OPTIMIZATION OF ^{236}U AMS AT CIRCE

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ABSTRACT. Actinide isotopes are present in environmental samples at ultra-trace levels (^{236}U concentration is quoted to be on the order of pg/kg or fg/kg). Their detection requires the resolution of mass spectrometry (MS) techniques, but only accelerator mass spectrometry (AMS) has the sensitivity required. In order to perform the isotopic ratio measurements of actinides, such as ^{236}U/^{238}U, an upgrade of the Center for Isotopic Research on Cultural and Environmental Heritage accelerator (CIRCE) in Caserta, Italy, has been performed. The system was originally equipped for radiocarbon AMS measurements. The main difficulty of AMS measurement of ^{236}U is the intense neighboring beam of ^{238}U. Although most of the ^{238}U ions are suppressed by means of magnetic and electrostatic elements, a small fraction of this intense beam can interfere with the rare isotopes. This paper reports the preliminary results of the ^{236}U/^{238}U isotopic ratio measurement limit ($<5.6 \times 10^{-11}$), aimed also to better understand the origin of background ions. For this purpose, a large 16-strip silicon detector providing spatial resolution has been used. In addition, calculations to assess the performance of the system obtained by adding a high-resolution time of flight-energy (TOF-E) detector are discussed.

INTRODUCTION

The processing of material for nuclear power plants (NPPs) and nuclear weapons (NW) together with all the activities correlated with production, transport, and reprocessing of nuclear fuel, has led to the release of a wide range of radioactive elements in the environment, such as uranium, plutonium, and fission and activation products. Since the relative concentrations of uranium and plutonium can vary from site to site, depending on the nature of the source material and on its subsequent irradiation history, all these sources do not contribute equally to the contamination in a given site. The long-lived radionuclide ^{236}U (the half-life of ^{236}U is 2.3×10^7 yr) is present in environmental samples at ultra-trace levels. Measurement of these isotopic ratios requires mass spectrometric techniques, but only AMS offers the sensitivity needed to measure ^{236}U/^{238}U at natural levels (10^9 to 10^{13}) because of its capability to suppress the background of isobaric molecules such as ^{235}UH$^+$.

Conventional mass spectrometry, like thermal ionization (TI-MS) or inductively coupled plasma mass spectrometry (ICP-MS), suffers from isobaric interferences of hydrides, ^{235}UH$^+$, and from ^{238}U; the detection limits of these techniques for ^{236}U/^{238}U are 10^{-10} and 10^{-7}, respectively. Recently, increasing attention is being paid to the determination of ^{236}U/^{238}U at the level typically present in the environment (Fifield 2008; Quinto et al. 2009; Sakaguchi et al. 2009); these studies have demonstrated the potential of ^{236}U to serve as a tracer for geomorphologic processes and as sensitive fingerprints of releases from nuclear facilities.

In nature, U stable abundant isotopes exist. For this reason, the sensitivity limit for the isotopic ratio depends on the U concentration in the sample. Thus, our task is, for environmental samples, to push the sensitivity in the isotopic ratio measurement down to natural abundances (^{236}U/^{238}U $\sim 10^{-12}$) in samples with sizeable amounts of U (~1 mg). On the other hand, for anthropogenically influenced samples, the required sensitivity for the measurement of the isotopic composition is alleviated, but significantly smaller amounts of U have to be used (down to 1 ng).

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Actinides AMS measurements were pioneered at the IsoTrace laboratory in Toronto (Zhao et al. 1994, 1997), where the ^{236}U content in an uranium ore was determined using the 1.6MV AMS system. Then, at the Australian National University (Fifield et al. 1996, 1997) the use of a higher terminal voltage (4 MV) improved the sensitivity of the method, both for the detection limit as the minimum detectable number of U atoms in the sample, and for the lower limit of isotopic ratio measurable in samples at high concentration. Similar detection systems have been developed at the Lawrence Livermore National Laboratory, USA (Brown et al. 2004), at ANSTO, Australia (Hotchkis et al. 2000), at the Vienna Environmental Research Accelerator (VERA) (Steier et al. 2002) and, at much lower energies, at the ETH in Zurich (Wacker et al. 2005). Two recent review papers (Fifield 2008; Steier et al. 2010) summarize the results obtained in the laboratories active in the fields of actinides AMS. Summarizing, the 2 systems aiming for the best isotopic ratio sensitivity (VERA and ANU) have shown that it is possible to reach a sensitivity of 10^{-12} for ^{236}U in samples including about 1 mg of U. The ANSTO and LLNL laboratories quote a sensitivity concentration of about 1 ng of U with isotopic ratio of ~10^{-8}. In this paper, we report the preliminary results of the ^{236}U/^{238}U isotopic ratio measurement limit obtained so far ($<5.6 \times 10^{-11}$), aimed, also, to understand the origin of background ions. A large 16-strip silicon detector providing spatial resolution has been used for this purpose.

