Determination of $^{210}$Po and uranium in high salinity water samples

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Received: 14 July 2010 / Published online: 19 August 2010 © The Author(s) 2010. This article is published with open access at Springerlink.com

Abstract A method for the determination of uranium and $^{210}$Po in high salinity water samples has been elaborated. Both radionuclides are preconcentrated from 0.5 dm$^3$ saline media by co-precipitation with hydrated manganese dioxide, followed by dissolution of the precipitate in 200 mL of 1 M HCl. Uranium isotopes $^{235}$U and $^{238}$U can be directly determined by ICP MS method with a detection limit of 0.01 ppb for $^{238}$U. Prior to a selective determination of $^{210}$Po, the majority of other naturally occurring $\alpha$-emitting radionuclides (uranium, thorium and protactinium) can be stripped from this solution by their extraction with a 50% solution of HDEHP in toluene. Finally, $^{210}$Po is simply separated by direct transfer to an extractive scintillator containing 5% of trioctylphosphine oxide in Ultima Gold F cocktail and determined by an $\alpha/\beta$ separation liquid scintillation technique with detection limit below 0.1 mBq/dm$^3$.

Keywords $^{210}$Po · Uranium radionuclide · Liquid scintillation counting with $\alpha/\beta$ separation

Introduction

According to the WHO protocol for radioactivity monitoring in drinking waters, in the first step the total $\alpha$ and $\beta$ activities are determined [1]. Gross $\alpha$, $\beta$ and $^{226}$Ra activity can be simply appraised by direct adding of the water samples to scintillation cocktails followed by a simultaneous counting of $\alpha$ and $\beta$ particles in the liquid scintillation devices with an $\alpha/\beta$ separation option [2]. However, if the total $\alpha$ activity exceeds 0.5 Bq/dm$^3$, the activities of the particular $\alpha$ emitters should be measured. In practice, for natural water it concerns the radionuclides of radium, uranium, $^{210}$Po and $^{226}$Th. Since thorium compounds are generally poorly soluble in water, the activities of $^{232}$Th and $^{230}$Th radioisotopes are extremely low in the order of $\mu$Bq/dm$^3$, and only the activity of $^{228}$Th is observable, due to the contribution of $^{226}$Ra decay to its total budget in natural water [3].

A number of analytical methods are available for the detection of radium isotopes [4]. However, liquid scintillation counting is often used for fast and reliable determination of radium and $^{222}$Rn determination in different kinds of natural water [5–8].

Military use of depleted uranium in wars in the Middle East, Afghanistan and the Balkans, and its environmental and human health impact [9, 10] are reasons for an increasing interest in the fast and routine methods for uranium determination in environmental samples. On the other hand, a terrorist poisoning by using $^{210}$Po drew attention to its high radiotoxicity, even in extremely low concentrations in water solutions [11].

Average $^{238}$U and $^{234}$U activities in surface and ground water are in the range of 1–20 mBq/dm$^3$, which corresponds to a concentration of elemental uranium from 0.1 to 1.5 $\mu$g/dm$^3$ [12]. These two uranium radionuclides in natural water are not often in a radioactive equilibrium. A major source of uranium is seawater, where its average concentration is equal to 3.4 $\mu$g/dm$^3$ [13]. However, higher concentrations of uranium are also detected in the drinking water in different countries, for example up to 150 Bq/dm$^3$ in some private wells in Finland [14].

$^{210}$Po radionuclide in a water environment occurs usually in much lower activities <1 Bq/dm$^3$. Typical activity
concentrations of $^{210}$Po in surface coastal seawater are in the range of 0.06–0.84 mBq/dm$^3$ [15, 16].

According to WHO recommendation, taking into account uranium chemical toxicity, its level in drinking water should not exceed 2 μg/dm$^3$ [1]. A more tolerable concentration of 30 μg/dm$^3$ for uranium is proposed by the US EPA [17]. Both these organizations recommend close values of permissible activity concentrations of $^{210}$Po in drinking water: 0.1 and 0.148 Bq/dm$^3$, respectively, which are among the lowest limits for any natural or anthropogenic radionuclides in drinking water.

Uranium can be measured in diverse matrices by radiometric, laser fluorimetry and mass spectrometric methods. Advances in the methods of uranium determination have been recently overviewed [18, 19]. There is no doubt that in recent years inductively coupled mass spectrometry (ICP MS) has been increasingly used for the accurate determination of trace levels of long-lived radionuclides, including uranium and even radium isotopes [20]. Thanks to the very low detection limit of the ICP MS method (<0.01 ppb), the majority of water samples can be instrumentally measured directly without any pretreatment. However, an important limitation of this method is the fact that the concentration of the total dissolved solids (TDS) present in the analyzed solution cannot exceed 0.1% (1 g/dm$^3$) [21]. Unfortunately, in some kinds of environmental samples such as seawater, geothermal or natural mineral drinking water, the TDS values are often higher than 1 g/dm$^3$.

