Status report on AMS Measurements of Plutonium Isotopes using the 1MV Tandetron Accelerator at IFIN-HH

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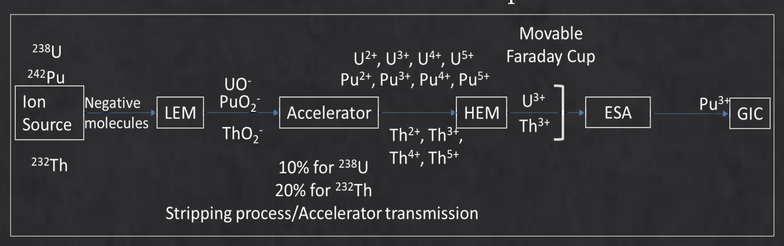
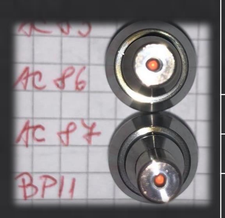
This report gives an overview regarding the progress that was made in the determination of Plutonium isotopes at IFIN-HH, and it starts with the studies performed for the transport and measurement of actinides using the 1MV Tandetron Accelerator [1, 2]. The Accelerator Mass Spectrometry is the most sensitive measurement method that allows us to determine $^{239, 240, 244}$Pu radioisotopes from environmental samples with extremely low isotopic concentrations.

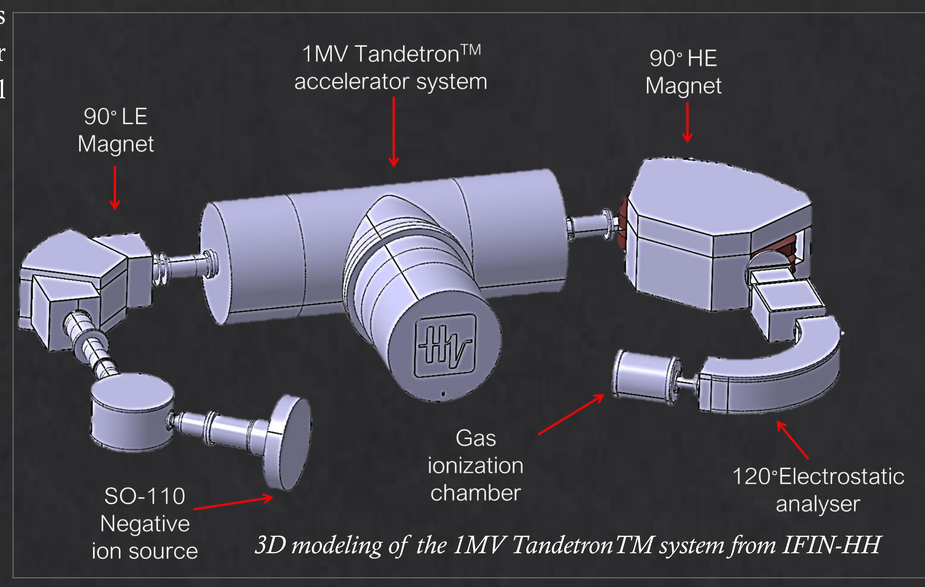
When measuring Plutonium isotopes, we focus our research activity on two main directions, monitorization of environmental nuclear pollution and nuclear astrophysics. In the perspective of measuring actinides for applications in nuclear astrophysics it is important to determine the detection efficiency of our system, the detection efficiency implies the ionization process, the losses through the accelerator and detection.



In order to determine the detection efficiency of our AMS system, we prepared several samples with precise quantities of each isotope of interest, also we quantified all the material lost during sample preparation so that it is known how much radioisotope we have in the cathode at the beginning of the measurement. The targets/ cathodes were measured until total exhaustion.

