Status report of the 1 MV AMS facility at the Centro Nacional de Aceleradores

Elena Chamizo Calvo [a,*], Francisco Javier Santos [a], José María López-Gutiérrez [a,b], Santiago Padilla [a], Manuel García-León [a,c], Jan Heinemeier [d], Christoph Schnabel [b], Grazia Scognamiglio [a]

[a] Centro Nacional de Aceleradores (Universidad de Sevilla, Consejo Superior de Investigaciones Científicas, Junta de Andalucía), Thomas Alva Edison 7, 41092 Seville, Spain
[b] Dpto. de Física Aplicada I, Escuela Universitaria Politécnica, Universidad de Sevilla. Virgen de África 7, 41011 Seville, Spain
[c] Dpto. de Física Atómica Molecular y Nuclear, Universidad de Sevilla, Reina Mercedes s/n, 41012 Seville, Spain
[d] AMS 14C Dating Centre, Department of Physics and Astronomy, Aarhus University, Ny Munkegade 120, DK-8000 Aarhus C, Denmark

ABSTRACT

SARA (Spanish Accelerator for Radionuclides Analysis) was the first multielemental AMS facility installed in Spain in 2005. Since then it has been dedicated to the routine analysis of several radionuclides, such as ^{10}Be, ^{14}C, ^{26}Al, ^{129}I and Pu isotopes. Tests have been carried out with other isotopes, such as ^{41}Ca, ^{236}U and ^{237}Np, and several changes have been made to the original facility to improve performance. First, an upgraded version of the ion source SO-110 has allowed us more stable measurement conditions for volatile elements (i.e. iodine), and a better general performance. Besides, changes in the target geometry have improved the ionization efficiency and long-term stability of the source output. Moreover, different software upgrades have been introduced to meet our routine operational needs. Finally, changing the movable Faraday-cup associated electronics now allows the measurement of smaller currents (in the range of pA), which has been key for the study of ^{236}U/^{238}U atomic ratio in environmental samples. Apart from these modifications it has to be noted that routine radiocarbon measurements have been moved to a Micadas system (200 kV) installed at CNA in 2012. In this paper we will illustrate the evolution of the facility up to now, and our future prospects will be introduced.

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1. Introduction

In the recent years, low-energy Accelerator Mass Spectrometry (AMS) systems, with accelerator terminal voltages below 1 MV, have become established in the AMS community. They feature with lower running costs, manpower requirements and ease of operation compared to conventional AMS facilities (accelerator terminal voltages of 3 MV and above). Among the so-called compact systems, two different types of facilities can be distinguished: (i) the ones dedicated to ^{14}C analysis and, (ii) the multielemental systems that, depending on their design also support the measurement of other AMS radionuclides. Among the first group, it is interesting to mention the 250 kV single stage system developed by National Electrostatics Corporation (NEC, USA) [1], and the 200 kV vacuum insulated terminal voltage Micadas currently supplied by Ionplus (Zürich, Switzerland) [2]. Among the multielemental facilities, NEC and High Voltage Engineering Europa (HVEE, Holland) lead the market of the commercially available compact AMS facilities.

The 1 MV AMS system installed at the Centro Nacional de Aceleradores (CNA, Seville, Spain) was designed and manufactured by HVEE in 2005 [3]. It was the prototype that set the fundamentals of a series of nine further 1 MV HVEE machines that are operative nowadays worldwide and some more to be installed. The CNA machine, called SARA (Spanish Accelerator for Radionuclides Analysis) has a very compact design (3.8×6.3 m^2) (Fig. 1). On the low energy side, a Cs-sputter ion source produces the negative ion beam. It is mass-analyzed by a 40 cm radius 90° sector magnet (low-energy magnet, LEM), and accelerated in a 1 MV Tandetron accelerator using argon gas in the terminal stripper. The beam is then analyzed for mass and energy by a 85 cm radius 90° sector magnet (high-energy magnet, HEM) and by a 120° electrostatic analyzer (ESA). The ions are finally counted and identified in a two-anode gas ionization chamber (GIC) provided with a 40 nm silicon nitride entrance window [3,4]. Later versions of this system incorporate further upgrades. For instance, the recently installed system in Aarhus, Denmark, includes a second sector-magnet on the high-energy side of the spectrometer and a dual gas system (Ar and He) for the terminal stripper [5–7].

