STANDARTIZATION OF THE RADIOCHEMICAL PROCEDURE OF POLONIUM - 210 DETERMINATION IN WATER SAMPLES

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Abstract

Naturally occurring radionuclides of terrestrial origin called primordial radionuclides are present in various quantities in the environment, including the human body. Polonium- 210 is one of the most toxic naturally occurring radionuclides and one of the most important environmental radionuclides due to its wide distribution and potential for human radiation exposure through ingestion and inhalation. It is a naturally occurring alpha emitter and exists in the environment, mostly found in water, soil and food because of the Lead-210 decay within the Uranium-238 decay chain. There are limited number of methods available for its determination in water samples, the most used being alpha-particle spectrometry. In this study are performed two procedures for chemical separation of polonium, solvent extraction, and Sr-resin extraction chromatography to determine the activity concentration of 210-Po by alpha-particle spectrometry. The other steps of the procedure including chemical procedures and measurement was the same.

Key words**:** *Polonium-210, alpha particle spectrometry, radiation, decay chain.*

Përmbledhje

Radionuklidet primordiale, të cilat janë radionuklide natyrore me origjinë tokësore, gjenden në sasi të ndryshme në mjedis, duke përfshirë edhe trupin e njeriut. Ndër to, Poloniumi-210 (^{210}Po) është një nga radionuklidet më toksike dhe më të rëndësishme për mjedisin për shkak të shpërndarjes së gjerë dhe potencialit për ekspozim të njeriut ndaj rrezatimit nëpërmjet gëlltitjes dhe frymëmarrjes. Ai është një emetues natyror i grimcave alfa dhe gjendet kryesisht në ujë, tokë dhe ushqim si rezultat i zbërthimit radioaktiv të Plumbit-210 (^{210}Pb) brenda zinxhirit të zbërthimit të Uraniumit-238 (^{238}U). Ekzistojnë

metoda të kufizuara për përcaktimin e ^{210}Po në kampionet e ujit, ku me e përdorura është spektrometria e grimcave alfa. Në këtë studim, u aplikuan dy teknika për ndarjen kimike të poloniumit — ekstraktimi me tretës dhe kromatografia e ekstraktimit me rrëshirë Sr — për të përcaktuar përqëndrimin e aktivitetit të ^{210}Po me anë të spektrometrisë së grimcave alfa. Hapat e tjerë të procedurës, përfshirë përpunimin kimik dhe matjet, ishin të njëjtë për të dyja metodat.

***Fjalë kyçe**: Polonium-210, spektrometria e grimcave alfa, rrezatim, zinxhiri i zbërthimit.*

Introduction

Polonium is a chemical element with 25 known radioactive isotopes, mass numbers of 192 to 218, of which only the 208, 209 and 210 isotopes have half-lives longer than 1 day. Of these three, it is ^{210}Po which is of most interest from an environmental impact viewpoint (Carvalho F et al., 2017), and its measurement is the subject of this paper.

The Lead-210 decay within the Uranium-238 decay chain is shown in Figure 1.

