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^{236}U identification in the new AMS beamline at the TANDAR accelerator

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Abstract. ^{236}U ($T_{1/2} = 23$ My) is an excellent monitor for nuclear contamination in the environment. Indeed, spent nuclear fuels present isotopic ratios ^{236}U/^{238}U several orders of magnitude higher than natural samples. ^{236}U also provides a useful fingerprint to identify and trace nuclear material for safeguards purposes. Here we describe a new beamline for the discrimination of ^{236}U using the accelerator mass spectrometry (AMS) technique. This system comprises a Wien velocity filter and a 6.7 m long time-of-flight (ToF) path with a focusing quadrupole. The ToF is determined by a time zero detector and a surface barrier detector, which also provides an energy measurement. The capability of the system to discriminate ^{236}U from the much more abundant isotopes ^{235}U and ^{238}U is shown. While sensitivity values of ^{236}U/^{238}U$\sim 10^{-8}$ were achieved, ongoing works aim to improve this performance.

1. Introduction

Radionuclides are useful tracers for several research fields since they provide not only spatial but also time information. Typical examples are the use of ^{14}C (5.7 ky) for archaeology, ^{10}Be (1.4 My) for geology and ^{129}I (16 My) for environmental and oceanographic studies [1]. To the latter application, ^{236}U (23 My) can add substantial information [2, 3].

^{236}U is mainly produced by thermal neutron capture on the abundant, primordial isotope ^{235}U. In nature, these neutrons can be provided by (α,n) reactions on light nuclei, induced fission of ^{235}U, spontaneous fission of ^{238}U and at Earth surface by cosmic rays. Thus, the production and concentration of ^{236}U are strongly dependent on the neutron flux, which in turn depends on several factors: concentration of light nuclei, the depth from the Earth surface, the amount of water in the ore necessary for an efficient neutron thermalization and the presence of elements with high neutron capture cross section. It is estimated that about 30 kg of naturally produced ^{236}U exists in the Earth surface and $\sim$0.5 kg in the oceans [4]. However, the anthropogenic contribution to the ^{236}U inventory is much larger than the natural one. Around 900 kg of ^{236}U have been released to the environment during the atmospheric nuclear weapon testing period between 1945 and 1963 [5], and about 10^6 kg were produced in power reactors [4], although only a fraction of the latter is expected to have leaked to the environment due to nuclear fuel reprocessing plants discharges (∼100 kg [6]) and accidents in power reactors.

In samples of preanthropogenical material, the isotopic ratio ^{236}U/^{238}U lies in the 10^{-14}-10^{-9} range, while in those stemming from man-made processes this ratio can be as high as 10^{-4}-10^{-2}. This feature makes the radionuclide ^{236}U a very sensitive monitor for nuclear contamination in the environment and a valuable tracer used to determine the origin of an unknown uranium material, contributing in this way to nuclear safeguards controls. In addition, in the past few years, the radionuclide ^{236}U became a complementary oceanographic tracer in conjunction with ^{129}I [2, 3].

Since its long half-life, sensitive measurements of ^{236}U abundances are not achievable by alpha-decay detection. On the other hand, molecular isobars, such as ^{204}Pb^{16}O$_2$ and ^{235}U^{1}H, interfere the measurement of low concentrations of ^{236}U by conventional mass spectrometry. Hence, accelerator mass spectrometry (AMS) turns out to be the most suitable technique for these measurements. When using a tandem accelerator, the stripper at the high-voltage terminal assures the dissociation of all molecular background.

In the following section, we describe the commissioning of a time-of-flight beamline at the TANDAR accelerator aimed at the detection of ^{236}U by the AMS technique. In section 3, the sample preparation and measurement procedures are explained. In section 4, results for detection of ^{236}U are shown and discussed. Summary and outlook are given in section 5.