CIRCE AMS FACILITY AND ELEMENTS OPTIMIZATION

The +3MV CIRCE tandem accelerator of Pelletron type (Terrasi et al. 2008), similar to others installed in the past years in Europe and USA, is shown in Figure 1. Since the installation of CIRCE in 2005, we have extended the measuring capabilities of the original system.

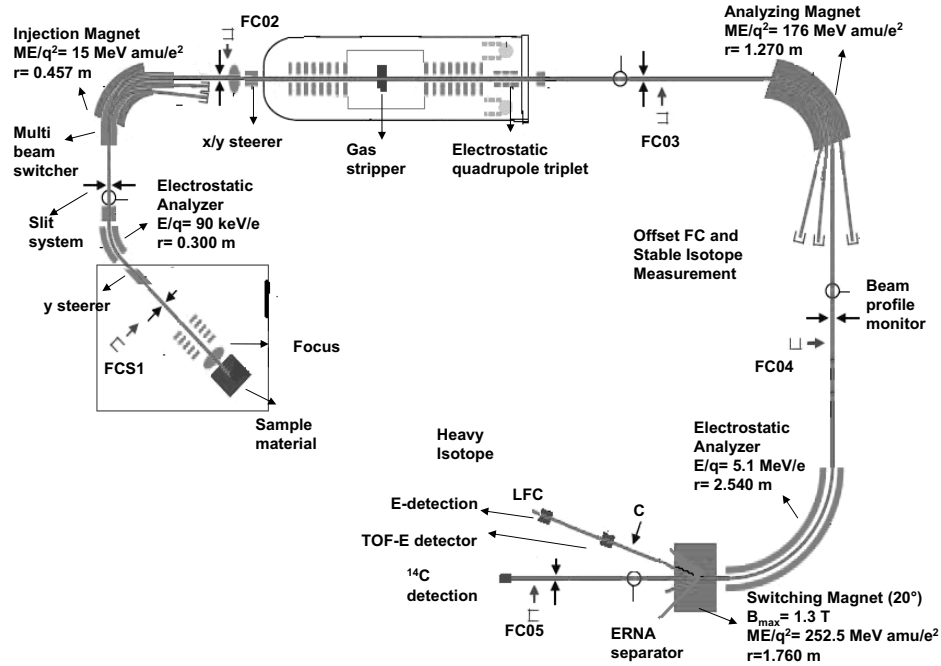

Figure 1 Schematic layout of the CIRCE accelerator and of the accelerator upgrade with the new heavy isotope detection layout: the switching magnet already inserted and the start and the stop TOF-E detector, not yet inserted (LFC in the heavy isotope line is the last Faraday cup). C denotes the collimator in the heavy isotope line, and the arrows indicate a slit system. ERNA is the acronym of European Recoil separator for Nuclear Astrophysics.

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The cesium sputter ion source is a 40-sample MC-SNICS. A typical output from 1-mm-diameter samples pressed in Al cathodes for $^{12}C^-$ ions is 30 μA at 6-kV cathode voltage and a total injection energy of 67 keV and 0.3 μA for $^{238}U^{16}O^-$ molecules with a total injection energy of 50 keV. These ions are energy selected by a ±45° spherical electrostatic analyzer (r = 30 cm, plate gap = 5 cm) that is operated up to ±15 kV. Maximum electric field strength is 6 kV/cm, resulting in an energy/charge state ratio of 90 keV/e. The 90° double-focusing low-energy (LE) injection magnet (r = 0.457 m, vacuum gap = 48 mm, ME/q^2 = 15 MeV amu/e^2) allows high-resolution mass analysis for all stable isotopes in the periodic table. The insulated stainless steel chamber can be biased up to 15 kV for beam sequencing (e.g. between ^{12}C, ^{13}C, ^{14}C or between $^{238}U^{16}O^-$, $^{236}U^{16}O^-$).