* Corresponding author.

During the more than 8 years the 1 MV AMS system at the CNA has been in operation, our group has worked not only on the optimization of the radionuclides tested on-site by HVEE (i.e. ^{10}Be, ^{14}C, ^{26}Al, [4]), but has also explored its possibilities for the measurement of other AMS radionuclides such as ^{129}I [8] and plutonium isotopes, ^{239,240}Pu [9]. To date, these techniques are well-established and those radionuclides are measured in different matrixes on a regular basis. Very recently, this list has been extended to other actinides, such as ^{236}U, ^{237}Np and ^{241,243}Am. Besides of this, the measurement of ^{41}Ca has been investigated. Special effort has been taken to improve the ionization efficiency through the study of the sputtering geometry. Also, the software and hardware of the AMS system have been upgraded by HVEE to improve its operation. The current setup of the AMS system, the study of the optimization of the sputtering geometry, a description of the technique used for the measurements of the different radionuclides, a review of the performance of the facility, and our future prospects, are discussed in detail in the following sections.

2. The current setup of the 1 MV AMS system at the CNA

2.1. Description of the 1 MV AMS facility

The main constructive characteristics of the 1 MV AMS facility at the CNA in Seville, Spain, have been extensively described in different publications so only a brief description is given here [3,4] (Fig. 1).

It is above mentioned that its low and high-energy kinematic filters are the fundamental ones to any AMS system (a LEM, a HEM and an ESA). The Cs-sputter ion source is provided with a 200 samples carrousel. It was updated in 2011 to the new version SO-110B [10]. Due to the compact design of the system, only an extraction cone, coupled to the ion source, and an einzel lens is required for conducting the negative ion beam to the LEM, that is equipped with a fast bouncing unit. The system features a 1 MV Tandetron accelerator, using the so-called Q-Snout lens at the entrance for an optimal focusing of the beam in the gas stripper operating on argon. At its exit, a quadrupole triplet lens (Q-Pole) is used to fulfill the ion-optics requirements of the high-energy spectrometer for every charge state. No other lenses are required

for tuning the system. Two sets of vertical electrostatic steerers are placed at the entrance and exit of the accelerator to adjust the beam position. The steerer on the low-energy side serves also as an ultra-fast beam blanking unit during the fast switching of the bouncer voltage (blanking steerer). A silicon nitride foil as passive absorber at the object position of the ESA is routinely used to suppress the ^{10}B isobaric background for ^{10}Be measurements.

A movable Faraday Cup (FC3) can be set manually to the deflection position of the stable isotope at the exit of the HEM. Its design allows the measurement of the ^{127}I current when ^{129}I is counted in the detector, which are 2 cm apart at the image of the HEM. Isotopic ratios are determined using the so-called Fast Sequential Injection (FSI) method, which is conventionally called bouncing and used when a single rare and one or more abundant stable isotopes are present (e.g. the ^{127}I beam currents during a ^{129}I analysis) [11]. In our case, injection periods of 100 ms are dedicated to the rare isotope and periods of 100 μs to the stable ones.

However, the high-energy spectrometer does not support the measurement of the ^{238}U current when the HEM is set for ^{236}U, as these beams are only 1 cm apart (i.e. the HEM mass resolving power is 1300 mm/(δM/M)). Therefore, the so-called Slow Sequential Injection method (SSI) is employed. It supports sequential adjustment of the ion energy by changing the accelerator voltage and the ESA field accordingly, allowing the measurement of several isotopes in the detector (e.g. ^{242}Pu, ^{239}Pu and ^{240}Pu) while keeping the magnet settings constant. The method also supports deflection of isotope beams to a given position of FC3, e.g. for a ^{238}U measurement when FC3 is shifted away not to intercept the ^{236}U. The measurement periods are at least 2 s in our case. This SSI software was implemented by HVEE in 2008 and is routinely used for actinides measurements [9,12].

