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Study of $^{236}U/^{238}U$ ratio at CIRCE using a 16-strip silicon detector with a TOF system

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Abstract. Accelerator Mass Spectrometry (AMS) is presently the most sensitive technique for the measurement of long-lived actinides, e.g. ^{236}U and ^{x}Pu isotopes. A new actinide AMS system, based on a 3-MV pelletron tandem accelerator, is operated at the Center for Isotopic Research on Cultural and Environmental Heritage (CIRCE) in Caserta, Italy. In this paper we report on the procedure adopted to increase the ^{236}U abundance sensitivity as low as possible. The energy and position determinations of the ^{236}U ions, using a 16-strip silicon detector have been obtained. A $^{236}U/^{238}U$ isotopic ratio background level of about 2.9×10^{-11} was obtained, summing over all the strips, using a Time of Flight-Energy (TOF-E) system with a 16-strip silicon detector (4.9×10^{-12} just with one strip).

1 Introduction

Anthropogenic long-lived alpha-emitting radionuclides have been (and still are) released into the environment by nuclear tests, nuclear accidents, operations of fuel reprocessing and plant decommissioning at ultra trace level. Moreover since U stable abundant isotopes also exist in nature, the sensitivity limit for the isotopic ratio depends on the U concentration in the sample. In turn, our task is for environmental samples with sizeable amounts of U (~ 1 mg), to increase the isotopic ratio sensitivity down to the natural abundances ($^{236}U/^{238}U \sim 10^{-13}$); on the other hand, for anthropogenic influenced samples, the required sensitivity for the measurement of the isotopic composition is alleviated, but significantly smaller amounts of U may be used. The detection requires the resolution of mass spectrometry (MS) techniques - e.g. Thermal Ionization (TI-MS) or Inductively Coupled Plasma (ICP-MS) - but only Accelerator Mass Spectrometry (AMS) has the sensitivity required.

Two recent papers [1, 2] summarize the results obtained in the laboratories in the field of actinides (e.g. ^{236}U) AMS. The two systems (VERA and ANU) aiming to the best isotopic ratio sensitivity have shown that it is possible to reach an $^{236}U/^{238}U$ sensitivity of 10^{-13} in samples including about 1 mg of U. The ANSTO and LLNL laboratories quote a much lower U concentration sensitivity with an isotopic ratio of about 10^{-8}.

In the framework of a research program aiming to design, realize and characterize an actinides dedicated beam line at CIRCE, we have already performed simulations and preliminary measurements to characterize the system [3, 4]. In particular it has been shown that the addition of a switching magnet just after the electrostatic analyzer (ESA) of the CIRCE system was able to reduce the background in the $^{236}U/^{238}U$ detection from $^{236}U/^{238}U \sim 3.0\times10^{-9}$ down to $^{236}U/^{238}U \sim 5.6\times10^{-11}$. This result was obtained using only a silicon strip detector without any Time of Flight system for the identification of the rare isotopes. In this paper we present measurements performed to assess the CIRCE U isotopic ratio detection limits of the TOF-E system with 16-strip detector. To this end, an important step has been the validation of the energy and position determinations of the ^{236}U ions in the 16 strip silicon detector. This validation has been obtained verifying the energy calibration of the detector by means of a comparison with the literature pulse height defect value [5].

2 Experimental methods

2.1 CIRCE accelerator and uranium procedure

CIRCE, described elsewhere [6–8], is a dedicated AMS facility based on a 3MV-tandem accelerator. Briefly the caesium sputter ion source is a 40-sample MC-SNICS and a total injection energy of 50 keV for $^{238}U^{16}O^{-}$ molecules are mass selected by a spherical electrostatic analyzer with a bending angle of ±45° operated up to ±15 kV. The 90° double focusing Low Energy (LE) injector magnet allows high resolution mass analysis for all stable isotopes in the periodic table and the insulated stainless steel chamber can be biased up to 15 kV for beam sequencing (e.g. between $^{238}U^{16}O^{-}$, $^{236}U^{16}O^{-}$). The accelerator is contained inside