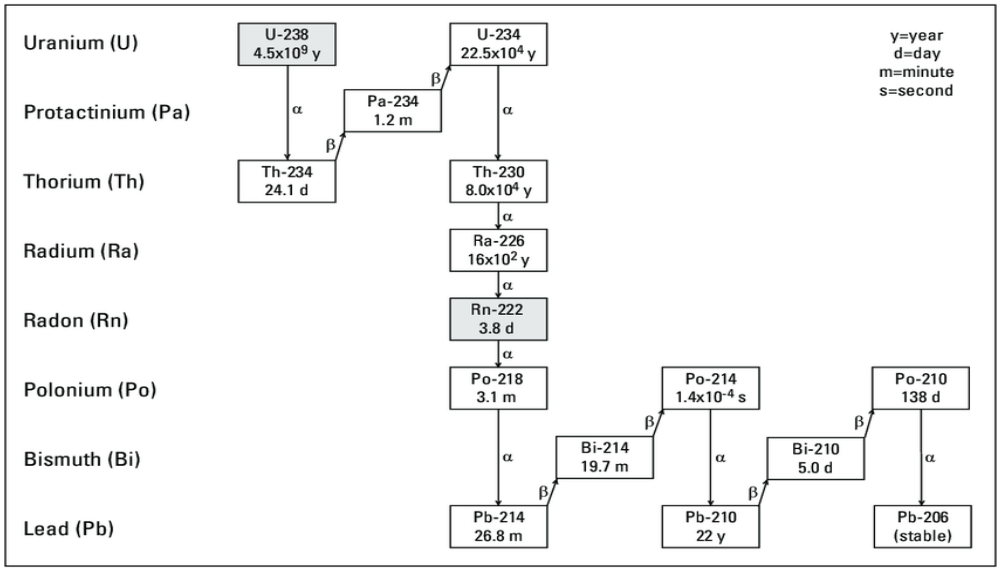

Figure 1. The uranium decay series

In addition, for many sample types (for example, soils and sediments) determination of ^{210}Po is commonly used as a means of determining its progenitor ^{210}Pb. Based on this review, two candidate methods for determination of ^{210}Po in water samples were selected for testing, refinement, and validation. The results of this work are the subject of this paper. For determination of a low-level activity concentration of ^{210}Po in a water sample, it is necessary to handle a large volume of sample. Direct evaporation of water samples has been used for reducing a small volume as a simple process. However, this method is very time consuming for a large amount of a sample (> 1 L).

Therefore, co-precipitation with iron hydroxide, or MnO_2 have commonly been used to preconcentrate Po from water samples. When iron hydroxide is used for preconcentration, it is necessary to remove Fe from the sample solution using a solvent extraction step with an extractant such as di-isopropyl ether, because bulk iron may interfere with the purification of Po using solvent extraction or extraction chromatography as well as in the auto-deposition of Po. In the case that MnO_2 is used, manganese can be removed easily from Po by several alternative chemical separation procedures. Therefore, in this work MnO_2 co-precipitation was selected as the method for preconcentration.

Materials and methods

Two ^{210}Po separation procedures, based on diethyl ammonium diethyldithiocarbamate (DDTC) solvent extraction (DDTC-SE) and extraction chromatography using Sr-resin (Sr-EC), were selected for testing (Chen Q et al., 2001) Reports in the literature indicate that both methods have been successfully used for determination of ^{210}Po in a variety of sample matrices. The method validation of both methods was carried out in terms of trueness, repeatability and reproducibility with tap water spiked with a known amount of ^{210}Po.

This study describes a method for measuring ^{210}Po in freshwater by alpha-particle spectrometry using chemical separation techniques. The method validation was carried out using spiked tap water. The detection limit of the method is 2 mBq L^{-1}, assuming the counting efficiency is 25%, the counting time is 250,000 s and 0.5 L of sample is analysed. Sample volumes up to 10 L can be processed by this method. The method was validated in an activity range of 10 to1200 mBq L^{-1} of ^{210}Po.

The activity concentration of ^{210}Po is determined by alpha-particle spectrometry. The procedure involves the following major steps:

1. Sample preparation, including co-precipitation with MnO_2 in order to concentrate the Po from the bulk sample.

2. Chemical separation and purification of the polonium in order to avoid interference from natural or artificial alpha emitters and stable elements in the test sample.

3. Source preparation by auto-deposition of Po onto a silver disk.

4. Measurement in an alpha spectrometer.

5. Calculation of the analytical result and combined uncertainty.

The chemical separation of polonium is carried out by one of two alternative methods:

(a) DDTC (Diethyl ammonium diethyldithiocarbamate) solvent extraction (Figure 2), or

(b) Sr-resin extraction chromatography (Figure 3).

The chemical separation step improves the reliability of the procedure in terms of both Po recovery and spectrum peak resolution by removing interfering elements present in the sample (E. Kavitha et al., 2017). Auto-deposition onto silver disks is the most used form of source preparation for determination of ^{210}Po by alpha-particle spectrometry as it is simple and ensures separation of Po from other alpha-emitting radionuclides and matrix elements which may not have been completely separated in the chemical separation step. The auto-deposition of Po is also possible onto disks of copper, stainless steel, or nickel but with a drop in deposition efficiency of about 7-8% under the same conditions.

