PRELIMINARY RESULTS ON THE MEASUREMENT OF PLUTONIUM ISOTOPIC RATIOS AT THE 1MV AMS FACILITY IN IFIN-HH

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The range of radioisotopes measured at the Accelerator Mass Spectrometry (AMS) facility based on the 1MV Tandetron™ of IFIN-HH has been extended to actinides. The paper presents the preliminary results obtained for measuring the isotopic ratios of ^{239}Pu / ^{242}Pu and ^{240}Pu / ^{242}Pu using a secondary standard prepared at CNA Seville.

Keywords: AMS analysis, plutonium isotopic ratios.

1. Introduction

As nuclear techniques have penetrated most of the scientific and technical fields, environmental monitoring has been enriched by detecting long-lived radionuclides, which is essential to track specific nuclear activities such as: operating nuclear reactors, testing weapons, nuclear power stations for reprocessing nuclear fuel, as well as nuclear waste storage [1-5].

After commissioning the 1MV AMS system from IFIN-HH [6], ^{129}I measurements were made to determine the zero-level concentration in the waters of the Danube Delta and the Black Sea [7], and to determine the high radionuclide content in the waters of the west coast of the Pacific (USA) two years after the Fukushima nuclear accident [8]. Recently, tritium measurements probed the contamination caused by nuclear power plants using tree rings, which are natural ^{3}H getters [9].

Investigating the concentrations of plutonium isotopes also represents a highly sensitive tool to track nuclear pollution in the environment (atmosphere, soil, water). In addition, to determine the Pu sources in the environment, due to nuclear activities, measuring the isotopic ratios of plutonium is useful to

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characterize the nuclear fuel used in nuclear power plants [10]. Furthermore, the ^{239}Pu /^{240}Pu ratio provides information with respect to the origin of plutonium and offers the possibility to study the dispersion and mobility of plutonium in the environment [11]. For example, the value of $^{240}Pu/^{239}Pu$ ratio for Chernobyl samples is about 0.56, while the same ratio related to the nuclear weapons is (0.03-0.06) [12].

Measuring the low concentration of radionuclides requires very powerful spectrometric techniques such as inductively coupled plasma mass spectrometry (ICP-MS), complemented by alpha spectrometry, to determine Pu isotopes present in a sample. Alpha spectrometry is used to give information on the ^{238}Pu isotope besides the joint $^{239+240}Pu$, because ^{238}Pu cannot be measured due to the presence of the isobar ^{238}U. Accelerator Mass Spectrometry (AMS) can achieve ICP-MS performance and it has the advantage that it is not so sensitive to the matrix effects as MS techniques [13]. The AMS sensitivity for ^{239}Pu is 2.0 fg / mg Fe and for ^{240}Pu is 0.5 fg / mg Fe [14].

In this paper, we present the first results for ^{239}Pu /^{242}Pu and ^{240}Pu /^{242}Pu isotope ratios obtained after the plutonium transport optimization tests at the 1MV AMS installation from IFIN-HH.

2. Experimental

2.1. Samples

Since, being anthropogenic, none of the plutonium isotopes provide high enough beam current to be measured with direct methods and specific instruments, such as Faraday cups, a uranium pilot beam was used to optimize transport through the accelerator assemblies. Natural uranium contained in an acidified aqueous solution was mixed with iron which acts as a carrier, and then the mixture was chemically and thermally processed to obtain highly oxidised uranium U_3O_8 in a Fe_2O_3 matrix. In the last phase, the material obtained was mixed with Nb powder for a better thermal and electrical conductivity target material that is required for AMS measurements. The ratio of U: Fe: Nb in the mixture was 50 µg: 1 mg: 3 mg [13].

For the transport of ^{239}Pu, ^{240}Pu and ^{242}Pu beams, in order to determine the isotopic ratios, a secondary standard prepared at the *Centro Nacional de Acceleradores* (CNA), Seville-Spain, was used. The isotopic ratios of this standard are $^{239}Pu/^{242}Pu = 0.534 \pm 0.011$ and $^{240}Pu/^{242}Pu = 0.281 \pm 0.005$ [15].

2.2. Experimental system

The AMS system, on which the measurements were made, is shown in Figure. 1. The description of the main components of this system has already been made in other studies [6, 16, 17]. In this paper, we describe briefly the tuning of the system for actinides beam transport and we also describe the selection procedure for a specific isotope.