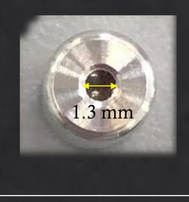


^{x}Ac (U and Th particles)= $^{x}Ac^{3+}$ (A)·t (s)/3·e (C) e=1.602 x 10^{-19}C	*Transmission/Detection Efficiency Ion source - FC3 (Movable Faraday Cup) / GIC*
^{232}Th (averaged for 2 cathodes)	$1.43 \cdot 10^{-4} \pm 0.51 \cdot 10^{-4}$
^{238}U (averaged for 2 cathodes)	$0.32 \cdot 10^{-4} \pm 0.03 \cdot 10^{-4}$
^{242}Pu (averaged for 4 cathodes)	$0.41 \cdot 10^{-4} \pm 0.07 \cdot 10^{-4}$



3D modeling of the 1MV TandetronTM system from IFIN-HH

The target material was obtained by mixing the new Plutonium Standard ColPuS [3, 4] that was kindly provided by Prof. Dr. Tibor Dunai and Dr. Erik Strub from Koln University, with 1mg/mL Iron solution, purchased from High Purity Standards, then Pu and Fe were precipitated with ammonia solution as hydroxides. The next step was to dry the material at 65°C for 2 hours and then calcinated at 800°C for 4 hours. Finally, the Pu dispersed homogenously in Iron Oxide was mixed with 3mg of Niobium 99.8% (Sigma Aldrich) and pressed in aluminum cathodes (HVEE). The ^{238}U and ^{232}Th isotopes were used as pilot beams for determining the ion transport parameters, the beam currents being measured by using the Gated Charge Pump Digitizers (GCPD), because it allow us the measurement of pA currents. For counting the plutonium isotopes was employed the so-called Slow Sequential Injection method (SSI), the measurement sequence being ^{242}Pu 5s, ^{239}Pu 5s, ^{240}Pu 5s and ^{244}Pu 20s, each sequence being repeated 5x6 times.



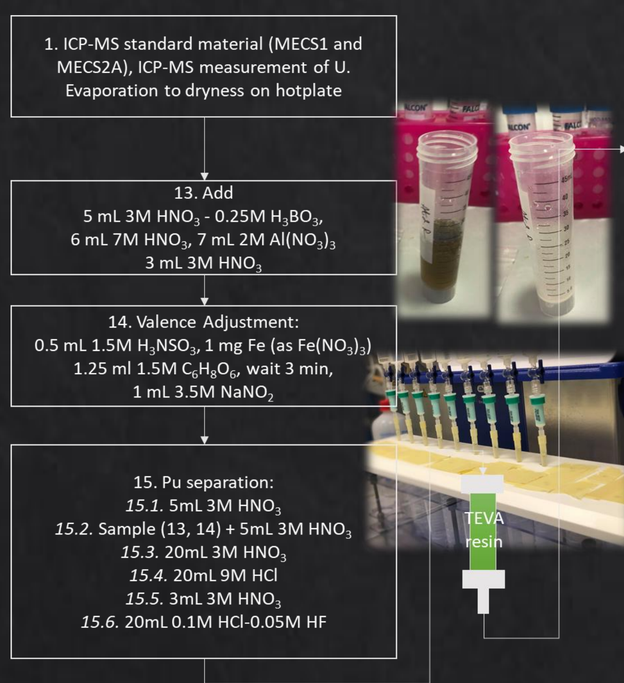
	^{239}Pu /^{242}Pu			*^{240}Pu /^{242}Pu*			*^{244}Pu /^{242}Pu*		
Cathode number	ratio	σ (ext)	δ (int)	ratio	σ (ext)	δ (int)	ratio	σ (ext)	δ (int)
ColPuS1	1.033	0.053	0.017	1.039	0.054	0.028	0.0986	0.0063	0.0024
ColPuS2	1.025	0.042	0.016	1.042	0.043	0.014	0.0925	0.0048	0.0030
ColPuS3	1.029	0.045	0.025	1.052	0.046	0.023	0.0937	0.0052	0.0030
ColPuS4	0.955	0.047	0.013	1.001	0.049	0.017	0.0896	0.0056	0.0021
ColPuS5	1.034	0.038	0.015	1.077	0.039	0.021	0.0915	0.0042	0.0018
ColPuS6	0.991	0.049	0.021	1.004	0.049	0.016	0.0930	0.0057	0.0031
ColPuS7	0.985	0.048	0.008	1.008	0.048	0.012	0.0876	0.0054	0.0011