2.2. Software and hardware upgrades

Several software and hardware upgrades have been implemented in the last 3 years by HVEE. The so-called Auto Range functionality allows automatic selection of the most convenient measurement range for FC3 for each individual cathode during a programmed sequence of analyses (called a batch measurement). Once a sample is inserted into the ion source, the system waits a
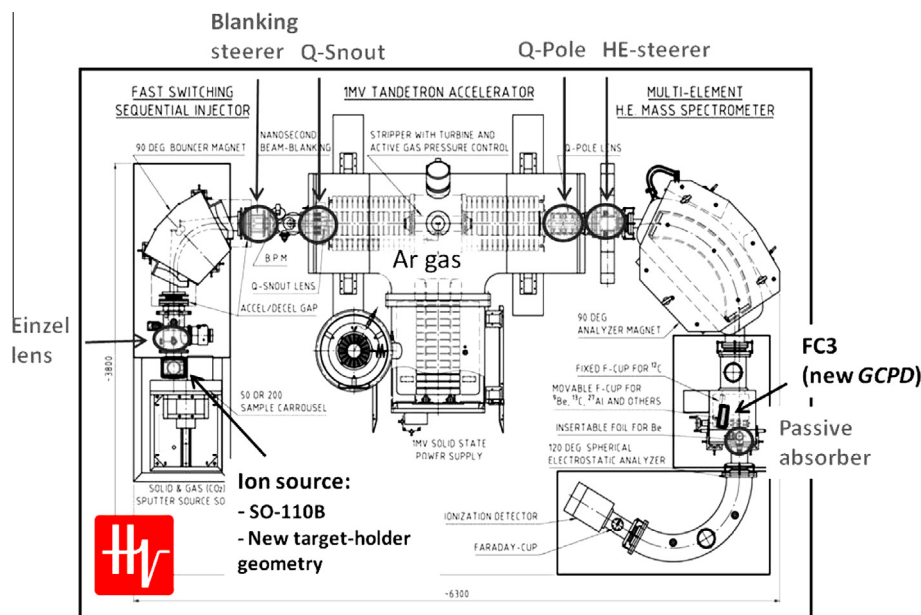

Fig. 1. Layout of the 1 MV AMS system at the CNA where the main characteristics of its design and the most recent updates are indicated [3].

few seconds for the measured current to settle, and then choses automatically the integration range that fits best. This way, overload of the so-called Gated Charge Pump Digitizers (GCPD, i.e. the electronics processing the FC3 current for measurement [13]) is avoided, and suitable scales can be used for every single current. This feature improved the measurement precision compared to the former version of the software, where the range was fixed for all samples regardless of their different output currents.

Four different integration ranges are available on our system, supporting maximum beam currents in FC3 of 10 nA, 100 nA, 1 µA and 10 µA. For the second high-energy offset Faraday cup that monitors ^{12}C during a ^{14}C measurement (FC2), the maximum currents for every range are higher by a factor of 100, following the natural isotopic ratio between ^{12}C and ^{13}C. For this upgrade, the three printed circuitry boards (PCBs) associated to the GCPD electronics were completely redesigned by HVEE in 2012. The 10 nA current range was incorporated to allow measurement of pA currents. Noise values of about 40 pA had been obtained with the former 100 nA range, which was the lowest one in the previous version of the GCPD. The newly available 10 nA range is exclusively used in SSI mode. During the FC3 current measurement, the bouncer magnet is set on a constant potential, and gating of the GCPD is avoided to keep the electronic measurement noise as low as possible. The operator can define this and other details of the measurement method in the software.

In order to study the performance of the new GCPD during a real measurement, an experiment with natural uranium samples was performed. A set of 4 cathodes with uranium contents ranging from 10 to 30 µg were prepared, and the ^{238}U and the ^{235}U currents, in 3+ charge state, were monitored in FC3 during 10 blocks each using the new 10 nA integration range. In Fig. 2, we show the obtained correlation between the ^{235}U and ^{238}U currents for every single block. The slope of the linear fit approximates the natural $^{235}U/^{238}U$ atom ratio of 7.2×10^{-3}, with a y-intercept of approximately -0.8 pA. Therefore, 0.8 pA is about the minimum true current that can be monitored in FC3 (i.e. offset limit). During a ^{236}U measurement, ^{238}U is monitored in this high-energy cup. According to our experience, a sample containing 3 µg of uranium produces a typical $^{238}U^{3+}$ current in FC3 of 45 pA. This is the amount of uranium that can be found in 1 l of seawater or in 1 g of soil, which evidences the relevance of the new GCPD as long as the measurement of $^{236}U/^{238}U$ atom ratios at environmental