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a vessel filled with sulphur hexafluoride (SF_6) at a pressure of 6 bar and at the terminal the ions loose electrons in the Argon (Ar) stripper. At the high energy side the double focusing 90° High Energy (HE) analyzing magnet efficiently removes molecular break-up products [3, 4]. Two 45° electrostatic spherical analyzers (ESA) are operated up to ±60 kV. A switching magnet (B_{max}= 1.3 T, r=1.760 m and ME/q^2= 253 MeV amu/e^2 at the 20° exit) is positioned after the ESA. Finally the selected ions at 20° are counted in a final detector. The control of the acquisition system is handled by the Fast Intercrate Readout (FAIR) system [9] via Ethernet interfaces.

The negative molecular ions $^{238}U^{16}O^-$ are accelerated in the LE side and $^{238}U^{x+}$ positive ions are accelerated in the post stripper high energy tube of the tandem. For $^{238}U^{5+}$ this results in an energy of E = 17.3 MeV at a terminal voltage of 2.900 MV. The working pressure of the Ar is about 1.3 mTorr [4] with a stripping yield of 3.1%. The tuning procedure is made by the optimization of HE magnet and ESA in the high-energy side, maximizing the $^{238}U^{5+}$ current in the Last Faraday Cup. Once the setup for the $^{238}U^{5+}$ pilot beam is found, the voltage at the chamber of the injection magnet, the terminal voltage and the voltage of the ESA are scaled to transmit $^{236}U^{5+}$.

2.2 TOF-E detection system

A Time of Flight-Energy (TOF-E) detection system with a flight path of 1.5 m has been installed, to discriminate $^{236}U^{5+}$ ions from ^{238}U, ^{235}U and other interferences. In our set up, the start signal for the TOF measurement is given by a MCP (MicroChannel Plate) positioned about 70 cm downstream from a 4 mm collimator. The energy information is given by the 16 strip silicon detector which also provides the timing for the stop signal. The TOF-E detector setup, the MCP mounted in the electrostatic mirror configuration, the COSY Infinity [10] simulation of the electrons that reach the MCP and the calculation of the detection of the TOF-E detector for ^{235}U, $^{236}U^{5+}$ and ^{238}U are shown in figure 1.

Briefly (see upper left part of figure 1) ions pass through a thin LPA [11, 12] Laser Plasma Ablation carbon foil (4 μg/cm^2 with a diameter of 45 mm) placed orthogonal to the beam axis and they are detected by a 16 strip silicon detector. Electrons are emitted from the carbon foil and they are accelerated to about 1 keV in the electric field generated by an acceleration grid (wires have a diameter of 25 μm and are separated by 1 mm, i.e. the grid has a transparency of 97.5 %) that is parallel to the carbon foil (see lower left part of figure 1). The electrons are bent, by means of an electrostatic mirror, towards the MCP, with a diameter of 40 mm, placed outside the beam axis. The mirror consists of two parallel grids, 5 mm apart, placed at 45° to the beam axis. The electrostatic field between these two grids bends the electrons through 90° towards the MCP. For the electrons that exit parallel to the foil with an energy of 10 eV and $\Delta V(V_{foil}\text{-}V_{accl.})$= 1.1 kV, we calculate that the exit angle from the acceleration grid is about 95 mrad [3]. In the lower right part of figure 1, the simulation of the electrons trajectory from the acceleration grid,