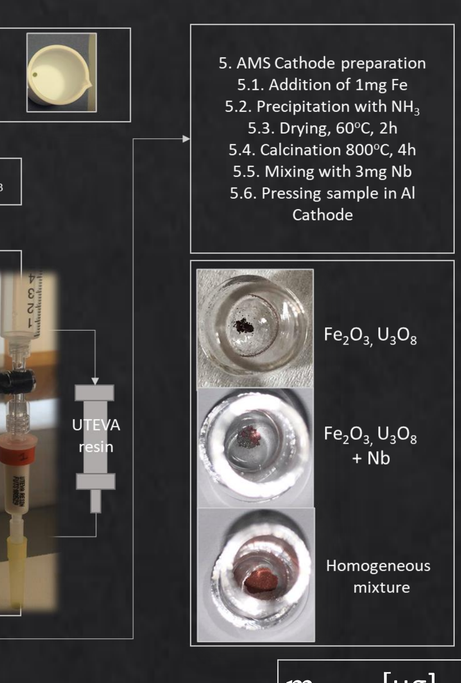
In the table in the right side are presented the results obtained for 7 sample targets measured in IFIN-HH, also, the statistic or internal uncertainty (δ), and the standard deviation of the weighted mean or external which informs us about the stability of the AMS system during the extended measurement (σ).

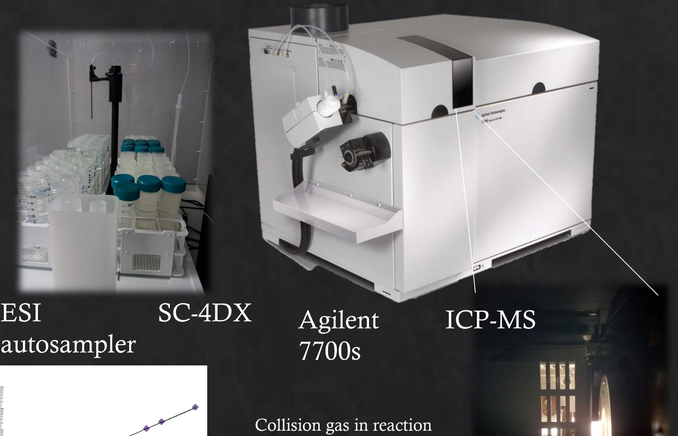
The results obtained for the isotopic ratios are listed in table on the right side and agrees with the values given by 7 AMS laboratories and with the consensus values of the standard reported in [4].

Isotopic ratio	*^{239}Pu /^{242}Pu*	*^{240}Pu /^{242}Pu*	*^{244}Pu /^{242}Pu*
Measured, 1MV IFIN-HH 7 cathodes	*1.007 ± 0.030 (3.03%)*	*1.032 ± 0.028 (2.76%)*	*0.0924 ± 0.0035 (3.74%)*
Mean normalized – 7 AMS LAB, Dittmann 2018	*1.067 ± 0.028 (2.62%)*	*1.076 ± 0.021 (1.95%)*	*0.1075 ± 0.0045 (4.18%)*
Consensus values, Dittmann 2018	*1.058 ± 0.008 (0.08%)*	*1.066 ± 0.009 (0.83%)*	*0.1051 ± 0.0021 (2.0%)*

Radiochemical Separation of Thorium, Plutonium and Uranium using TEVA and UTEVA resins at IFIN-HH, AMS Laboratory, DFNA







Collision gas in reaction cell: He (5.5 = 99.9995 %); Molecules, Oxides < 1% Doubly charged species: < 2%

	TEVA		UTEVA
	^{232}Th	^{238}U	^{238}U
m_{added} [µg]	40.10	40.15	120.62
$m_{measured}$ [µg] ± SD	37.92 ± 0.49	35.48 ± 0.43	115.14 ± 1.38
Chemical recovery [%]	88.45	94.55	98.31

