete. The remaining solution was totally removed and diluted for measurements by ICP-MS and HG-AAS to determine the selenium total concentration and its oxidation state, respectively. The amounts measured after mineralization were (4.80 ± 0.15) μg by ICP-MS and (4.59 ± 0.10) μg by HG-QFAAS, respectively. These results have showed that 90% of initial Se(IV) was present in the mineralized solution and more than 92% of total selenium remains under +IV form.

3.2. Separation

3.2.1. Evaporation/concentration

Some physico-chemical forms of selenium can be volatile, for example under oxidation degree II as $H_2Se(g)$ [20] or oxidation degree +IV in hydrochloric acid medium [21]. For this reason, it was important to check the stability of Se(IV) during an eventual evaporation step which could be necessary to concentrate the sample and/or to remove volatile halide compounds.

According to previous works described in the literature [22], tests were made on hot plate at low temperature ($T = 150\,^{\circ}C$, open Teflon vial) in order to evaporate the sample as slowly as possible. They were carried out with 1 μg of Se(IV) prepared in 10ml of concentrated HNO_3 to simulate the solution obtained after the mineralization step. After evaporation (4–5 h), the remaining solution was removed in 1 or 2 ml of HNO_3 0.1 mol l^{-1}. The yields obtained for selenium in various media were evaluated after ICP-MS measurement and are presented in Table 1. Selenium yield is higher than 90% even in samples presenting a high salinity. A measurement by HG-AAS also showed that selenium oxidation degree did not change. Moreover, measurements of Br concentrations showed that 95 to 98% of the initial content were removed as $Br_2(g)$ during the evaporation step.

3.2.2. Decrease of total activity and salinity

In order to decrease total activity and salinity, a separation on cationic exchange resin was carried out on AG50-X8 column. Cationic forms have a higher affinity for R-SO3$^-$ functional groups than selenium which is under selenic acid form ($H_2SeO_3/HSeO_3^-$, p$K_a = 2.68$) in HNO_3 0.1 mol l^{-1} medium and thus is not retained on AG$^{®}$50 resin. According to the relative selectivity of various counterions given by the manufacturer and the literature [23], the separation was performed with diluted HNO_3 as eluent. Different concentrations of eluent (0.1 to 0.5 mol l^{-1}), amounts of resin (2 to 10 g), flow rates (without regulation, 0.5 and 1 ml min^{-1}), were tested with a solution containing 500 ng of Se(IV) and 5 μg of Cs, Co and Sb and an important salinity (5 g l^{-1} of NaCl). The results showed that a simple elution by gravity induces a co-elution of the species whatever the mass of resin or eluent concentration is. A regulation of flow rate is then necessary to allow the chemical equilibriums to be established in the column. Moreover, in these conditions, the concentration of eluent has no influence on Se(IV) elution profile, which confirms the very low affinity of selenium for the stationary phase. After optimisation, the following conditions were chosen: sample volume: 2 ml, mass of resin: 10 g, eluent: HNO_3 0.1 mol l^{-1}, flow rate: 0.5 ml min^{-1}. In these conditions, 90% of initial selenium content is recovered in a 12 ml fraction. Only 0.02% of Co, 0.01% of Cs and 0.5% of Sb are present in this fraction.

Table 1
Selenium yield after evaporation step in nitric acid (3 mol l^{-1}) and in various conditions (%) (mean on three experiments ± standard deviation)

	Solution composition	Se(IV) yield (%)
1	1 μg Se(IV)	95 ± 3
2	1 μg Se(IV) + 1 μg Br$^-$	93 ± 4
3	1 μg Se(IV) + 1 μg Br$^-$ + 500 μg NaCl	94 ± 3
4	1 μg Se(IV) + 1 μg Br$^-$ + 1 mg NaCl	94 ± 4