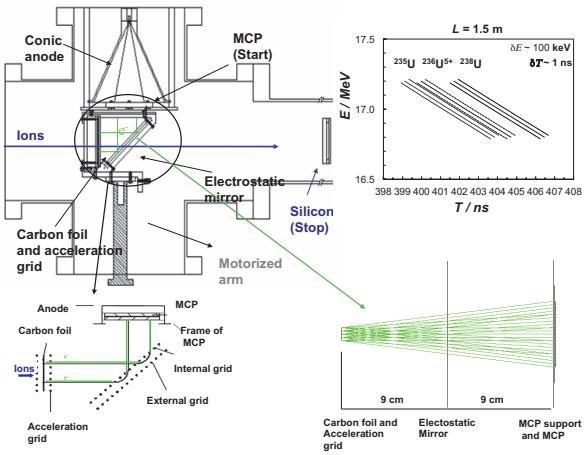

Figure 1. TOF-E system (upper left), MCP in electrostatic mirror configuration (lower left) and simulation of electron trajectories (lower right) are shown. A calculation of the spectrum is shown in the upper right part. Energy (E) vs Time of Flight (TOF) of the three different uranium isotopes are shown, where δT is the time resolution and δE is the energy spread introduced from the carbon foil, see text.

where the beam spot is assumed to be 8 mm in diameter, to the 40 mm diameter MCP is shown. There are no problems to collect all the electrons.

2.2.1 Energy validation

An energy and position validation is always an important step in ion detections, especially for heavy ions. For charged particles, silicon detectors are commonly adopted because of their simplicity. An energy resolution as low as 0.5% can be achieved for light ions and for very heavy ions, such as uranium, a much worse resolution is achieved (about 15%) due to the energy loss straggling in the dead layers at the detector surface. In addition, the high charge density leads to incomplete charge collection giving the well known pulse height defect. The experimental CIRCE results are illustrated in figure 2. The silicon detector has a linear response for light ions but not for the heavy $^{236}U^{5+}$. The signal from the 17.3 MeV $^{236}U^{5+}$ ions is equivalent to that from a light ion with an energy of only 10.2 MeV. This gives a pulse height defect of 6.9±0.7 MeV in agreement with the literature [5].

3 $^{236}U/^{238}U$ measurement results

3.1 $^{236}U/^{238}U$ sensitivity limits with 16-strip silicon detector

The two dimensional TOF-E spectrum of the silicon strip with the highest $^{236}U^{5+}$ counts (number 11 in figure 4) for the KkU VERA in house U standard $^{236}U/^{238}U$= (6.98±0.32)×10^{-11} [13], is shown in figure 3a. A 4 mm collimator was positioned at the image point of the ESA, i.e. the beam waist after the switching magnet to restrict the number of ^{238}U and ^{235}U background ions that could

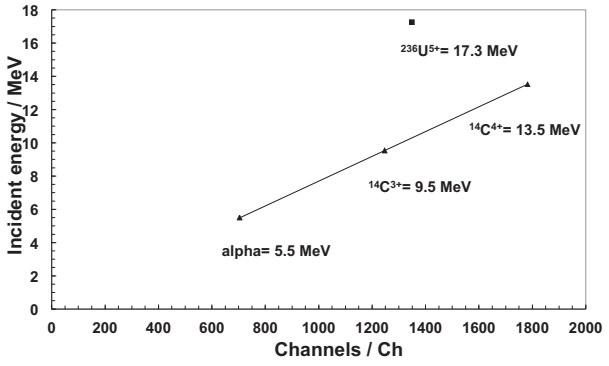

Figure 2. The three triangles are respectively alpha-particles at 5.5 MeV, $^{14}C^{3+}$ at 9.5 MeV and $^{14}C^{4+}$ at 13.5 MeV. The square is 17.3 MeV $^{236}U^{5+}$ ions. A regression line is calculated E_I= 0.0075·Ch + 0.2399 and the difference between the incident energy of the ions and the energy calculated by the regression line is zero for the light ions and not for the 17.3 MeV $^{236}U^{5+}$ ions, see text.