2. Experimental setup

In fig. 1 schemes of the 20 MV TANDAR accelerator and of the new AMS beamline are shown. Negative atomic or molecular ions are produced by a 40-cathodes sputtering ion source. A first mass selection is performed at the injection double-focusing magnet (bending radius $\rho = 305$ mm and $(B\rho)^2_{max} = 17.5$ amu×MeV/e^2) which, in conjunction with a 10 mm slit, provides a selectivity of $\Delta m/m \sim 1\%$. After a preacceleration voltage of 165 kV, ions are injected into the main accelerator where terminal voltages of around 8 MV are used. Carbon foil strippers are often damaged by heavy ion beams and introduce large angular straggling to the beam. Instead, N$_2$ gas stripper (~ 1 μTorr, 1 m long) is used. The analyzing double-focusing magnet ($\rho = 2$ m and $(B\rho)^2_{max} = 500$ amu×MeV/e^2) selects the charge state of uranium ions ($q = 8+$ was used in these experiments). Using a 5 mm slit at the image focal point, a selectivity of $\Delta m/m \sim 0.2\%$ can be expected. The analyzing magnet mechanically rotates to direct the beam to each experimental line, without the use of a switching magnet.

It is worth mentioning that in the injection magnet the suppression of neighboring masses is not complete. In the mass region of interest (mass of ^{236}U^{16}O, 252 a.m.u) the selectivity of 1% allows the injection of molecules with mass between 250 and 254. An effective mass selection can be expected in the analyzing magnet, since its mass selectivity ($\Delta m/m \sim 0.2\%$) is adequate for discriminating mass 236 from 235. However, charge-exchange collisions in the high energy side of the accelerator can provide ^{235}U (and ^{238}U) ions the magnetic rigidity necessary to be accepted by the analyzing magnet (see Sect. 4).

In this facility, a new Wien velocity filter and a ToF system were installed in a beamline conducting to a QDD magnetic spectrometer. This setup is intended for the detection of ^{236}U and ^{129}I. These radionuclides suffer no interference from stable isobars[1], so they can be discriminated from their more abundant isotopes by a ToF measurement. The Wien filter, acquired from Danfysik®, is composed by a 1-m-long magnet and an electrostatic dipole with an electrode gap of 50 mm. Electric and magnetic forces counterbalance for a selected ion velocity, thus deviating interfering ions. The Wien filter was placed at the exit of the first magnetic quadrupole triplet following the analyzing magnet (MQ-L1, see fig. 1). Since the

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[1] ^{129}Xe produces no negative ions in the ion source

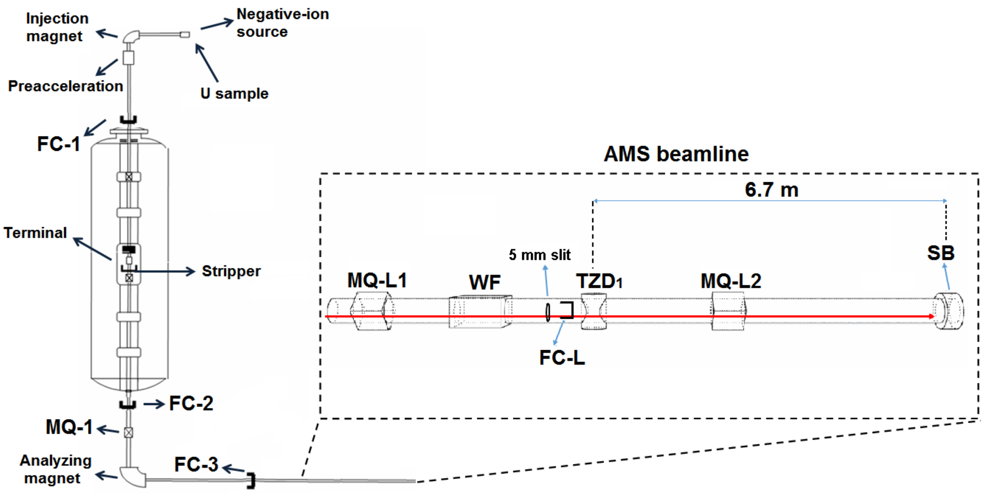

Figure 1. TANDAR accelerator and AMS beamline. MQ=magnetic quadrupole; WF=Wien filter; FC=Faraday cup; TZD=time zero detector; SB=surface barrier detector. Not in scale.

