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Actinides AMS at CIRCE and ^{236}U and Pu measurements of structural and environmental samples from in and around a mothballed nuclear power plant

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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 ^{239}Pu. A new actinide line is in operation at the Center for Isotopic Research on Cultural and Environmental heritage (CIRCE) in Caserta, Italy. Using the actinide line a uranium mass sensitivity of around 4 μg has been reached measuring with a 16-strip silicon detector, and a ^{239}Pu background level of below 0.1 fg has been obtained.

In this work we also discuss preliminary results for environmental and structural samples from in and around the Garigliano nuclear power plant (GNPP), presently in the decommissioning phase. Measurements on environmental samples from the vicinity of the plant allow the assessment of contamination, if any, over the years. Measurements of structural samples from the plant are relevant to the optimization of the decommissioning program for the GNPP.

1. Introduction

Long-lived anthropogenic radionuclides have been released into the environment by nuclear weapons testing, nuclear accidents, fuel reprocessing and decommissioning of NPPs. In Italy no NPPs are in operation, but the four shutdown NPPs are now being decommissioned. An overriding concern in the dismantling process is to avoid possible contamination of the site by radionuclide release.

At the GNPP several radiological assessment campaigns have been carried out using conventional radioactive decay counting techniques to determine the extent, if any, of environmental contamination by β- and γ-emitters. Long-lived α-emitters, specifically uranium-236 and isotopes of plutonium, are however difficult to measure with adequate sensitivity using α-particle counting techniques, and the ultra-sensitivity of AMS is crucial for the evaluation of any contamination from these important nuclides. In particular, the measurement of the activities in structural materials of the plant is required to inform the appropriate procedures for dismantling.

Pu isotopes and ^{236}U are currently used in a broad domain ranging from tracing nuclear releases, through nuclear safeguards, to tracing soil loss and sediment transport [1–3].

The CIRCE [3,4], located in Caserta (Italy), in collaboration with SoGIN (Nuclear Plant Management Company) started a research program to establish a highly sensitive system for measuring the concentration and isotopic ratios of U and Pu isotopes based on AMS, applicable to the analysis of both environmental and structural samples (i.e. from the reactor building and related infrastructures) to quantify and determine the origin of any U or Pu.

For ^{236}U, the measured quantity is ^{236}U/^{238}U, and the sensitivity limit for this ratio depends on the mass of uranium in the sample. To measure the isotopic ratio in environmental samples it is therefore desirable to push the sensitivity down to natural abundance levels (^{236}U/^{238}U $\sim 10^{-13}$) in samples with sizeable amounts of U ($\sim$1 mg). On the other hand, for anthropogenically influenced samples, higher ratios are expected so the required sensitivity may be relaxed, and significantly smaller amounts of U may then be used [3].

In this paper we present the performance of the CIRCE system, the sensitivity reached and the measurement results on samples from the GNPP site, obtained both at the Australian National University (ANU) [3] and at CIRCE.

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2. Results of AMS measurements

In a first phase of the project [4–6] aiming to build a beam line dedicated to actinides AMS at CIRCE based on a 3-MV AMS Pelletron tandem system, we have performed preliminary measurements to characterize the system, shown in Fig. 1. In particular it has been shown that the addition of a switching magnet just after the ESA of the CIRCE AMS system was able to reduce the background in the ^{236}U detection from $^{236}U/^{238}U \sim 3.0 \times 10^{-9}$ to $< 5 \times 10^{-11}$, even without the use of a TOF-E detection system, that is in any case planned for the near future. The determination of the background was achieved using a silicon strip detector that provides the spatial distribution of the incident ions to assist in the identification and assessment of isotopic interferences.

2.1. CIRCE accelerator: ^{236}U and Pu measurement procedures

UO or PuO molecular negative ions were extracted from an NEC 40-sample MC–SNICS cesium sputter ion source and pre-accelerated to a total injection energy of 50 keV. Typical beam currents are 50–300 nA for $^{238}U^{16}O^-$ ions. The ions are energy 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; mass resolution is $M/\Delta M \sim 500$ for a slits aperture of ±1 mm [4]. The insulated stainless steel chamber can be biased up to 15 kV for beam sequencing (e.g. between $^{238}U^{16}O^-$, $^{236}U^{16}O^-$ or between $^{239}Pu^{16}O^-$, $^{240}Pu^{16}O^-$, $^{242}Pu^{16}O^-$). An argon gas stripper in the high-voltage terminal of the accelerator is used to dissociate the molecules and strip the resulting atomic ions to high positive charge states. After acceleration, the double focusing 90° high energy (HE) analyzing magnet ($M/\Delta M = 725$ with slit opening of ±1 mm both at object and image points), efficiently removes molecular break-up products [4,5]. Subsequently, the two 45° electrostatic spherical analyzers select ions of the correct energy with an energy resolution $E/\Delta E = 700$ for a typical beam size of 3–4 mm. A switching magnet is positioned after the ESA. Finally the selected ions at 20° are counted in an appropriate detector. The control of the acquisition system is handled by the Fast Intercrate Readout (FaIR) system [7] via Ethernet or AccelNet interfaces.