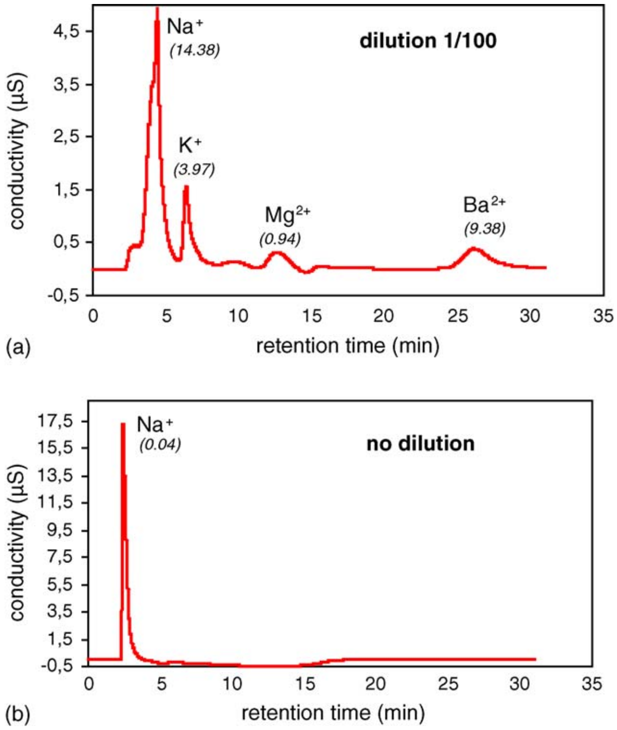

Fig. 1. Typical chromatograms obtained by cationic chromatography before and after step no. 3 for the glass filter sample: (a) before cationic exchange resin; (b) after cationic exchange resin-concentrations in mg (cation) l^{-1}.

In order to confirm the decrease of salinity with a “real sample”, the protocol was also applied to a glass–fiber filter spiked with 500 ng of Se(IV) (after mineralization and evaporation). The major cations (Na^+, K^+, Ca^{2+}, Mg^{2+}) were quantified before and after separation. The results presented on chromatograms (Fig. 1) show an important decrease of salinity after separation.

3.2.3. Separation from isobaric interferent ^{79}Br

The ultimate step of the separation aims at eliminating species, which can induce interferences during ICP-MS measurement, particularly the main isobaric interferent ^{79}Br. According to the previous results, more than 90% of bromide ions initially present in the solution could be removed as $Br_2(g)$ during the evaporation step. However, a complementary step might be added in order to eliminate bromide compounds, even at trace level. Two methods were tested to separate selenium from Br species: (i) precipitation of silver halides and (ii) separation on anion exchange resin.

Due to their low solubility in diluted nitric acid media [24], silver halides were precipitated with $AgNO_3$. The reaction was performed in HNO_3 2% by adding some excess of $AgNO_3$ 2% to different amounts of Se(IV), Br$^-$ and a fixed amount of Cl$^-$ (about 300 mg), used as a carrier. After stirring, the solution was centrifuged and the supernatant was measured by ICP-MS. Results are presented in Table 2. Selenium was not

Table 2
Selenium and bromide yields (%) in the supernatant after halides precipitation by $AgNO_3$ in HNO_3 2% medium (mean on three experiments ± standard deviation)

Initial amount (ng)	% in the supernatant	
Se (IV) and Br^-	Se (IV)	Br^-
20	99.0 ± 0.5	5.0 ± 1.0
500	99.0 ± 0.5	0.3 ± 0.2

co-precipitated with silver halides as 99% of the initial amount was removed in the supernatant in both tested conditions. This step allows eliminating more than 99% of Br^- in case of high initial amounts but only 95% if Br^- was present at trace level. According to these results, the precipitation step appeared not to be sufficient to get rid of bromide compounds. Even if the precipitation reaction might be added to the protocol in case of rich halides samples, a complementary step is necessary to remove traces of Br compounds in view of an ICP-MS measurement.

Tests for the separation of selenium and bromide at trace level were performed with an AG® 1-X4 anion exchange resin by using the same eluent than during cation exchange step (HNO_3). The operating conditions were optimised: the amount of resin, the concentration of eluent, the flow rate and the volume of sample. Three typical elution profiles obtained in different conditions are presented in Fig. 2. More than 90% of selenium is eluted in the first 30 ml in case (a) and in the first 40 ml in case (b). The increase of stationary phase amount allows to improve the resolution but these conditions do not allow a complete separation. More than 65% of Br^- is co eluted with selenium in case (a) and 30% in case (b). So, a resin amount of 10 g was chosen and the influence of eluent concentration was then tested. According to the manufacturer values, the relative selectivity of $NO_3{}^-$ ($k' = 65$) is higher than Br^- one ($k' = 50$). When the eluent concentration increases, the number of sites taken up by $NO_3{}^-$ is higher, this induces a faster elution of anionic species with a lower affinity for the resin. A concentration of 0.1 mol l^{-1} allows a satisfactory separation of Se(IV) and Br^-. More than 80% of selenium is recovered in a 25 ml fraction which contains less than 1% of initial bromide. The decrease of flow rate down to 0.5 ml min^{-1} do not allow to improve the separation between both compounds.