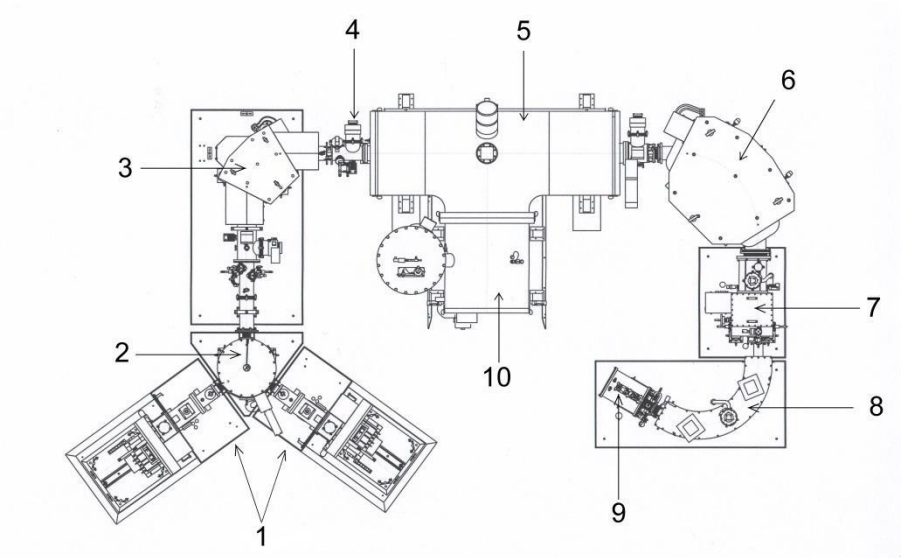

Fig. 1: The general layout of 1 MV HVEE AMS facility in Bucharest, Romania. (1) two ion beam sources; (2) electrostatic switching system; (3) injection magnet with multi beam switcher (bouncer); (4) Faraday cup; (5) 1 MV Tandetron^{TM} accelerator; (6) analysing Magnet; (7) three offset Faraday cups; (8) electrostatic analyser; (9) particle detector; (10) Cockcroft–Walton type HV power supply [16].

A cathode which contains the material of interest, once introduced into the carousel of the ion source (1), is bombarded with a Cs^+ primary beam, a process that will initiate the sputtering of the cathode. By applying and extraction potential, the negative ions and ionic molecular fragments with charge -1, are extracted and accelerated out of the source. Because the two negative ion sources are positioned at 120^o, in order to keep the negative ion beam on the optical axis of the system, we use an electrostatic switching system (2). The last ion-optics element on the low energy side is represented by 90 degrees magnetic analyser that makes a selection of the accelerated ions according to their momentum to charge ratio (3). The negative ion beam is then measured with a Faraday cup, mounted at the entrance of the acceleration system (4).

The analysed particles are then accelerated by the positive potential applied on the acceleration tubes of the Tandetron^{TM} 1 MV accelerator system (5). The accelerated negative ions are passing through a gas stripping channel (filled with Ar) where they lose part of their electrons becoming positive ions. Then the electric field will repel the positive ions that have different charge states.

After that the beam is once more analysed by momentum/charge state ratio with a 90° sector magnet (high-energy magnet, HEM) and by energy/charge ratio with a 120° electrostatic analyser (ESA) (8). The ions are finally detected and measured in terms of energy and energy loss in a two-anode gas ionization chamber (GIC) provided with a 75 nm silicon nitride entrance window (9). In case of plutonium measurements, the second anode is not used because all the ions are stopped along the first anode.

The optimization of ion transport is achieved by varying the electric and magnetic fields of the above-mentioned components until a maximum intensity of the pilot beam current is obtained in the Faraday cups (4) and (7).

Usually for the transport of rare isotopes (e.q. ^{14}C, ^{10}Be, ^{26}Al, etc.) intensity of the electric (E_e) and magnetic (B) fields are determined from the calculation, based on the balance between the centrifugal force and the electric force ($mv^2 / R = qE_e$), respectively magnetic ($mv^2/R = qvB$), where R, m, v and q represent the bending radius of the particle trajectory, mass, velocity and electric charge of the ion.