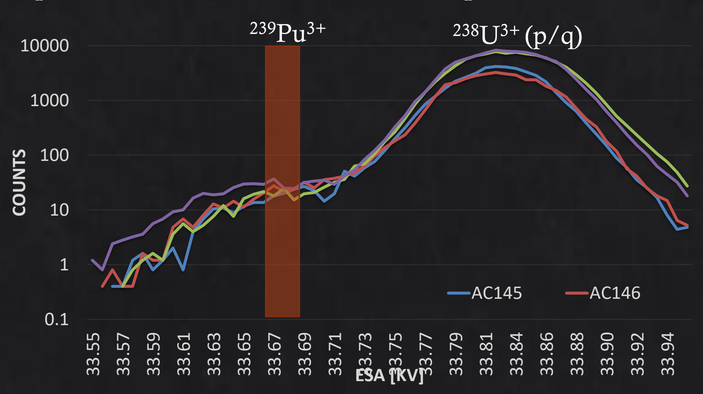
The natural Radioisotopes Uranium-238 and Thorium-232 are used in our laboratory as pilot beams. In order to obtain clean, in-house produced materials, we tested two chemical procedures for the separation and extraction of the needed actinides by using TEVA and UTEVA resins from Eichrom. For the determination of the radiochemical recoveries, Uranium and Thorium fractions were measured using Inductively Coupled Mass Spectrometry [5].

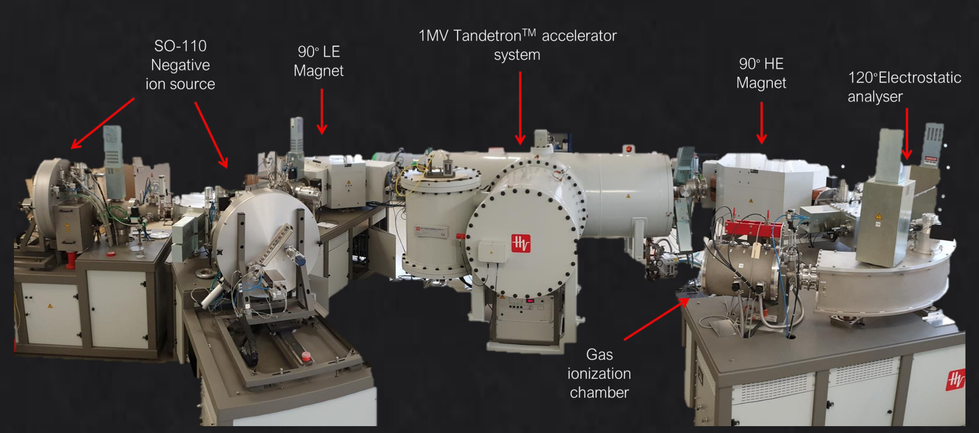
The results for the chemical recoveries obtained for the radiochemical resin separation are showed in the table and for all situations the values are above 90%, a reason for these good results is the fact that the sample preparation implies only a simple evaporation. Also, these values show a good perspective for using the procedure for real environmental samples.

*MECS 1 and 2A – Multi Element Certified Material

When, in an AMS system, we take the tuning parameters of the ^{239}Pu and we introduce a ^{238}U sample into the ion source, counts are recorded in the detector. We have seen that this interference is caused by molecular fragments of $^{238}U^{3+}$ that undergo dispersive and/or charge exchange processes in the high energy zone, so that its kinematic properties, apparently distinguishable by mass analyzers, they undergo certain changes that compensate for the mass deficit or necessary energy.

In order to investigate this phenomenon, in the graph in the right side is showed the ESA voltage scan around the optimal value for the mass 239u while the other AMS system elements are set for the same mass. The samples used have different quantities of ^{238}U - 10µg for AC145 and AC146 and 20µg for AC148 and AC149. The results showed us that the molecular fragments that have the same magnetic rigidity as 239 mass can be quantified, when discussing about environmental samples it is negligible and when necessary, the slit settings at the high-energy side of the Spectrometer are modified.





Conclusions

This work gives an overview regarding the progress that was made in the determination of Plutonium isotopes at IFIN-HH by the successful measurement of the new Plutonium ColPuS Standard and chemical procedures for the separation of Uranium and Thorium. Also, important steps were performed in the perspective of nuclear astrophysics applications by determining the detection efficiency of the 1MV Accelerator from IFIN-HH, Romania and finally a brief result about the molecular interference that can occur while measuring ^{239}Pu.

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References
1. D. Păcesilă et al., Romanian Reports in Physics, 70, 2018.
2. D. Păcesilă et al., U.P.B. Sci. Bull, Series A, 82, 2020.
3. B.A. Dittmann, et al., Nucl. Instr. Meth. B 361 (2015) 327–331.
4. B.A. Dittmann, et al., Nucl. Instr. Meth. B 438 (2019) 189–192.
5. Serban, A. et al., J Radioanal Nucl Chem (2021).

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