According to the obtained results, a radiochemical protocol was proposed (Fig. 3) including five steps. At the end of the protocol, a 25 ml fraction was finally removed in HNO_3 0.1 mol l^{-1} for measurement by ICP-MS and/or gamma spectrometry.

3.3. Application to spiked waste samples

The radiochemical procedure was applied to two types of matrices which could be selected during controls of waste packages: a glass–fiber filter (rich silicium sample) and an ion exchange resin, both spiked with natural selenium.

The yields obtained for selenium were evaluated at different stages of the separation by ICP-MS measurement. The results are given in Table 3. The global yields obtained are 83 and 84%, respectively for the filter and the resin, so they confirm the efficiency of the extraction. A loss of 14% can be noted between steps 1 and 5 and about 9% between steps 3 and 5.

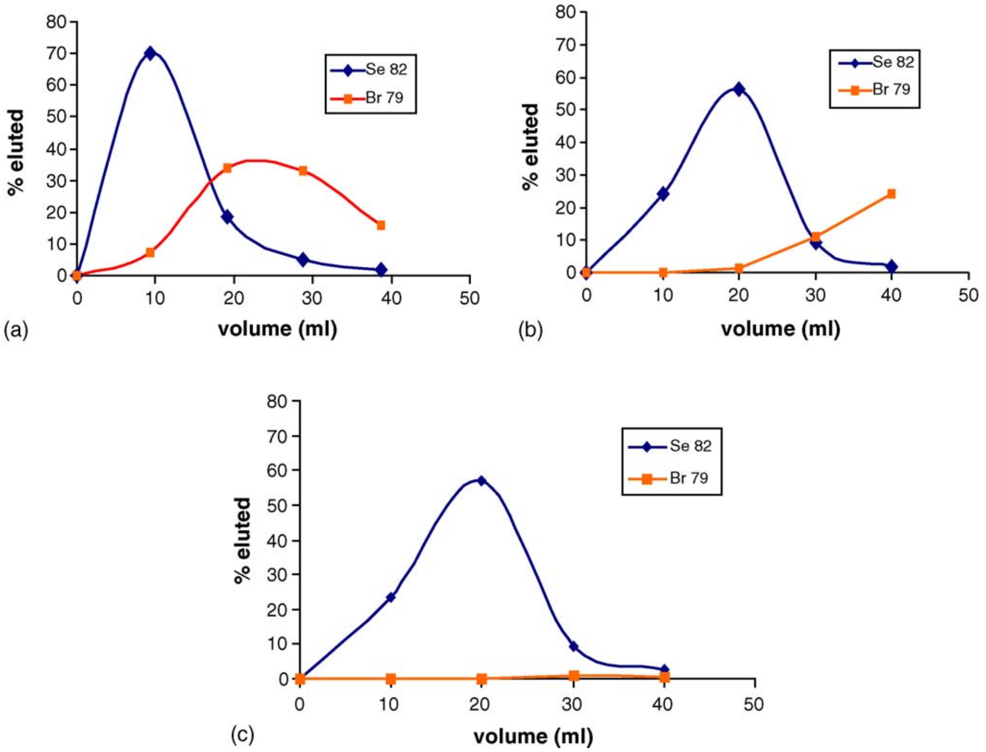

Fig. 2. Elution profiles of Se(IV) and Br^- on anion exchange resin (AG1-X4) in the following conditions: sample volume, 2 ml; initial amount, 500 ng of Se(IV) and Br^-; flow rate, 1 ml min^{-1}; (a) mass of resin, 5 g; eluent, HNO_3 0.1 mol l^{-1}; (b) resin mass, 10 g; eluent, HNO_3 0.3 mol l^{-1}; (c) resin mass, 10 g; eluent, HNO_3 0.1 mol l^{-1}.