For the measurements of the uranium and plutonium isotopes, which have much higher masses than the commonly used isotopes, due to the hysteresis phenomenon, it is not recommended to change the magnetic field on low and high analysing magnets. The balance between the centrifugal force and the magnetic force can also occur if the magnetic field is kept unchanged but the energy of the particle is changed. On the low energy side this is achieved by applying an electrical voltage to the analyser magnet chamber (bouncer voltage, U_b) (3). In the high energy side, it is done by changing the positive voltage applied to the accelerator terminal (U_T). We worked with terminal voltage of 650 kV and charge state +3 for ^{238}U used as pilot beam.

To determine the ratio of plutonium isotopes, the Slow Sequential Injection (SSI) method implemented by High Voltage Engineering Europe (HVEE, Holland) [18], allows the sequential regulation of ion energy by changing the U_b, U_T and U_{ESA} voltage applied to the electrostatic analyser plates (8) [19].

In this way, several rare isotopes can be measured in the particle detector without changing the magnetic field settings and also the pilot beam current can be measured in a Faraday cup placed outside the trajectory of the rare isotope beam [20].

3. Results

After the entire system is optimized with a ^{238}U pilot beam, in order to optimize the transport for each of the three isotopes, ^{239}Pu, ^{240}Pu and ^{242}Pu, the parameters, U_b, U_T and U_{ESA}, corresponding to the transported isotope were manually set, until the rate of events obtained in the particle detector was maximized. The spectra obtained for the separate injection of the three isotopes are shown in Figures 2 to 4. In figure 2 is presented the spectrum of the ^{239}Pu isotope.

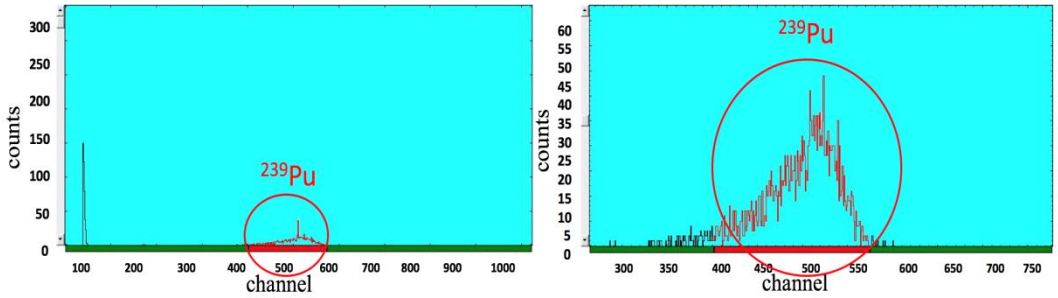

Fig. 2: Spectrum obtained for measuring ^{239}Pu after optimizing the ion beam transport parameters.

In Figure 3 it is observed that the spectrum obtained for ^{240}Pu is accompanied at low energies by an isotope signal corresponding to mass 160, which has the energy 2/3 of the one of ^{240}Pu. Depending on the nature of the sample and the chemical separation of plutonium, the $^{160}Dy^{2+}$ peak may be smaller or more intense [10, 15]. Even if the peaks of the two isotopes are well separated, if the peak of Dy is very intense, pile-up events could occur which affect the ^{240}Pu peak. To avoid this influence, optimize tuning should be done.

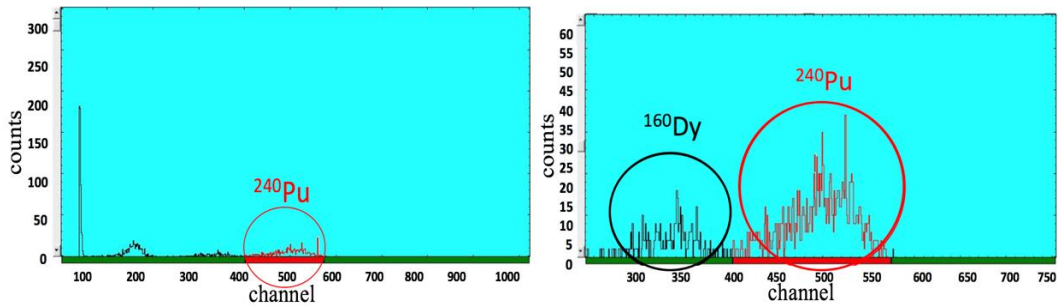

Fig. 3: Spectrum obtained for measuring ^{240}Pu after optimizing the ion beam transport parameters. On the right side, the first signal is corresponding to $^{160}Dy^{2+}$ isotope.