Measurement of the $^{236}U/^{238}U$ ratio is effected by counting ^{236}U ions in the final detector and periodically measuring the ^{238}U current in the FC04 Faraday cup after the analyzing magnet. The transmission efficiency between the FC04 and FC5 at 20° is 80%, with a 4 mm collimator in place. A tuning of the transport elements up to the Faraday cup in front of the final detectors (LFC) is performed by setting the parameters of the beam line to the detection of ^{238}U. For $^{238}U^{5+}$ an energy of $E = 17.3$ MeV with a terminal voltage of $V = 2.900$ MV is reached. The working pressure of the Ar in the stripper is about 1.3 mTorr for $^{238}U^{5+}$ at 2.875 MV [5] and the stripping yield achieved for $^{238}U^{5+}$ is around 3.1%.

Once the setup for the pilot $^{238}U^{5+}$ beam is determined, the voltage at the chamber of the injection magnet, the terminal voltage and the voltage of the ESA are scaled to transmit $^{236}U^{5+}$.

For Pu isotopes, the system is also set up with a $^{238}U^{5+}$ pilot beam, and then the same parameters as above are scaled to transmit sequentially $^{239,240,242}Pu$.

2.2. Internal calibration and ^{236}U and ^{239}Pu mass sensitivity

2.2.1. Internal calibration

In order to test the linearity of the response of the system, a series of samples (Ratio Series) with nominal isotopic ratios from $\sim 5 \times 10^{-8}$ down to $\sim 1 \times 10^{-10}$ were prepared by mixing different

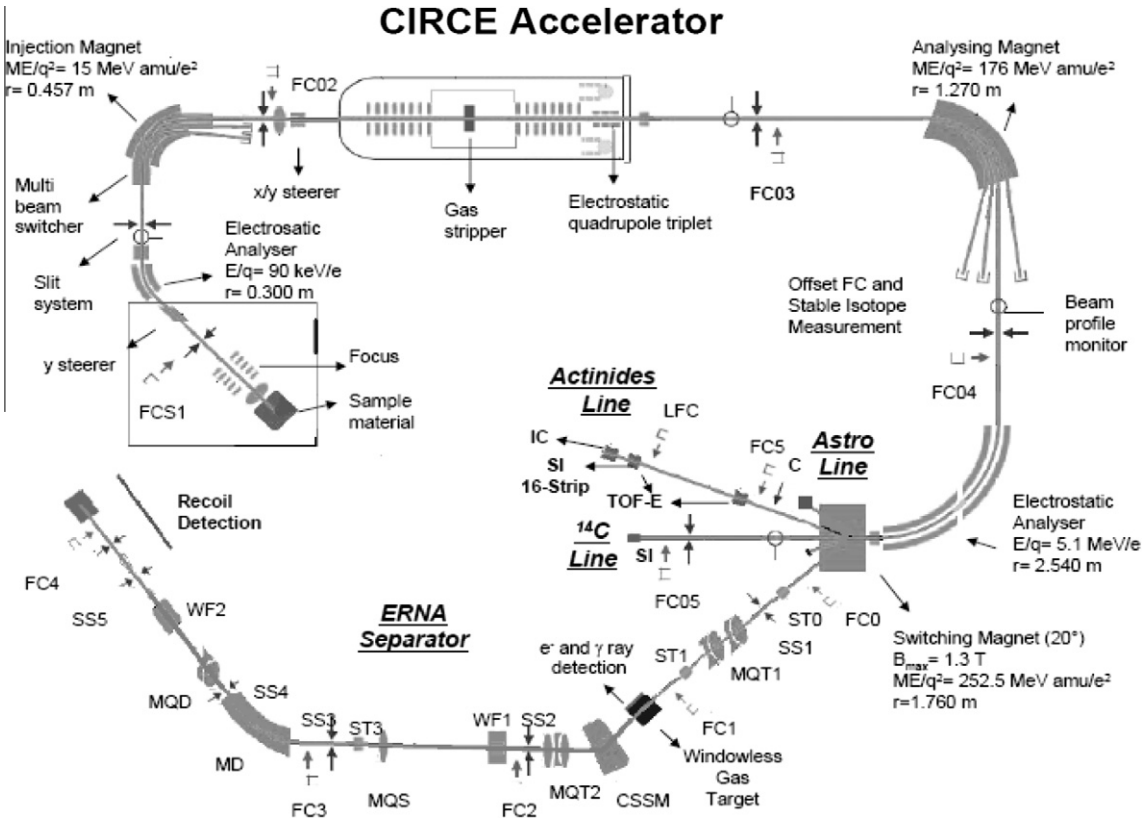

Fig. 1. Schematic layout of the CIRCE accelerator with the actinides line layout, including the switching magnet, start and stop TOF-E detector and the Ionization Chamber. FC denotes Faraday Cup; arrows indicate system slits and collimators are identified by a C. ERNA is the acronym of European Recoil separator for Nuclear Astrophysics.