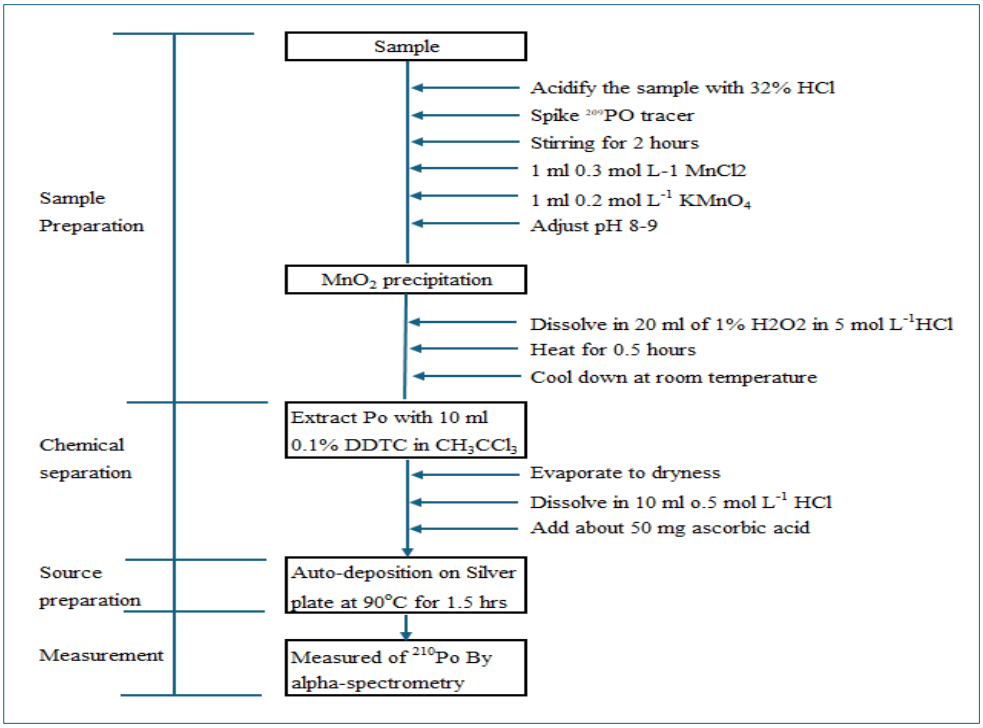

Figure 2. A flow chart for chemical separation of Po by DDTC solvent extraction

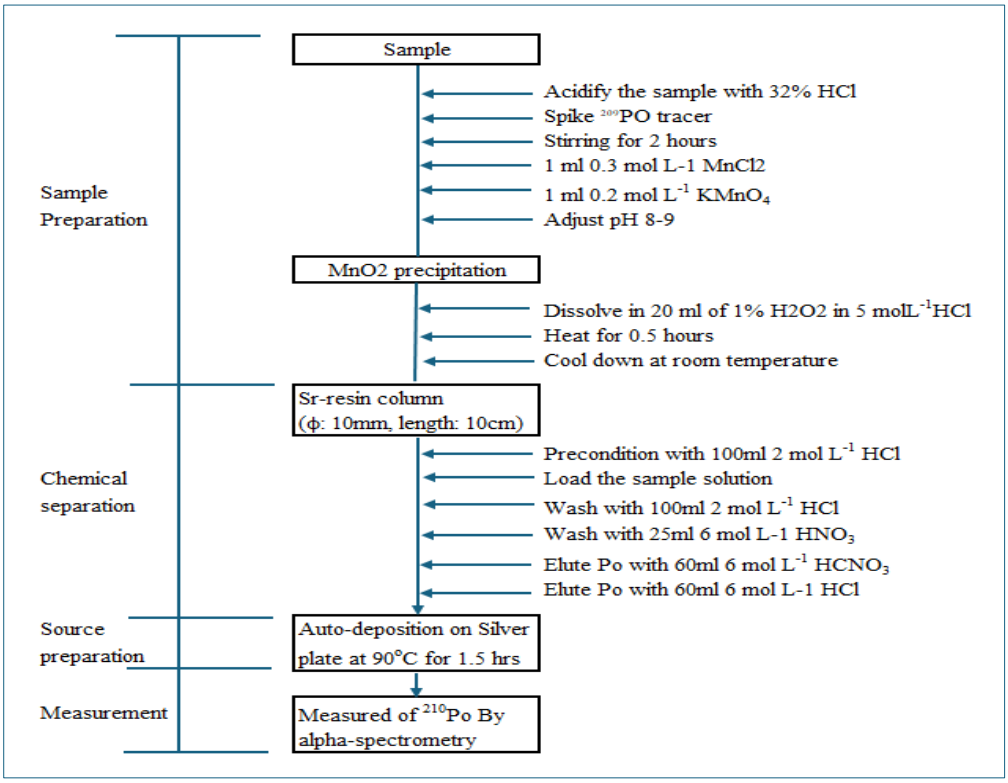

Figure 3. A flow chart for chemical separation of Po by Sr-resin extraction chromatography

The two tracers commonly used for ^{210}Po determination are ^{208}Po and ^{209}Po (Sherrod, et al., 2013). ^{209}Po has a clear advantage over ^{208}Po in energy separation from the ^{210}Po peak and is the preferred tracer, if available. It should also be noted that ^{208}Po solutions normally contain trace activities of ^{209}Po. Unfortunately, the long half-life of ^{209}Po has a disadvantage in relation to detector contamination.

Sources prepared by auto-depositions of polonium are measured in an alfa spectrometer Alpha Analyst Integrated Alfa spectrometer with PIPS (Figure 4).

Calculation of the activity concentration and the combined uncertainty of ^{210}Po on the separation date:

The ratio of the net count rates of the ^{209}Po and ^{210}Po peaks is used to calculate the activity concentration of ^{210}Po in the sample on the date of chemical separation, considering the specific activity of the tracer solution, the volume of the sample and the mass of the tracer solution used, the decay of ^{210}Po between separation and counting, and the decay of the tracer between its calibration date and counting.

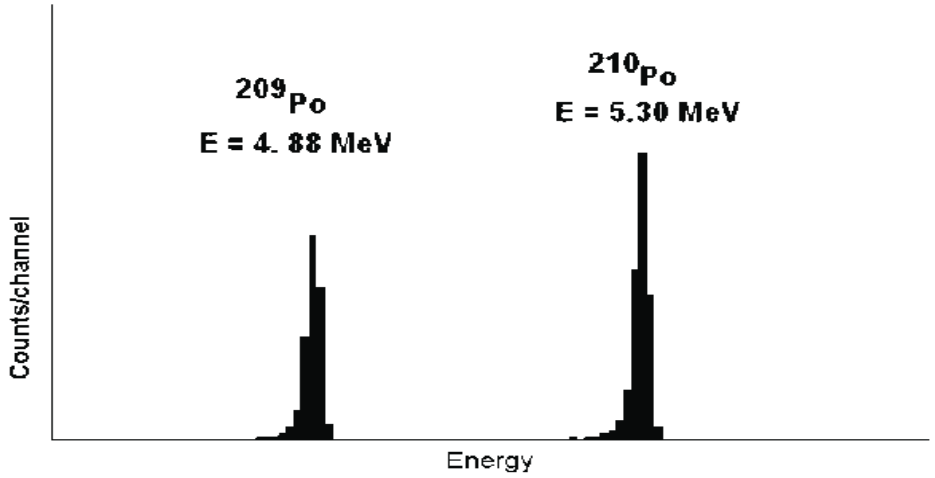

Figure 4. Alpha spectrum for polonium

Conclusions

- The results show that the radiochemical separation performed by two methods were identic. So, they both can be used for the determination of the activity concentration of polonium-210 in water samples.
- The all steps of the procedure must be performed correctly.
- The two picks of ^{209}Po and ^{210}Po are clearly defined with both methods used in this study.

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