analyzing magnet selects the beam energy bending it in the vertical axis, the Wien filter was installed to exert its forces in the horizontal axis, so as to be decoupled. The maximum magnetic and electric fields are 0.3 T and 24 kV/cm, respectively. The coil current and electrodes voltages are set by PLC-controlled power supplies. For its initial calibration, ions of several masses (^{12}C, ^{35}Cl, ^{127}I and ^{197}Au) and velocities ($0.02 < v/c < 0.10$) were tuned. For selecting 70-MeV ^{236}U ions a coil current of 150 A (0.234 T) and electrode voltages of ±52 kV were adopted. Thus, under the condition in which ^{236}U is tuned, interfering ^{235}U and ^{238}U ions are deviated ~1.3 mm and ~2.6 mm at the image focal point of the quadrupole MQ-L1 (which is the start of the ToF system). At this point a 5 mm slit was placed to partially suppress these interferences.

The ToF system comprises a time zero detector (TZD) as start and a surface barrier silicon detector as stop, separated by 6.7 m. The silicon detector also provides an energy measurement. An alternative configuration using a second TZD (close to the surface barrier detector) to provide a faster stop signal was also tested, achieving lower total detection efficiency values.

The TZD is composed by a thin carbon foil ($\sim 20\ \mu$g/cm^2) and a Z-stack microchannel plate (MCP) assembly. Three different configurations were tested: a) direct collection of forward electrons, b) direct collection of backward electrons, and c) electrostatic mirror [7] (see Fig. 2). We run the measurements using the latter configuration since it showed a better intrinsic efficiency[2].

The surface barrier detector, which is 1 mm thick and 19.5 mm diameter, is over-biased to 220 V to improve its timing performance. Because the flight path of 6.7 m is rather large, a magnetic quadrupole triplet half-way between the TZD and the surface barrier detector was used to minimize the loss of ions due to angular straggling in the carbon foil.

The signal processing was performed with standard nuclear instruments electronic modules (NIM). The time signals from the TZD and the surface barrier detector were furnished to constant fraction discriminator modules to produce negative logic pulses. The output signal from the TZD discriminator was delayed about 1.5 μs using a gate and delay generator and was fed together with the output pulse from the surface barrier detector discriminator to a time-to-

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[2] Defined as the ratio between the number of output signals and the number of impinging ions. The former is estimated by the coincidence events in the TZD and the surface barrier detector and the latter by the total events in the surface barrier detector.

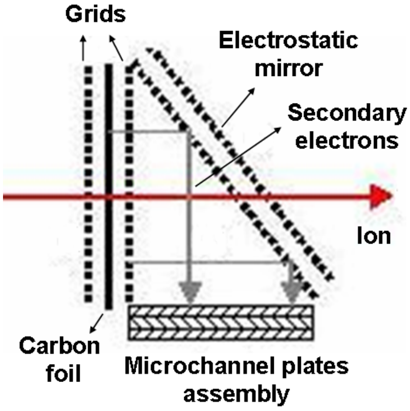


Figure 2. Electrostatic mirror configuration of the time zero detector.

amplitude converter (TAC) used in the inverse start-stop mode, i.e. the surface barrier detector starts the TAC and the TZD stops it. In this way, TAC dead time due to ions triggering the TZD but not reaching the surface barrier detector is avoided. This is important if the TZD rate is high, which is our case due to the high isotopic ratio of the samples used as standards.