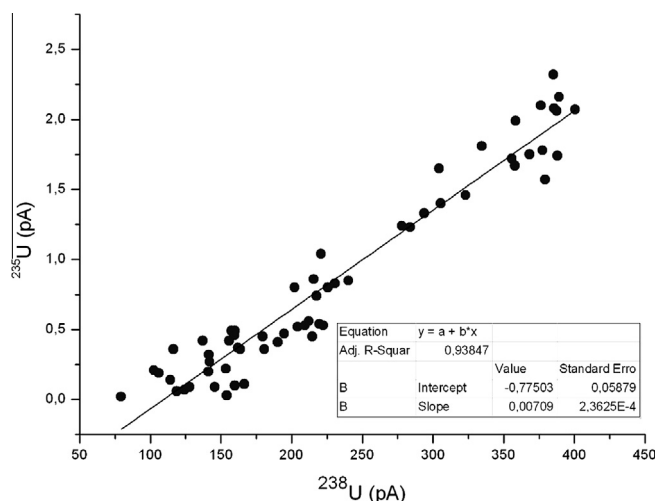levels is concerned. The determination of the $^{235}U/^{238}U$ isotopic ratio is also feasible for samples containing at least 3 times more uranium (more than 9 µg of ^{238}U), providing additional information about the uranium source in specific applications.

3. Ion source performance

One of the most critical items to the efficiency of the measurement in our case is the ion source. During the first 6 years we operated the system, we run the SO-110 model that was included in the original AMS setup. From our experience, this model needed a very frequent maintenance service. Also, the cross-contamination was a severe problem for volatile elements such as iodine. On the other hand, samples were not always efficiently used, as we found it difficult to center the Cs$^+$ beam on the sample. This is because correct placing of the ionizer, which has to be parallel and aligned within very restrictive specifications, was a difficult task.

In order to overcome these problems, in 2012 the upgrade version SO-110B was installed on our system [10]. This ion source has allowed us to measure longer without maintenance. However, alignment of the ionizer stays difficult despite the incorporation of a specific alignment tool. Another item is the dimension of the Cs$^+$ beam focus, as discussed next.

3.1. Sputtering geometry study

One of the major issues in the performance of the ion source is the capacity of the sputtering process to use as much of the present material as possible. For this purpose, the caesium sputtering should take place over the largest possible area of the sample without affecting the surrounding material. Besides, a well-focused Cs$^+$ beam ensures lowest memory and cross-contamination effects. On the other side, a too wide extracted beam may not have the optimal optical properties for the extraction, which may turn in a reduction of the extracted current. Although there are many factors affecting the sputtering process, one of the most obvious is the distance between the cathode surface and the ionizer, since this distance together with voltages applied on each item is a key-factor affecting the caesium beam focusing on the target.

The SO-110B ion source is supplied with two different cathode holders; the standard one and a short one specifically designed for volatile samples, which are pressed into a cathode with a shield that makes it about 3 mm longer. In effect, both ensembles have the same length and fit in the vacuum chamber where the samples are stored. If this short holder is used in combination with the normal cathode, a different sputtering geometry is obtained. For convenience, we will call this standard and short configuration. In the short configuration, the cathode is 1.7 mm further away from the ionizer than in the standard configuration (Fig. 3).

In order to compare both configurations, different experiments were performed. Graphite targets made from oxalic acid (Ox-II) and double spar carbonate (DBSP) containing about 1 mg of carbon were prepared at the Aarhus Radiocarbon Laboratory and used for the tests. Cathodes with the standard configuration were used to tune the system. One Ox-II and two DBSP were measured in the standard configuration, and two Ox-II and one DBSP were measured in the short one. Each sample was measured for 10 cycles of 60 s before changing to the following target. In this way, each target was measured multiple times, depending on its behavior.