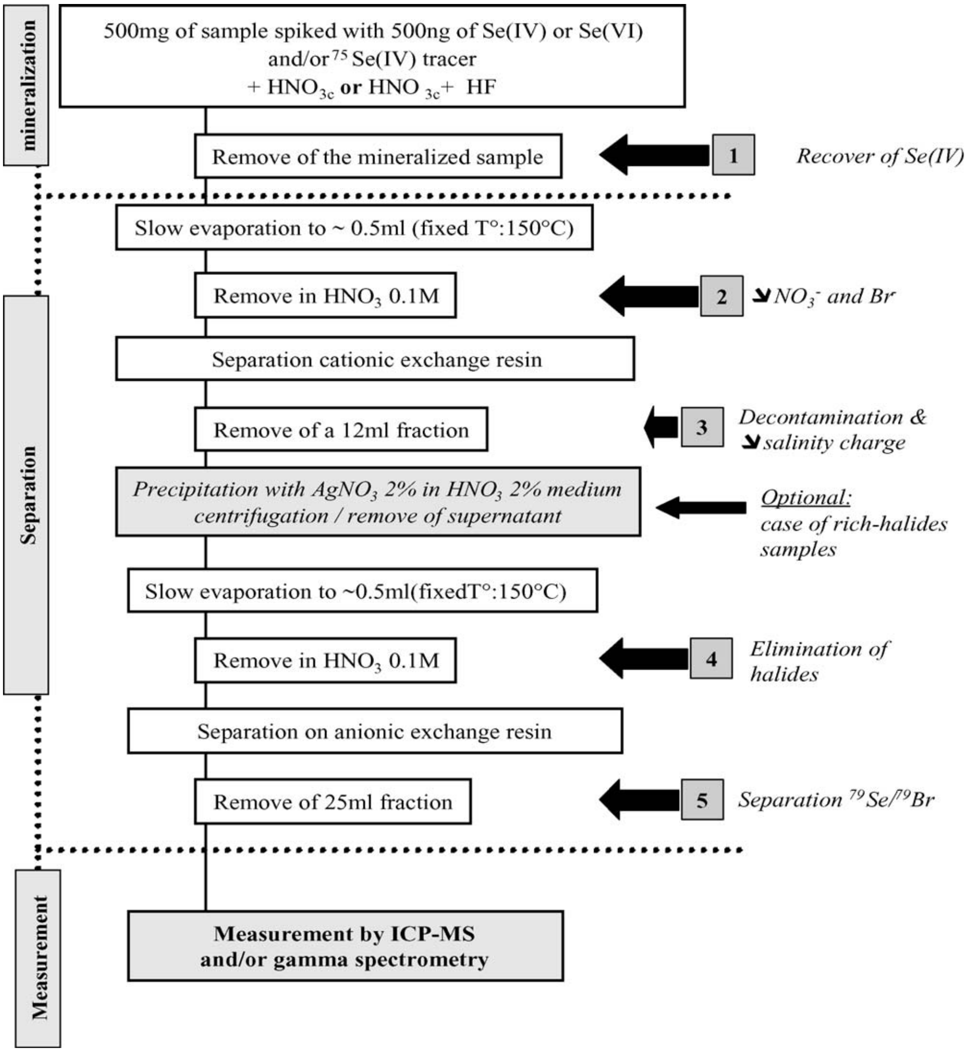

Fig. 3. Scheme of the optimised analytical procedure: mineralization, separation and measurement.

Typical ICP-MS spectra are also presented in Fig. 4. They were obtained for the spiked glass–fiber filter sample at different stages of the separation. A residual presence of Br, detected at the mass 79 and 81 can be noted after step 2. The concentration assessed on mass 79 was about 25 $\mu g\,l^{-1}$. At the end of the protocol, the isobaric interferent is removed and mass 79 is free of any interferences which confirms the efficiency of the separation. Moreover, in case of an eventual presence of Br at trace level in the final sample, its quantitation could be checked on mass 81 in order to correct the measurement on mass 79. Then,