3. Experimental procedure

The ToF detection system was characterized using samples with different content of uranium and concentrations of ^{236}U: (a) samples with 1-5 milligrams of non-enriched uranium, (b) samples with about 20 ng of non-enriched uranium, (c) samples ranging from 20 ng to 20 μg of enriched uranium, and (d) samples with 1-5 milligrams of enriched uranium with high ^{236}U concentration (^{236}U/U= 0.03%).

Samples (a) were used to generate a ^{238}U pilot beam (intensities of several nanoamperes) necessary for tuning the ion optic elements of the accelerator, including the Wien filter, by optimizing the reading in the Faraday cups (FC). Samples (b) and (c) were used for tuning low-intensity beams (rates $\lesssim$ 1 kHz) for the calibration of the detection system using the ^{235}U and ^{238}U ToF and energy signals. Samples (d) were used as calibration standards for the detection of ^{236}U. Measuring the ^{238}U beam intensity of these samples in FC-L1 (immediately before the TZD$_1$, see fig. 1) the beam transmission of the system can be determined. This transmission is necessary for correcting systematic effects when determining isotopic ratios ^{236}U/^{238}U of unknown samples. The use of a calibration standard corrects for any isotopic fractionation effect, even though it is expected to be negligible in this mass region.

Nanogram samples (b) and (c) were prepared dissolving 1 mg of uranyl nitrate ($UO_2(NO_3)_2$) in 1 l of a 10^{-4} M HCl solution and 50 mg of iron oxide (Fe_2O_3) in 50 ml of concentrated HCl (36.5% m/m). In these samples, iron is used as a carrier material for the coprecipitation and to form a macroscopic bulk for the handling of the sample material. An aliquot of 50 μl of the uranyl solution was added to 400 μl of the iron solution and, by the addition of NH_3, the solution was neutralized (pH $>$ 7). This induces coprecipitation of iron as iron hydroxide ($Fe(OH)_3$) and of uranium in the form of ammonium diuranate ($(NH_4)_2U_2O_7$). After centrifugation, the supernatant was extracted and the precipitate was washed and subsequently dried in vacuum at ∼60°C. Then, it was transferred to a ∼1 ml quartz crucible, where it was heated to 850°C in a muffle furnace to convert it into the oxide form. After cooling it at room temperature, approximately 1 mg of metallic aluminum powder was added to the crucible and mixed with the oxide, which is composed of ∼0.5 mg of Fe_2O_3 and ∼20 ng of uranium in the form of triuranium octoxide (U_3O_8). Finally, the solid was scrapped from the crucible and loaded in aluminum cathodes.

Milligram samples (a) and (d) were prepared heating 1-5 mg of uranyl nitrate to 800°C in a muffle furnace to convert it in U_3O_8. Once cool, the solid was mixed with ~2 mg of metallic aluminum powder and loaded in aluminum cathodes.

Uranium samples were loaded into the ion source, where uranium ions were extracted as UO^-. An initial tuning of accelerator beam optics can be done with a $^{197}Au^{7+}$ beam, which fits the same magnetic rigidity as $^{238}U^{8+}$ for a similar terminal voltage (see table 1). Gold samples deliver more intense beams than uranium ($I \sim 1$ μA at FC-1, after the injection magnet), easing the optimization of the ion optics elements. After it, a pilot beam of $^{238}U^{8+}$ ($I \sim 50$ nA at FC-1) is tuned up to FC-L1 and optimized. Then, nanogram samples (b) are used to produce low-intensity beams of ^{235}U and ^{238}U which allow the calibration of the ToF system, determining the expected spectrum region for ^{236}U events. Finally, from samples (c) ^{236}U was tuned and their events were identified and discriminated from the interfering ^{235}U and ^{238}U events, which are originated in charge-exchange collisions within the accelerator tube (see Sect. 4).