$^{210}$Po is an $\alpha$-emitter with weak gamma-rays abundance (803.1 keV – 0.0011%) and can be measured by $\alpha$-spectrometry with solid detectors or by liquid scintillation counting (LSC) with $\alpha/\beta$ separation using special extractive scintillation cocktails [22]. Usually a simple separation of $^{210}$Po from other interfering radionuclides present in mineral and drinking water is achieved by its spontaneous deposition on silver discs. Such disks can be measured by an $\alpha$-spectrometry systems with 30% efficiency and a very low background < $10^{-4}$ cps [23–26]. The alternative method, LSC allows 100% detection efficiency to be achieved, but the background is higher (>10$^{-3}$ cps) and the resulting limits of detection are also slightly higher [27].

The aim of this study was to develop a fast method for routine determination of uranium and $^{210}$Po radionuclides in high salinity water samples (mineral, geothermal and seawater) after its preconcentration on hydrated manganese dioxide. It is well known that in such media uranium occurs mainly as a UO$_2$(CO$_3$)$_{2-}$ anions [28], which are quantitatively scavenged by precipitated manganese dioxide [29]. Sorption of $^{210}$Po from seawater by this procedure is also well documented [30]. After dissolving the obtained precipitate in a sufficient volume of 1 M HCl to get the final TDS < 1 g/dm$^3$, one part of solution can be used for direct determination of uranium by ICP MS, whereas from the second portion of solution $^{210}$Po can be transferred to a silver disc deposition device or to an extractive scintillation solution after removal of the other interfering $\alpha$-emitting radionuclides.

### Experimental

#### Materials and method

A standard solution of $^{210}$Pb in equilibrium with $^{210}$Bi and $^{210}$Po was purchased from Amersham (RBZ.44). A secondary uranium standard (238U), 200 ppm, was prepared by dissolution of 0.422 g of uranyl nitrate (UO$_2$)(NO$_3$)$_2$·6H$_2$O (p.o. Chemapol) in 1 dm$^3$ of high purity deionizing water. All other chemicals used in this work were analytical grade purity compounds.

The standard reference material, Irish Sea Water: IAEA-381—“Radionuclides in seawater”, was supplied by the International Atomic Energy Agency (IAEA). For determination of the $^{210}$Po and $^{238}$U radionuclide recoveries from high salinity water, the artificial seawater solution was prepared by dissolving 27.2 g NaCl, 3 g MgSO$_4$, 2.65 g MgCl$_2$, 1.1 g CaCl$_2$ and 0.755 g KCl in 5 dm$^3$ of high purity deionizing water. Bottled mineral water with a high salt content were purchased from local stores.

#### Co-precipitation of $^{210}$Po and uranium from saline solutions

Simultaneous co-precipitation of $^{210}$Po and uranium from 0.5 dm$^3$ solutions was carried out by a slightly modified method, described elsewhere [26, 31, 32]. The artificial seawater solution was acidified to pH = 1 by concentrated nitric acid, and 2.5 mL of 0.1 M diethylenetriaminepentaacetic acid (DTPA) was added to form complexes of Ca$^{2+}$ and Mg$^{2+}$ ions followed by a further adding of 7 mL of 0.2 M solution of potassium permanganate with intense 1 h mixing. After this time the solution was alkali by concentrated ammonia, and 2.5 mL of 0.3 M manganese chloride was added. The mixture was heated for 1 h and left for overnight precipitation. The precipitate was filtered, dried and weighed. The addition of DTPA almost completely prevents the precipitation of calcium and magnesium hydroxides from saline solutions. Therefore, the total mass of dried manganese hydroxides precipitated from different water solutions, even those with a high salt content, varied in the narrow range from 0.1 to 0.2 g.

The 0.5 L of artificial seawater was spiked with uranyl nitrate and $^{210}$Po markers for the determination of their recoveries after the preconcentration step. For these purposes a hydrated manganese oxide precipitate with
preconcentrated radionuclides was dissolved in 5 mL of 1 M HCl. One mL of this solution was taken for polonium recovery determination and the remaining 4 mL of solution was diluted to 200 mL for uranium recovery determination by ICP-MS.

Instruments

Uranium determination

Elemental uranium concentration was routinely measured by ICP MS method using a Thermo Electron Corporation X-Series device. For analytical purposes the device was calibrated with a set of secondary standard uranium solutions covering the range up to 100 ppb, prepared from the Merck standard uranium solution.