The accelerator is contained inside a vessel filled with sulfur hexafluoride (SF_6) at a pressure of about 86 psig. The terminal voltage (TV) is achieved and stabilized by GVM feedback on the charging system high-voltage supply. The TV can be kept constant at 3.000 MV with a ripple of 1 kV. In the stripper, argon (Ar) is recirculated by 2 turbopumps; the working pressure is ~7 mTorr for $^{14}C^{3+}$ at 2.550 MV and ~1.3 mTorr for $^{238}U^{5+}$ at 2.875 MV. Figure 2 shows the scan of the gas stripper pressure while measuring $^{238}U^{5+}$ ion current in Faraday cup FC04 (located right after the high-energy magnet), injecting $^{238}U^{16}O^-$ at TV= 2.875 MV.

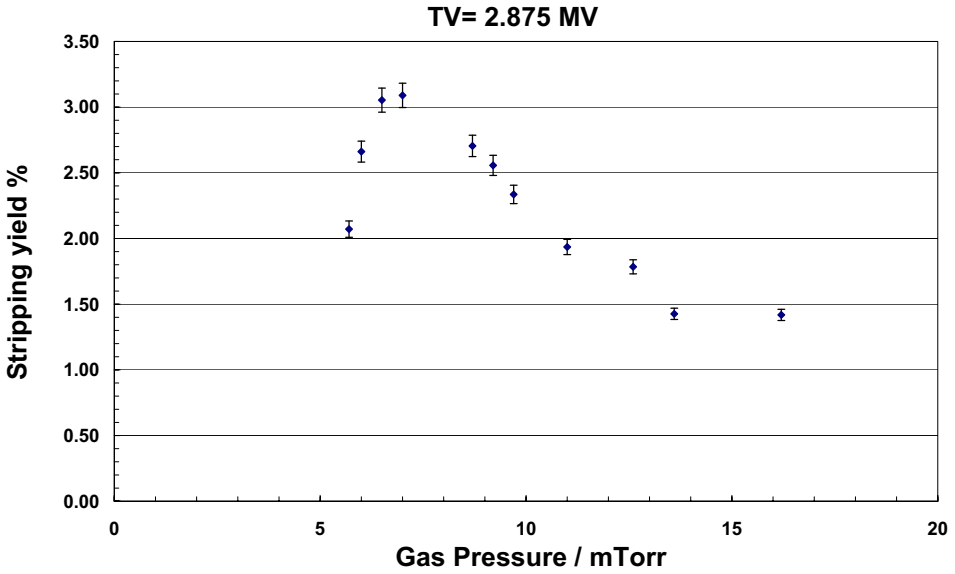

Figure 2 Scan of the gas stripper pressure while measuring $^{238}U^{5+}$ ion currents in the Faraday cup FC04, injecting $^{238}U^{16}O^-$. The terminal voltage of the accelerator is 2.875 MV. The stripping yield is the ratio between the $^{238}U^{5+}$ FC04 divided for the 5+ charge state current and $^{238}U^{16}O^-$ FC02 current. An offset of ~6 mTorr is visible.

The double-focusing 90° high-energy (HE) bending magnet has r = 1.27 m, ME/q^2 = 176 MeV amu/e^2, and $M/\Delta M$ = 725, with a slit opening of ±1 mm both at object and image points, so that e.g. $^{236}U^{5+}$ at 3 MV can be analyzed. The two 45° electrostatic spherical analyzers (r = 2.54 m and gap = 3 cm) are operated up to ±60 kV; energy resolution is $E/\Delta E$ = 700 for typical beam size. Finally, the selected ions are counted in a surface barrier detector. Control of the acquisition system is handled by the Fast Intercrate Readout (FAIR) system (Ordine et al. 1998) via ethernet interfaces.

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Low- and High-Energy Magnet Scans

The scan of the LE magnet while measuring positive ion currents in Faraday cup FC03 (De Cesare et al. 2010) is usually performed using an uranium oxide sample (U_3O_8). The $^{238}U^{16}O^-$ peak (mass 254) yields the highest current, while for ^{236}U measurements the injected mass is 252 ($^{236}U^{16}O^-$). In this case, the strongest contribution to the observed peak is due to $^{238}U^{14}N^-$.