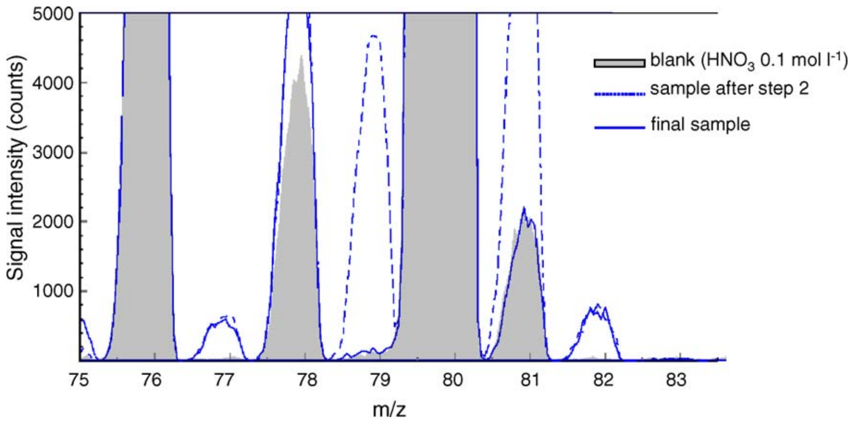

Fig. 4. Typical spectra obtained by ICP-MS for a glass–fiber filter spiked with Se(IV) at different stages of the separation procedure.

Table 3
Application to two samples spiked with 500 ng of natural Se(IV)

	Yield ± standard deviation (%)	
	Glass–fiber filter	Ion exchange resin
Step 2	–	98 ± 1
Step 3	92 ± 3	–
Step 5	83 ± 3	84 ± 1

Selenium yields (%) after the different steps of the separation (mean on three experiments ± standard deviation).

Table 4
Application to a synthetic solution of ^{75}Se and an effluent spiked with ^{75}Se

	Activity ± standard deviation (Bq)	
	Synthetic solution	Effluent
Initial activity	65.8 ± 9.7	71.1 ± 7.8
Step 3	71.7 ± 3.4	58.6 ± 2.8
Step 5	63.0 ± 3.0	50.2 ± 2.5

Activities after the different steps of the separation.

this complementary information allows to confirm the accuracy of the result. In these conditions, the measurement of selenium 79 appears to be feasible as the radiochemical procedure allows the total elimination of the isobaric interferent.

At last, the protocol was tested with two samples spiked with the gamma emitter ^{75}Se, used as a radioactive tracer: a synthetic solution (^{75}Se prepared in HNO_3 0.1 mol l^{-1}) and an effluent obtained from a waste treatment plant. The main objective was to check the behaviour of selenium as a radionuclide during the different steps of the procedure and verify the adequation with stable isotope. The ^{75}Se activities are given in Table 4 at different stages of the radiochemical protocol. Final yield was about 95% for the synthetic solution and about 71% for the effluent. According to these results, the yields appear to be dependant on the complexity of the matrix. In case of the effluent, the solid content was estimated to 3.78 g l^{-1}, which can induce difficulties and losses during the manipulation (e.g. high salinity during evaporation or ion exchange steps).

4. Conclusion

A radiochemical process was proposed for the determination of ^{79}Se in low and intermediate wastes samples by Q-ICP-MS. The protocol was developed on synthetic solutions prepared with natural selenium and/or ^{75}Se used as a radioactive spiker. After a mineralization in a microwave oven, the sample was submitted to a five steps separation procedure. It allows to (i) preconcentrate the sample and eliminate volatile species; (ii) reduce the global activity and the salinity by cation exchange separation; (iii) eliminate isobaric interferent ^{79}Br by precipitation and/or anion exchange resin. The protocol was applied to non active samples. The yields obtained were ranged from 70 up to 80% according to the matrix and the ICP-MS measurement showed that mass 79 was free of any interference after separation.

The validation of the radiochemical protocol is now carried out on active samples spiked with selenium which can present an initial global activity up to 10^5–10^6 Bq g^{-1}: effluent, concentrate from distillation process, technological wastes. The first results obtained on these types of samples show good yields for selenium (from 60 up to 80%). Decontamination factors about 11,000 can be reached for samples with an initial global activity of 10^3–10^4 Bq g^{-1}. The elimination of the radionuclides which generate the main activity (ex: ^{60}Co, ^{137}Cs, ^{241}Am, ^{90}Sr) is achieved, except for ^{125}Sb (co extracted with selenium during the different steps). As a consequence, investigations are needed in view of a LSC measurement. The addition of a complementary step allowing a complete purification is under study. These results will be the subject of a next paper about the validation of the protocol on ILW samples.

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