In order to determine the isotopic ratios of plutonium, each isotope is measured in the ionization gas chamber. After that the system software is

automatically set to quantify each isotope integrating the corresponding area to each mass of interest. In figure 4 is presented the spectrum for the ^{242}Pu, the isotope of Plutonium that is used to report the first two isotopes, being considered the reference isotope.

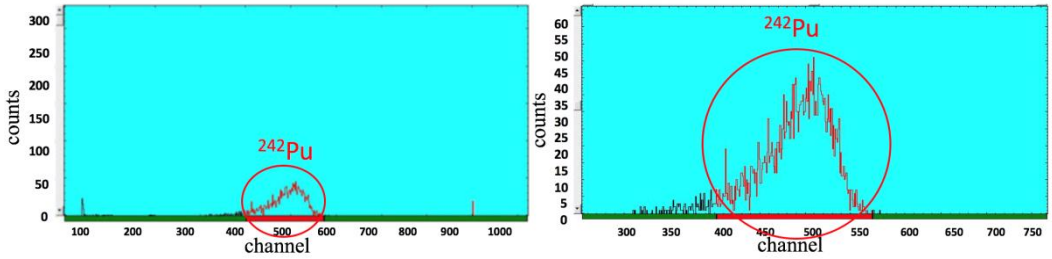

Fig. 4: Spectrum obtained for measuring ^{242}Pu after optimizing the ion beam transport parameters.

Experimental results obtained for the isotopic ratios of plutonium ^{239}Pu/^{242}Pu and ^{240}Pu/^{242}Pu are presented in Table 1, Figure 5 and Figure 6. For the experimental part, three samples (cathodes) were used, each containing the CNA standard for plutonium isotopes. Three sets of measurements were made, the only difference between these sets is given by the temperature applied on the cesium reservoir. Initially the measurements were made using the Cs temperature at 100^{o} C. For this temperature, the count rate obtained in the detector for the ^{242}Pu isotope was about 5 cps. In order to increase the count rate and accordingly measurement accuracy, the Cs temperature was raised up to 120^{o} C. At this value, the count rate for ^{242}Pu increased to about 13.2 cps. For the same purpose, measurements were made also at 130^{o} C, but the cps values did not increase significantly.

Table 1.

Experimental results obtained for plutonium isotopic ratio ^{239}Pu/^{242}Pu and ^{240}Pu/^{242}Pu

Cathode no.	T_{Cs} (℃)	^{239}Pu/^{242}Pu		^{240}Pu/^{242}Pu	
		Average value	Relative error (%)	Average value	Relative error (%)
1	100	0.5417	4.04	0.2801	4.42
2		0.5291	4.14	0.2841	4.36
3		0.5176	5.45	0.2876	6.28
1	120	0.5295	2.27	0.2839	1.32
2		0.5242	2.00	0.2832	1.52
3		0.5205	2.08	0.2847	1.80
1	130	0.5362	1.85	0.2904	2.92
2		0.5145	4.06	0.2790	1.87
3		0.5600	2.36	0.2853	1.67

In Table 1 the first column shows the number of the cathode used during the measurements, T_{Cs} represent the values of the Cesium temperatures and in the last four columns are presented the average values and the relative errors obtained for the isotopic ratios of $^{239}Pu/^{242}Pu$ and $^{240}Pu/^{242}Pu$.