In Figure 3, scans of the HE magnet while injecting $^{238}U^{16}O^-$ and $^{236}U^{16}O^-$, respectively, are shown. The currents were measured in Faraday cup 04. The ordinate on the left side of the figure corresponds to the $^{238}U^{16}O^-$ injected mass. Besides the peaks, which correspond to ^{238}U charge states from 5+ to 11+, one clearly observes measurable currents originating from ^{238}U ions that underwent charge exchange reactions. The ions that have undergone charge exchange after the acceleration tube are shown as peaks. In addition, there is also a continuum of ions that underwent charge exchange in the HE tube (Betz 1972; Vockenhuber et al. 2003); those ions are not visible in our FC04. It is interesting to note how the intensity of the charge exchange peaks are lower with respect to that reported in De Cesare et al. (2010); the ratio between ($^{238}U^{3+}$–> $^{238}U^{4+}$)/$^{238}U^{5+}$ is decreased by a factor of 2. This is mainly due to the better vacuum before the HE magnet, 9×10^{-9} Torr, with respect to the previous 3.5×10^{-7} Torr (De Cesare et al. 2010). The *y* axis on the right side of Figure 3 shows the current in FC04 when mass 252 was injected (i.e. $^{236}U^{16}O^-$ or $^{238}U^{14}N^-$), scaling the TV to 2.900 MV, which gives to $^{236}U^{5+}$ the same magnetic rigidity as $^{238}U^{5+}$ when injecting $^{238}U^{16}O$ at TV = 2.875. If Figure 3, it is possible to see the peaks, which correspond to charge states from 5+ to 8+ of ^{238}U coming from $^{238}U^{14}N^-$. Since the $^{236}U^{5+}$ peak when mass 252 is injected has a magnet field value very close to the $^{238}U^{5+}$ peak (~70 G), and since the $^{238}U^{5+}$ will undergo similar charge changing processes in the accelerator tube as those originating from $^{238}U^{16}O^-$, some exchanged ions of ^{238}U can pass the HE magnet and reach the final detector.

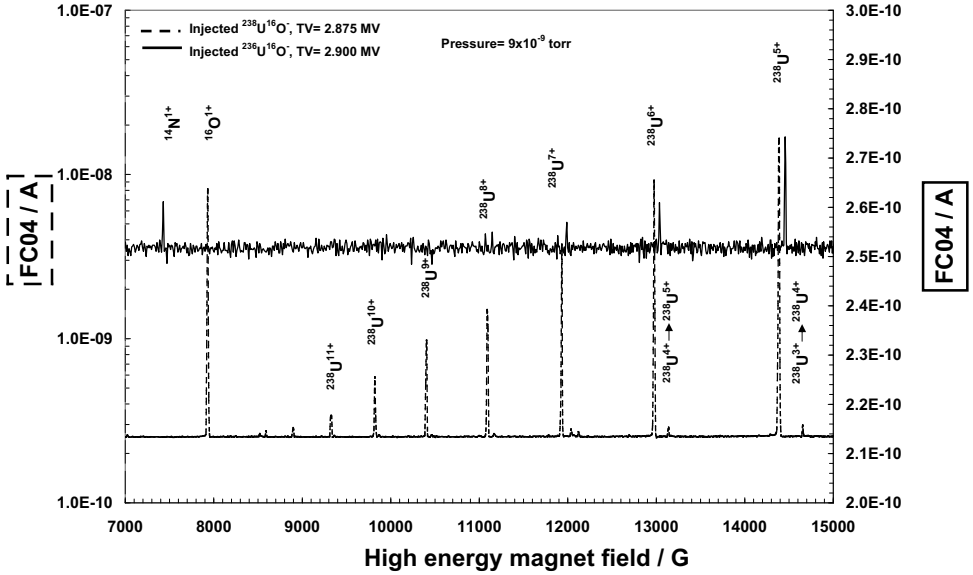

Figure 3 Positive ion current versus high-energy magnetic field. The scan of the high-energy magnet was monitored in FC04. The dashed line corresponds to the injection of $^{238}U^{16}O^-$ (left *y* axis, logarithmic scale), the solid line to $^{236}U^{16}O^-$ (right *y* axis, linear scale). The terminal voltage was set to TV = 2.875 MV when injecting $^{238}U^{16}O^-$ and it was set to TV = 2.900 MV when injecting $^{236}U^{16}O^-$. The $^{14}N^{1+}$ peak from injection of $^{238}U^{14}N^-$ at the same time as $^{236}U^{16}O^-$ is clearly visible (see text).