While the terminal voltage can be regulated in slit mode when pilot beams ($I > 0.1$ nA) are tuned, low intensity beams require the use of the generating voltmeter (GVM). ^{235}U and ^{236}U tuning parameters were calculated from the pilot beam parameters, maintaining the same magnetic rigidity. Hence, the analyzing magnet and the high-energy side magnetic optic elements are kept constant. The magnetic field of the Wien filter is kept fixed as well. Thus, the parameters to be changed for switching different ion masses are the injection magnet field, the terminal voltage and the electric field of the Wien filter.

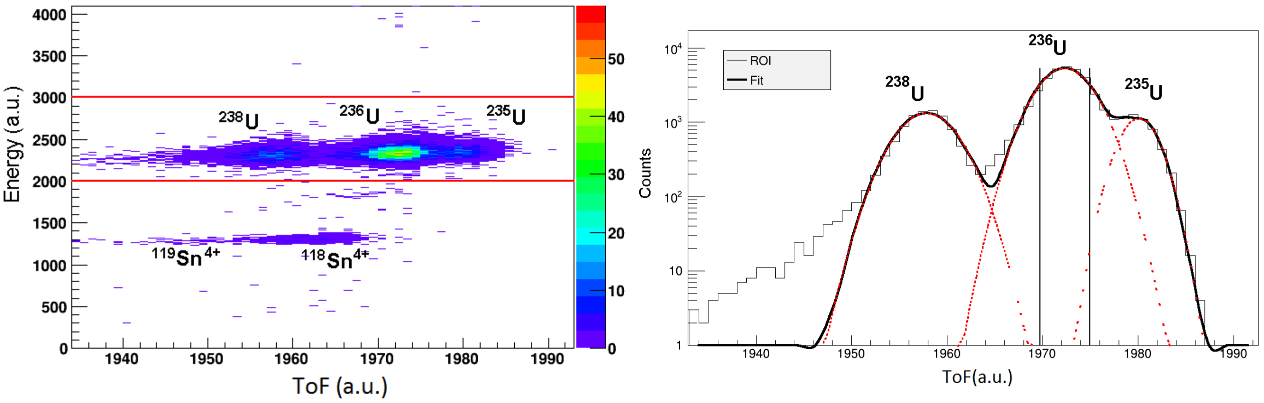
Table 1. Parameters used for the tuning of the different ions for a magnetic field of 11581 G in the analyzing magnet. M_{inj} is the molecular mass extracted from the ion source, B_{inj} is the magnetic field of the injection magnet, V_{Term} is the terminal voltage and V_{WF} is the plate voltage of the Wien filter.

Ion	M_{inj} (uma)	B_{inj} (G)	V_{Term} (MV)	Energy (MeV)	V_{WF} (kV)	ToF (ns)
$^{197}Au^{7+}$	197	7395	8.036	64.47	±53.9	842.6
$^{235}U^{8+}$	251	8348	7.880	70.58	±51.8	879.4
$^{236}U^{8+}$	252	8364	7.846	70.28	±51.7	883.2
$^{238}U^{8+}$	254	8398	7.780	69.69	±51.2	890.7

4. Results and discussion

The two dimensional spectrum shown in Fig.4(a) was taken tuning mass $A = 236$ from a sample containing 0.03% of ^{236}U and 3.5% of ^{235}U. Fig. 4(b) corresponds to the projection of uranium events onto the ToF axis. The interfering ^{235}U ions enter as molecules such as $^{235}U^{17}O^-$, $^{235}U^{16}O^-$ or $^{235}U^{16}O^{1}H^-$, while ^{238}U ions may stem from the low energy tail of the $^{238}U^{16}O^-$ beam. After stripping to a charge state 7+ or 9+, a subsequent charge-exchange collision with a residual gas molecule can result in $^{235}U^{8+}$ and $^{238}U^{8+}$ ions. When the collision occurs at a determined place of the high-energy side of the acceleration tube, these interfering ions may acquire the same magnetic rigidity as the tuned $^{236}U^{8+}$, and hence be accepted by the analyzing magnet [9]. Although the Wien filter slightly deviates interfering ^{235}U and ^{238}U (see sect. 2), a considerable amount of these ions still goes into the ToF, which must further discriminate their signals.