Radioactivity measurements

The separation efficiency of U, Th and Pa radionuclides from 210Po by 50% HDEHP (bis(2-ethylhexyl)hydrogen phosphate) in toluene extraction was checked by measurement of 234Th, 234Pa, 235U and 238U radionuclides in organic phase by HPGe γ spectrometry system described elsewhere [32].

The activity of 210Po transferred to an extractive scintillator was measured using a BetaScout counter (Perkin Elmer). The optimal counting parameters for α-particle measurement were settled after a preliminary 1-h count. The background in this region was equal to 0.2 cpm and the standard counting time was 80,000 s.

Quality assurance of the elaborated method

The accuracy of the developed method for uranium was evaluated for the standard reference materials IAEA-381 Irish Sea Water. The results for uranium concentration are presented in Table 1. The results of 210Po determination in the standard reference materials are also included although there are no reference values for this radionuclide.

Therefore, the accuracy of the method for 210Po determination was checked by measuring 210Po in the water samples supplied by IAEA in the frames of their intercomparison studies. Five different water samples with 210Po were provided by IAEA of Seibersdorf (Vienna). In the samples 01–04 210Po activities were in the range of 1 to 10 Bq/dm³, whereas for sample #05 the 210Po concentration activity was <0.1 Bq/dm³. The results of these measurements are presented in Table 2.

Results and discussion

Choice of the optimal condition for preconcentration of uranium prior to its determination by ICP-MS

In order to avoid an additional precipitation of calcium and manganese hydroxides, during the co-precipitation procedure with manganese hydrated oxide, two well-known complexing agents, EDTA and DTPA, were checked. The results of the uranium recoveries from artificial sea water with and without use of these complexing agents are presented in Table 3.

Using 5 mL of 0.1 M EDTA is connected with a decrease of uranium recovery in comparison to untreated sea water, while the addition of 2.5 mL of 0.1 M DTPA increases this recovery to 84.7%. Simultaneously, a reduction of the total amount of precipitate to 0.2 g was observed. Therefore, in the standard procedure for high saline water samples, the DTPA solution was used as a masking agent for Ca⁺² and Mg⁺² ions.

In order to achieve the final solution with TDS below 1 d/dm³, the hydrated manganese dioxide (0.2 g) with adsorbed radionuclides should be dissolved in 200 mL of

| Volume of complexing agent (mL) | Mass of precipitate (g) | Recovery (%) |
|-------------------------------|------------------------|--------------|
| 0.1 M EDTA                    | 0.4690                 | 72.8 ± 0.3   |
| 2                             | 0.0789                 | 48.0 ± 0.2   |
| 3                             | 0.2018                 | 84.7 ± 0.3   |
| 4                             | 0.1408                 | 59.1 ± 0.2   |
1 M HCl with 3% H$_2$O$_2$. A portion of this solution was taken for direct uranium determination by ICP-MS, while the rest was used for $^{210}$Po determination.

Removing of U, Th, Pa and $^{210}$Po from 1 M HCl solution by HDEHP in toluene

The standard solution containing 303 μg/mL UO$_2$(NO$_3$)$_2$ with $^{234}$Th and $^{234}$Pa radionuclides in a radioactive equilibrium was prepared for U, Th and Pa separation studies from $^{210}$Po. The thorium, uranium and protactinium nuclides were extracted from 20 mL and 100 mL samples of this solution by twofold extraction of 10 mL 50% solution HDEHP in toluene. The results of these experiments are shown in the Table 4.

As is evident from Table 4, all radionuclides of Th, U and Pa are completely removed from the solution containing $^{210}$Po, however, the $^{210}$Po remains almost quantitatively in the water phase.

Choice of optimal beta scout parameters for $^{210}$Po determination by LSC with an extractive scintillator

In order to ensure optimal counting conditions for complete separation of $\alpha$ from $\beta$ pulses in the BetaScout device, the total radioactivity in scintillator solution should be checked first. On the basis of the 3D spectrum, the proper PLI (Pulse Length Index) parameter of 440 for $^{210}$Po was settled on (see Fig. 1).

After twofold extraction of $^{210}$Po from dissolved manganese precipitate, the total volume of extractive scintillator rose to 20 mL. Therefore, the influence of the scintillator volume on the detection efficiency of $^{210}$Po in the Beta Scout device was also checked. The results of these measurements showed that the increase of the volume of scintillation solution does not influence detection efficiency, and the two-step extraction by 10 mL of an extractive scintillator ensures the complete transfer of $^{210}$Po.