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16-STRIP SILICON DETECTOR MEASUREMENTS

Following the results of the simulations reported in De Cesare et al. (2010), a switching magnet (B_{max} = 1.3 T, r = 1.760 m, and ME/q^2 = 252.5 MeV amu/e^2 at the 20° exit) was installed, which provides both the option for several dedicated beamlines and, in the heavy isotope beamline, an additional purification stage.

Figure 4 shows the results of measurements performed with a position-sensitive silicon detector with an active area of 58 × 58 mm^2, positioned 1.9 m downstream the 4-mm-diameter collimator, positioned in turn 80 cm from the switching magnet. To measure the ^{236}U/^{238}U ratio, a tuning of the transport elements up to the final detector is performed in order to maximize the ion optical transmission. The ^{236}U events are counted in the final detector and ^{238}U current is measured in FC04.

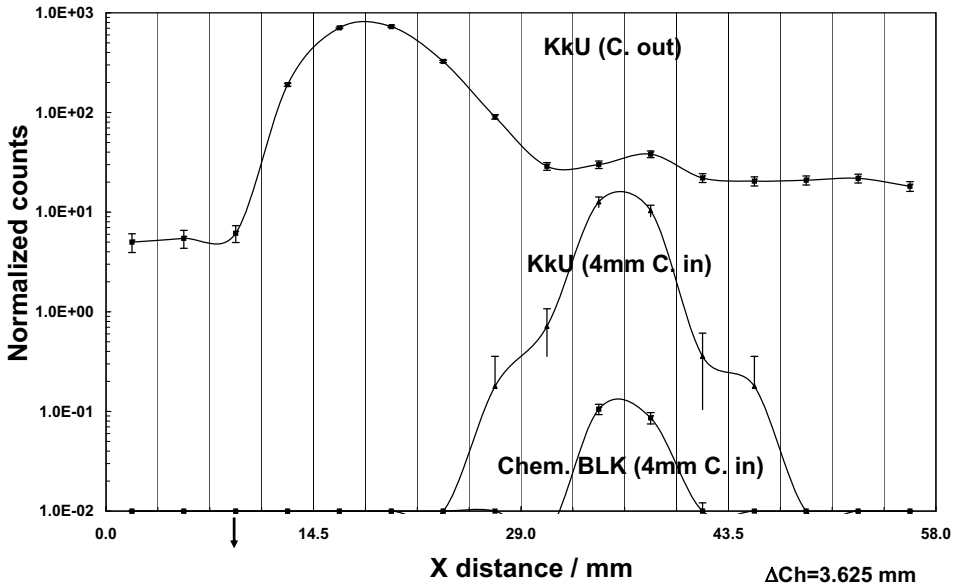

Figure 4 Normalized counts (counts in the detector in 300 s over FC04 current corrected for the transmission ~80% between FC04 and LFC) versus horizontal position of the 16-strip silicon detector. ΔCh = 3.625 mm is the distance between the center of 2 adjacent strips. The ^{238}U peak is clearly visible in the KkU (collimator out) spectrum to the left of the position of the ^{236}U peak (see text). The arrow indicates that the normalized counts are lower than 1 × 10^{-2} counts/nA.

Tuning is done by setting the parameters of the beam line of the ^{238}U current to the last Faraday cup (LFC) positioned in front of the detector, using as target material uranium oxide (U_3O_8) obtained from uranyl nitrate ($UO_2(NO_3)_2 \cdot 6\,H_2O$) baked at 800 °C. This material originated from the uranium mines Kk Uranfabric Joachimisthal, and is the VERA in-house U standard. In our case, the sample preparation provides uranium in the form U_xO_y+Fe_2O_3 from which $^{x}U^{y}O^{-}$ is sputtered. To select different injected masses, the injection magnet bouncing system is used. The stripping yield for $^{238}U^{5+}$ achieved by the Ar gas stripper of CIRCE is ~3.1% (Figure 2). At 3 MV terminal voltage, ME/q^2 is ~170 MeV amu/e^2 for $^{238}U^{5+}$ and ME/q^2 is ~221 MeV amu/e^2 for $^{238}U^{4+}$; since the double-focusing HE magnet reaches a maximal ME/q^2 = 176 MeV amu/e^2, the 5+ represents the lowest charge state that can be bent by the HE magnet. For heavy ion measurements, the object and image slits of the injection magnet are closed to ±1 mm, the slits of the HE magnet are closed to ±2 mm, and a collimator of 4 mm diameter is positioned just at the waist of the ESA. The current transmission between