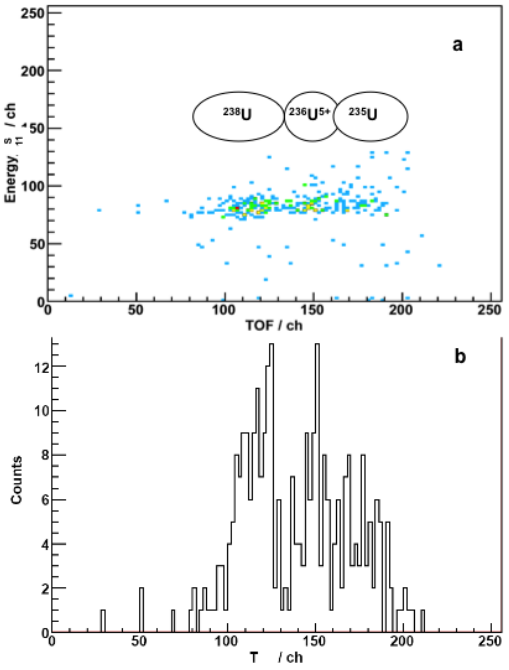

Figure 3. In the figure is shown the matrix of the TOF-E detector obtained from one strip of the 16 strip silicon detector. The y-axis represents the energy of the ions as measured by the 16 strip silicon detector and the x-axis is T=t_d-TOF where t_d is the delay applied to the MCP signal, 1 ch= 80 ps. In the figure 3b the projection of the previous matrix on the T axis is shown, see text.

reach the Si detector. A one-dimensional projection on to the time axis is shown in figure 3b.

The time calibration and the resolution of the time (T) spectrum were obtained with a $^{234}U^{5+}$ pure beam from the KkU sample. A gaussian shape was observed and the characteristics of the TOF-E system are: time resolution of 1.6 ns and energy resolution of about 15 %. In the figure 3 is clear that the two spots of $^{236}U^{5+}$ and ^{238}U are separated, while it is more difficult to separate the ^{235}U spot from $^{236}U^{5+}$ one. The ^{238}U and ^{235}U contributions to the $^{236}U^{5+}$ counts, obtained with a gaussian fitting, are negligible considering the ^{235}U tail and less than 7% for the ^{238}U one; this provides a background level isotopic ratio of about 4.9×10^{-12}. The ^{238}U and ^{235}U interference peaks are mainly due to ions that undergo charge exchange process [3, 14]. For a flight path of 1.5 m, the time difference between different interference peaks are $\Delta T(^{236}U^{5+}\text{-}^{235}U)\sim$ 1.7 ns, $\Delta T(^{238}U\text{-}^{236}U^{5+})\sim$ 3.4 ns for the same p/q (p= momentum and q= charge state of the ions), and $\Delta T(^{236}U^{5+}\text{-}^{235}U)\sim$ 0.8 ns, $\Delta T(^{238}U\text{-}^{236}U^{5+})\sim$ 1.7 ns for the same E/q (E= energy and q= charge state of the ions). From the separation of the peaks in figure 3, the majority of the ^{235}U and ^{238}U are due to ions with the same p/q, although smaller contributions due to ions with the same E/q cannot be ruled out. A significant improvement in time resolution would be required to separate the two contributions. As a matter of fact, summing over all the strips, in the $^{236}U^{5+}$ projection, the extrapolated tails of the ^{235}U and ^{238}U are respectively about 2% and 40% of the $^{236}U^{5+}$ counts. This means that using the present TOF-E system, the background level is reduced to an equivalent isotopic ratio of about 2.9×10^{-11}.