**Table 4** Removing of the interfering radionuclides after two-fold extraction by 10 mL of 50% (w/v) HDEHP in toluene solution

| Radionuclide | Yield of extraction from 20 mL solution (%) | Yield of extraction from 100 mL solution (%) |
|--------------|------------------------------------------|------------------------------------------|
| $^{234}$Th   | 95.7 ± 0.1                                | 98.6 ± 0.1                                |
| $^{235}$U    | 100.5 ± 3.4                               | 100.3 ± 1.0                               |
| $^{235}$U + $^{226}$Ra | 100.1 ± 0.2                           | 100.3 ± 0.1                               |
| $^{234}$Pa   | 98.8 ± 23.6                               | 100.1 ± 3.7                               |
| $^{210}$Po   | 0.191 ± 0.001                             | 0.037 ± 0.001                             |

Fig. 1 Establishing of the optimal setting parameters of BetaScout device for $^{210}$Po determination

**Table 5** $^{210}$Po extraction into 5% solution of trioctylphosphine oxide in Ultima Gold F

| Lp. | Step                                      | Recovery of $^{210}$Po (%) |
|-----|------------------------------------------|----------------------------|
| 1   | First extraction from 0.1 mL of $^{210}$Po in 100 mL 1 M HCl | 89.2 ± 0.1                 |
| 2   | Second consecutive extraction from the same solution | 99.4 ± 0.1                 |
Diagram scheme of the elaborated procedure for radio-nuclide determination in the high salinity water samples is shown on Fig 2.

**Table 6** Activity of $^{210}$Po and uranium in mineral water samples

| Mineral water samples | TDS (mg/dm$^3$) | Activity of $^{210}$Po (mBq/dm$^3$) | Activity of $^{238}$U (mBq/dm$^3$) | Reported $^{238}$U activity (mBq/dm$^3$) |
|-----------------------|-----------------|-------------------------------------|-------------------------------------|----------------------------------------|
|                       |                 | After extraction                     | After deposition                    |                                        |
| Kropla Beskidu        | 386.6           | 10.16 ± 1.01                        | 10.00 ± 2.09$^a$                    | 3.30 ± 0.27                            | 2.80 ± 0.4 [7]; 1.1 ± 0.2 [34]       |
| Ustronianka           | 423.32          | 8.60 ± 1.06                         | 8.36 ± 1.66                         | na                                     | 10.54 ± 0.76 [7]                     |
| Multi Vita            | 812             | 5.60 ± 0.85                         | 5.71 ± 1.71                         | 1.39 ± 0.21                            | 19.2 ± 3.5 [7]; 0.6 ± 0.2 [34]; 0.49 ± 0.01 [26] |
| Wielka Pieniawa       | 1,394.7         | 11.16 ± 1.04                        | 12.38 ± 2.18                        | 5.57 ± 0.45                            | 4.40 ± 1.2 [7]                       |
| Piwniczanka           | 2,314.49        | 4.75 ± 0.82                         | 4.60 ± 1.81                         | na                                     | 2.20 ± 0.2 [7]; 1.4 ± 0.2 [34]       |
| Staropolanka 2000     | 2537.2          | 13.84 ± 1.10                        | 13.65 ± 2.23                        | 7.25 ± 0.61                            | 7.68 ± 0.9 [7]                       |

$^a$ Uncertainties were calculated on the base of one standard deviation of activity measurements

Po and uranium in mineral water

The method has been used for the determination of the $^{238}$U and $^{210}$Po activities in some bottled mineral water samples distributed in Poland. The concentration of uranium in the majority of Polish mineral water samples was reported previously [7, 26, 34], while $^{210}$Po was determined only in a few types of bottled mineral water [26].

In our experiments, in three kinds of mineral water TDS exceeded 1 g/dm$^3$, and in the remaining three samples mineralization was lower. Our results (see Table 6) are generally in sufficient accordance with the previous reported data for these mineral water samples.
Conclusions

The elaborated method for $^{238}\text{U}$ and $^{210}\text{Po}$ determination consists on co-precipitation with hydrated manganese dioxide. Uranium radionuclides can be measured by ICP-MS technique after reducing the total TDS below 1 g/dm$^3$. Uranium recovery, even from high salinity water samples, averaged $\sim 86\%$. $^{210}\text{Po}$ was extracted by 5% (w/v) solution TOPO in scintillation cocktail Ultima Gold F, after removing all interfering $\alpha$-emitting radionuclides by two-fold extraction with 50% solution of HDEHP in toluene. The obtained detection limit for a routine liquid scintillation counting of $^{210}\text{Po}$ is equal to 1 mBq/dm$^3$, and is sufficient for determining the natural levels of this radionuclide in the majority of natural drinking water samples.

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