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FCO4 and LFC is ~80%. Once the setup for the pilot beam $^{238}U^{5+}$ is found, the voltage at the chamber of the injection magnet, the terminal voltage and the voltage of the analyzer ESA are scaled to the other wanted mass ($^{236}U^{5+}$).

The preliminary measurements for the isotopic ratio $^{236}U/^{238}U$ were performed by cycling between ^{238}U, which is measured by means of the current in the Faraday cup just after the HE magnet FC04, and ^{236}U, which is measured in the final 16-strip surface barrier detector. We measured 2 samples: one with a nominal ratio $^{236}U/^{238}U \sim 10^{-8}$, obtained by adding a spike of ^{236}U to the KkU VERA in-house U standard, and the KkU itself with $^{236}U/^{238}U = (6.98 \pm 0.32) \times 10^{-11}$ (Steier et al. 2008). In Figure 4, the points represent the counts in different positions of the 16-strip silicon detector, obtained with two KkU samples, KkU(C.out) and KkU(4mmC.in), and with a "chemistry blank" prepared by the same treatment as the other two. The contribution of the latter is ~1% of the KkU sample signal. Moreover, comparison with the spatial distribution obtained when the collimator was removed shows that interference from ions of higher magnetic rigidity with respect to $^{236}U^{5+}$ are largely suppressed by the 4-mm collimator; one can estimate that the residual interference background below the $^{236}U^{5+}$ peak, obtained as the relative difference between the sum of the 4 central strips of the KkU (4 mm C. out) and KkU (4 mm C. in), see Table 1, is <80% of the total, which is in turn the interference measured without the collimator. That indicates that "interference + chemistry" background in our $^{236}U/^{238}U$ measurements is $<5.6 \times 10^{-11}$. This value has to be compared with the previous result (3×10^{-9}, De Cesare et al. 2010) obtained at 0°, before the installation of the switching magnet. Figure 5 shows the comparison of the strips distribution obtained with the collimator in the position, using the "spike" and the KkU samples. The ratio between the sum of the integrated number of the 6-strip counts (Table 1) is 125 ± 12, yielding a lower limit of $(9 \pm 1) \times 10^{-9}$ for the spike sample isotopic ratio, in agreement with the expected value. These results show that with a background $<5.6 \times 10^{-11}$, we are able to measure samples with an isotopic ratio value $\geq 10^{-10}$, without the use of a TOF-E system.