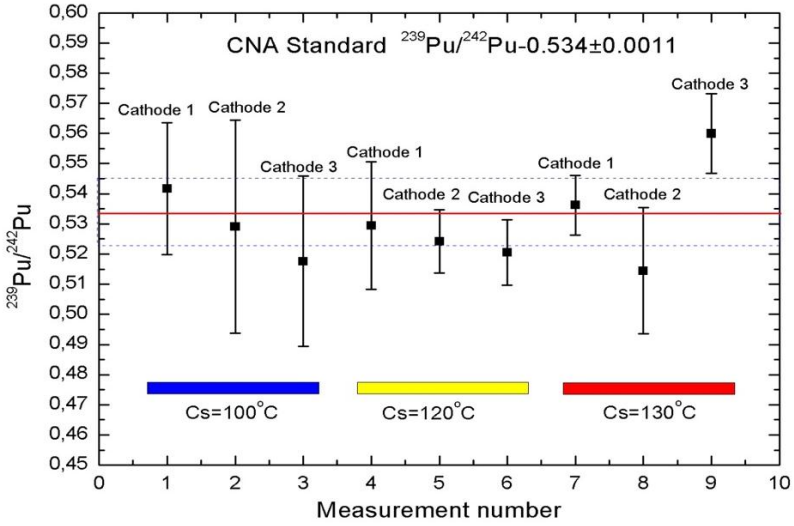

Fig. 5: Experimental results obtained for $^{239}Pu/^{242}Pu$ ratio. Red line represents the reference provided by the standard; blue line represents the error interval.

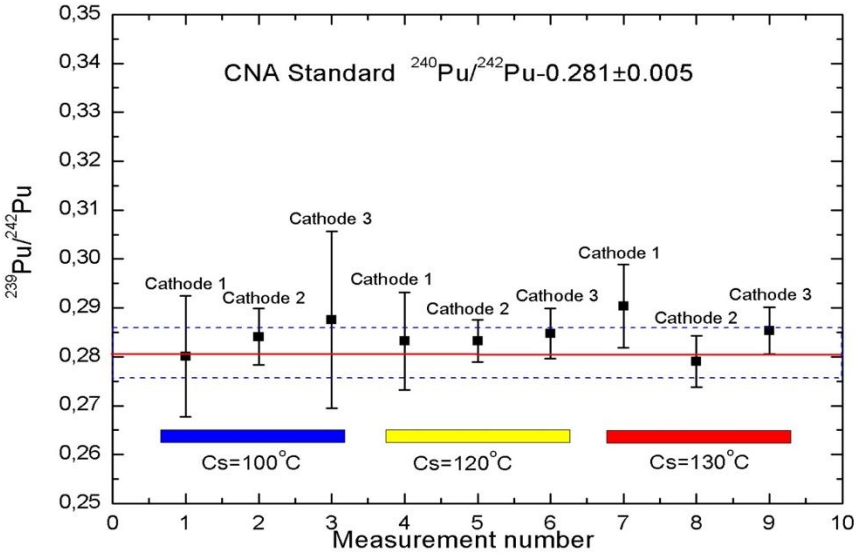

Fig. 6: Experimental results obtained for $^{240}Pu/^{242}Pu$ ratio. Red line represents the reference provided by the standard; blue line represents the error interval.

From the Figures 5 and 6 can be observed the isotopic ratios values for the Cs temperature 120^{o} C and 130^{o} C. At the temperature of 130^{o} C there are some fluctuations of the ion beam current, which indicate possible instabilities in the ion source functioning.

The value of the ^{239}Pu/^{242}Pu ratio obtained from present measurements of the three samples was 0.5244±0.0064, and that of ^{240}Pu/^{242}Pu was 0.2839±0.0025.

The average of the values obtained, in a normal operating mode of the ion source (120^{o}C) is in agreement with the nominal value reported by CNA Seville Laboratory [15], 0.534 ± 0.011 for ^{239}Pu / ^{242}Pu and 0.281 ± 0.005 for ^{240}Pu / ^{242}Pu. The measurement error of these two isotopic ratios of plutonium could be improved by measuring for a longer time each sample in order to have better statistics. This will be possible after our lab from IFIN-HH will procure standard solutions.

4. Conclusions

After more than five years of measurements of ^{14}C, ^{10}Be, ^{26}A and ^{129}I, tests for the transport of plutonium isotopes were carried out in order to determine its isotopic ratios for nuclear environmental pollution studies. For the first time in our laboratory the isotopic ratios of plutonium were measured. The average values obtained for ^{239}Pu / ^{242}Pu and ^{239}Pu / ^{242}Pu ratios, in a normal operating mode of the ion source (120^{o}C) are in agreement with the nominal values reported by CNA Seville Laboratory [15]. As a result of these measurements, it was concluded that at the 1 MV AMS system from IFIN-HH, the isotopic ratios of plutonium can be determined, and such measurements can be extended to other actinides.

In the near future optimization tests to improve measurement errors and reduce the background will be performed. For that we will have to purchase standard solutions for each isotope to be measured. Samples with multiple isotopic concentrations will be made in enough quantities such that each sample can be measured as long as it is required.

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