The events having the same ToF than uranium but approximately half of their energy can be identified as $^{118}Sn^{4+}$. When tuning $^{236}U^{8+}$, the ^{118}Sn present in trace amounts in the aluminum



(a)

(b)

Figure 3. (a) Energy vs. ToF two-dimensional spectrum. Horizontal lines show the gate used to project uranium events to the ToF axis. (b) Projection of uranium events into the ToF axis. Vertical lines show the ROI for the ^{236}U events. Dashed lines are the individual gaussian functions for each peak obtained by the fit. The solid line is the sum of the three gaussians.

alloy of the cathode can be extracted from the ion source as the molecular ion $^{118}\mathrm{Sn}_2^{18}\mathrm{O}^-$, which has the same mass of the tuned ion $^{236}\mathrm{U}^{18}\mathrm{O}^-$, thus being accepted by the injection magnet. After stripping to the 4+ charge state in the terminal, it will reach the analyzing magnet with half of the energy of the $^{236}\mathrm{U}^{8+}$ ions and, since both ions have the same mass to charge ratio, it will be accepted. Moreover, it will have the same velocity than $^{236}\mathrm{U}^{8+}$, so it will not be deviated by the Wien filter. However, these events are easily discriminated by the energy measurement and therefore pose no inconvenience to the ^{236}U identification.

The sensitivity, i.e. the minimum isotopic ratio $^{236}\mathrm{U}/^{238}\mathrm{U}$ that the system is capable to measure, is around $^{236}\mathrm{U}/^{238}\mathrm{U} = 10^{-8}$. This sensitivity was estimated as follows: the ToF spectrum of fig. 4(b) was fitted with a sum of three Gaussian functions. The region of interest (ROI) associated with the ^{236}U events was defined as the interval $\mu \pm \sigma$, where μ and σ are the mean and the standard deviation of the ^{236}U peak obtained by the fit. Using that the isotopic ratio of the sample was $^{236}\mathrm{U}/^{238}\mathrm{U}$=0.03%, it was determined that the tail of ^{235}U and ^{238}U peaks contribute at the level of 2×10^{-8} in the ratio.

Although the achieved sensitivity is still far from that of other dedicated AMS laboratories [8], there are many issues to be improved. The improvements will require the use of thinner carbon foils in the TZD$_1$ and a more restrictive vertical slit at image focal point of the quadrupole MQ-L1, to improve the Wien filter selectivity prior to the TZD$_1$. Additionally, the tuning of lower charge states, which have a higher yield at the stripper, will be tested. This will also make the difference between the ToF of the uranium isotopes larger, improving their discrimination. Due to the limitation in magnetic rigidity of the analyzing magnet and beam optic elements, the use of lower charge state as, for example, $q = 5+$, will require voltages of about 4 MV. Operating at such low voltages requires the use of shorting rods at the second half of both low- and high-energy sector of the accelerator.

5. Summary and perspective

In this work, we presented the commissioning and initial tests of a new dedicated AMS beamline at the TANDAR accelerator aimed at the detection of the radionuclide ^{236}U. It includes a Wien filter and a 6.7 m-long ToF system composed by a time zero detector and a silicon surface barrier detector, which also provides an energy measurement. To study the performance of this system, samples with different amounts of uranium and ^{236}U concentration were tested. Experimental results show that a sensitivity of ^{236}U/^{238}U=10^{-8} was achieved, but ongoing work is being done to improve this performance. The ToF system is also suitable for the detection of other radionuclides without interfering isobars, such as ^{129}I. For the discrimination of radionuclides that suffers from isobar interference, such as ^{10}Be, a segmented anode ionization chamber for atomic number discrimination and a gas-filled magnetic spectrometer are available at the end of this AMS beamline.

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