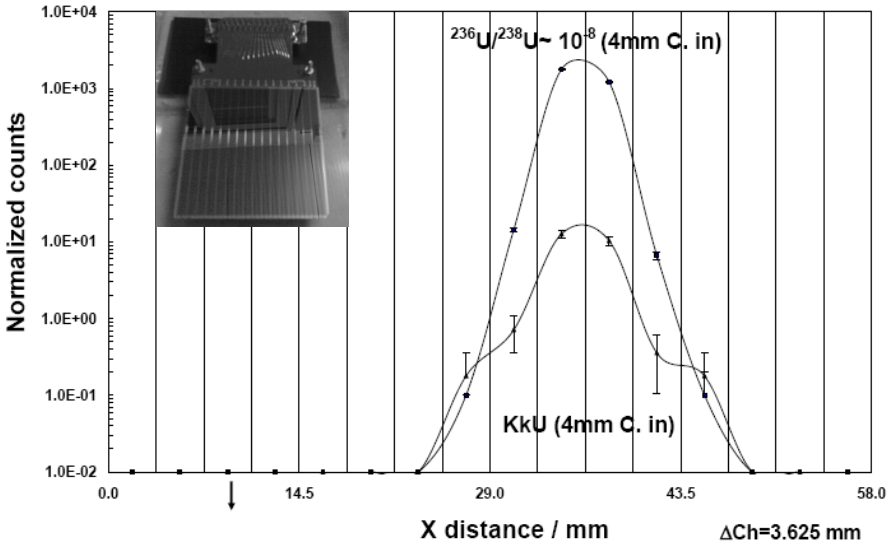

Figure 5 Normalized counts (counts in the detector in 300 s over FC04 current corrected for the transmission ~80% between FC04 and LFC) versus horizontal position of the 16-strip silicon detector. ΔCh = 3.625 mm is the distance between the center of 2 adjacent strips. A photo of the 16-strip detector is also shown. The bigger peak represents the position on the detector of the ^{236}U obtained with a spike sample; the nominal ratio is $^{236}U/^{238}U \sim 10^{-8}$. The lower ^{236}U peak is obtained with the KkU VERA in-house U standard (see text). The arrow indicates that the normalized counts at that position are lower than 1×10^{-2} counts/nA.

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Table 1 Measured raw strips data for the measured samples: KkU (4 mm C. in), Chem.BLK (4 mm C. in), KkU (4 mm C. out) and $^{236}U/^{238}U{\sim}10^{-8}$ (4 mm C. in) with the relative errors bar. The FC04 current for each cathode is also shown.

		$^{236}U^{5+}$ counts in 300 s in the STRIPS SILICON DETECTOR/FC04 current															
Cathode type	$^{238}U^{5+}$FC04 current (nA)	St. 0	St. 1	St. 2	St. 3	St. 4	St. 5	St. 6	St. 7	St. 8	St. 9	St. 10	St. 11	St. 12	St. 13	St. 14	St. 15
KkU (4 mm C. in)	2.8								0.2	0.7	13	10	0.4	0.2			
Errors									0.2	0.4	2	1	0.3	0.2			
Chem.BLK (4 mm C. in)	~1% of KkU (4 mm C. in)									0.006	0.11	0.09					
Errors										0.003	0.01	0.01					
KkU (4 mm C. out)	2.4	5	5	6	190	707	726	325	90	29	30	38	22	20	21	22	18
Errors		1	1	1	7	13	13	9	5	3	3	3	2	2	2	2	2
$^{236}U/^{238}U{\sim}10^{-8}$ (4 mm C. in)	6.8								0.1	14	1779	1218	6.7	0.1			
Errors									0.1	1	11	10	0.7	0.1			

Further investigations are needed to identify the amount of ^{238}U that is under the peak of $^{236}U^{5+}$ and the amount of ^{235}U, which is also present. A time of flight-energy (TOF-E) detection system (De Cesare et al. 2007, 2010) will be used, to discriminate between ^{236}U, ^{238}U, and ^{235}U, with the start signal for the TOF measurement given by a MCP (MicroChannel Plate), in electrostatic mirror configuration, positioned about 70 cm from the 4-mm collimator. The energy information is given by the 16-strip silicon detector, which also provides the timing for the stop signal. With an energy resolution of δE ~100 keV (substantially given from the spread of the beam itself and from the spread introduced from the 4 μg/cm^2 laser plasma ablation [LPA; Maier-Komor et al. 1997, 1999]) carbon foil and time resolution δT ~1 ns, for a flight path of L = 1.5 m, the different interference peaks can be resolved for ^{238}U and $^{236}U^{5+}$, since $\delta T(^{236}U^{5+}\_^{235}U)$~0.8 ns and $\delta T(^{238}U\_^{236}U^{5+})$~1.7 ns, for the same energy.

SUMMARY AND CONCLUSIONS

The main upgrade so far has been the addition of a switching magnet placed 50 cm after the exit of the ESA. The ratio $(^{238}U^{3+}\text{–}> {}^{238}U^{4+})/^{238}U^{5+}$ is decreased by a factor of 2 via improvement in the vacuum between the accelerator and the HE magnet. The preliminary results, using a 16-strip silicon detector, have shown that, in the upgraded CIRCE heavy ions beamline after the switching magnet installation, a background level $<5.6 \times 10^{-11}$ has been reached, compared to 3×10^{-9} obtained previously. The CIRCE laboratory is more that 1 order of magnitude higher with respect to the 2 systems (ANU and VERA) providing the best $^{236}U/^{238}U$ isotopic ratio sensitivity of 10^{-12}, in samples including about 1 mg of U. We aim to reaching this goal with the utilization of a TOF system. An overview of the upgraded CIRCE facility has been described using a TOF-E system, with a flight path of 1.5 m, aiming to reach the necessary $^{236}U/^{238}U$ isotopic ratio sensitivity.

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