Moreover this result was interpreted, besides the 1.6 ns time resolution, as a consequence of the angular straggling due to the thickness of the carbon foil, which deteriorates the spatial separation of the interfering ions with respect to $^{236}U^{5+}$. For 99% of particles scattering angles between 20 and 30 mrad were calculated, with 90% probability angles less than 10 mrad. This gives a distribution strips number that is about 2 times (~ 8 strips) of what we have obtained in [4]; a distribution of the ^{235}U, $^{236}U^{5+}$ and ^{238}U ions, fitted value of figure 3b, along the silicon strip are shown in figure 4, in order to better understand the background origin simulating them back from the end point (silicon detector). The interfering ions that were out of the trajectory of the $^{236}U^{5+}$ are brought back inside the $^{236}U^{5+}$ peak by the 4 μg/cm^2 carbon foil thickness. Measurements with a thinner DLC carbon foil (0.6 μg/cm^2) and/or a longer path (3 m) are planned in the near future. For L= 3 m the time difference $\Delta T(^{236}U^{5+}\text{-}^{235}U)\sim$ 1.7 ns and $\Delta T(^{238}U\text{-}^{236}U^{5+})\sim$ 3.4 ns, for the same E/q. The divergence of the beam for particles passing a carbon foil of 0.6 μg/cm^2 thickness, calculated by SRIM [3], is 5 times smaller then the value calculated for the 4 μg/cm^2 thickness, i.e. 99% of particles have scattering angles less then 10 mrad and 90% scatter through less than 2 mrad. This alleviates the angular problem as well as the energy one, and for this thinner DLC foil, the relative time spread (next section) introduced by the foil is 10 times lower, i.e. 0.01 %, 0.04 ns for 17 MeV ions and 1.5 m flight path.

3.2 TOF-E time resolution

The time resolution (in this case 1.6 ns) is constituted from three contributions: the energy spread introduced from the

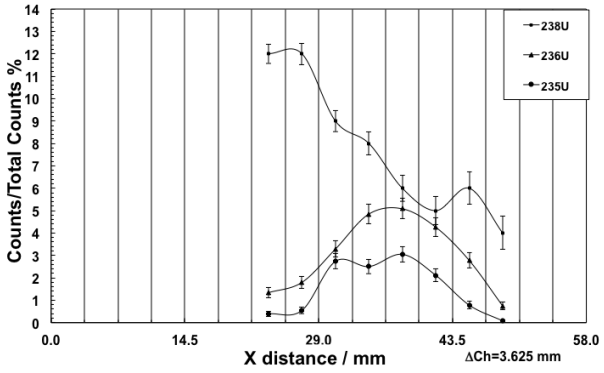

Figure 4. In the figure the distribution of the ^{238}U, $^{236}U^{5+}$ and ^{235}U along the 16 strip detector is shown when the collimator is inserted in such a way to better understand the background (see text).

thickness of the carbon foil, the spread of the beam itself and the electronics. The relative total time spread is of the order of 0.4 %. The energy spread calculated by SRIM [15], introduced from the 4 μg/cm^2 carbon foil, is about 33 % of the energy loss. The energy loss is about 30 keV/μg/cm^2 for 17 MeV particles and an energy spread of about 40 keV is obtained. The relative energy spread is about 0.2% giving a relative time spread of 0.1 %, i.e. 0.4 ns for 17 MeV ions and 1.5 m flight path. The spread of the beam itself, introduced from the ESA resolution is of the order of 0.05%, i.e. 0.2 ns for 17 MeV ions and 1.5 m flight path. The last contribution is due to the electronics and to the intrinsic resolution of the system, which can be improved in the near future with more rigorous attention to the optimization of the individual circuit components.

4 Summary

In this work it has been shown that using the actinide line an abundance sensitivity background level of about 2.9×10^{-11} on ^{236}U/^{238}U isotopic ratio can be reached using a Time of Flight-Energy system; an isotopic ratio of 4.9×10^{-12} was obtained just with one strip. The CIRCE laboratory is close to the two systems (ANU and VERA) that provide the best ^{236}U/^{238}U isotopic ratio sensitivity.

Moreover, with this upgrade, the idea to use the one dimensional information of the TOF-E detector is particularly useful to better understand the trajectory of the ions, simulating, in the near future, them back from the end point (silicon detector). An upgrade of the CIRCE TOF-E system is planned, with a flight path of about 3 m and a thinner (0.6 μg/cm^2) DLC carbon foil.

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