After the measurement, targets were observed under the binoculars and a clear difference could be seen. In order to check the sputtering spot, one target of each configuration was measured during only 90 min (Fig. 4). The standard configuration produced a very sharp and deep sputtering hole as expected (Fig. 4a), while the short one showed a ring-like sputtering area (Fig. 4b). The rest
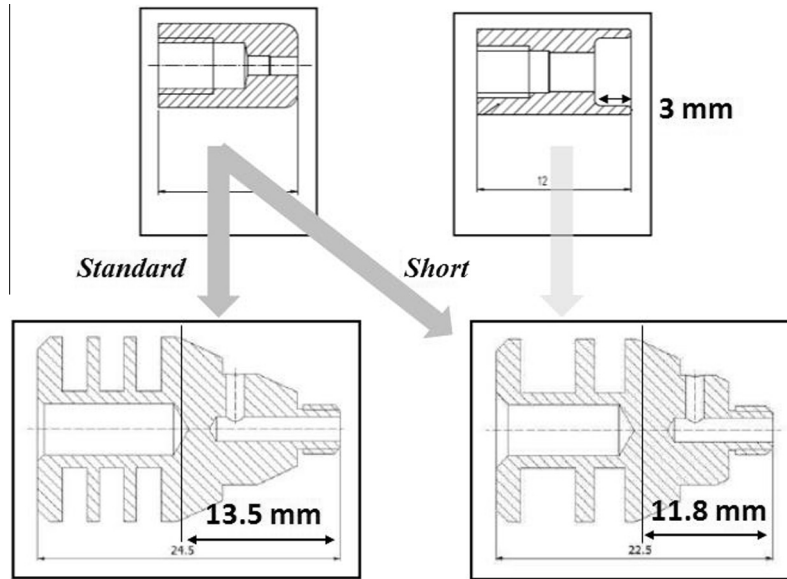

Fig. 2. Correlation between the $^{235}U^{3+}$ and $^{238}U^{3+}$ currents measured in FC3 for a set of natural uranium samples with total uranium contents ranging from 10 to 30 µg. Both isotopes were measured using the new 10 nA measurement range for the GCPD and the SSI software.

Fig. 3. Drawing of the two available configurations from HVEE for the cathodes and the holders for the SO-110B ion source. The so-called "short configuration" refers to the normal cathode combined with the short holder, which was originally conceived to hold the shielded cathodes. The so-called "standard configuration" refers to the conventional one, where the normal cathode is combined with the normal holder.
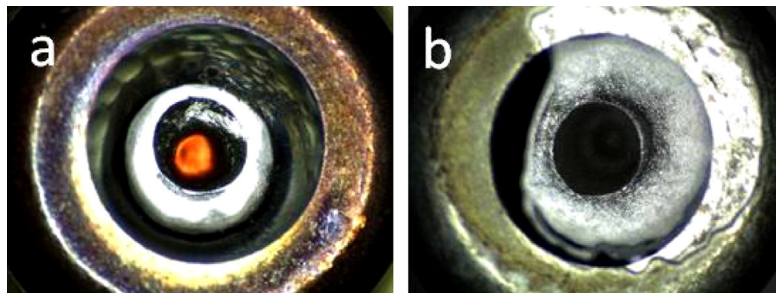

Fig. 4. Pictures showing the surface of two graphite samples after 90 min of sputtering with the standard (a) and the short configuration (b). The cupper pin is visible in the first case, due to the Cs$^+$ focusing on a small spot.
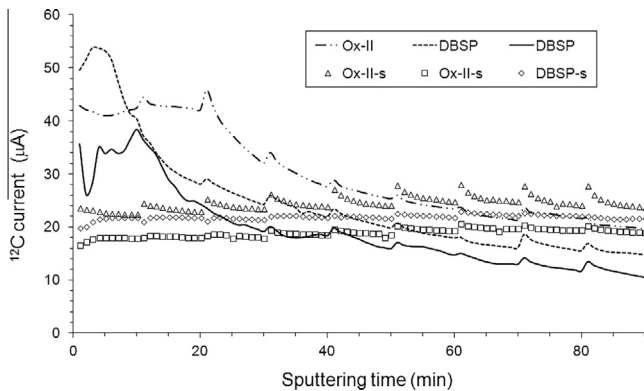

Fig. 5. Graphic showing the time evolution of the $^{12}C^{2+}$ current in FC3 after 90 min of sputtering for three cathodes with the short (-s) and the standard configuration.
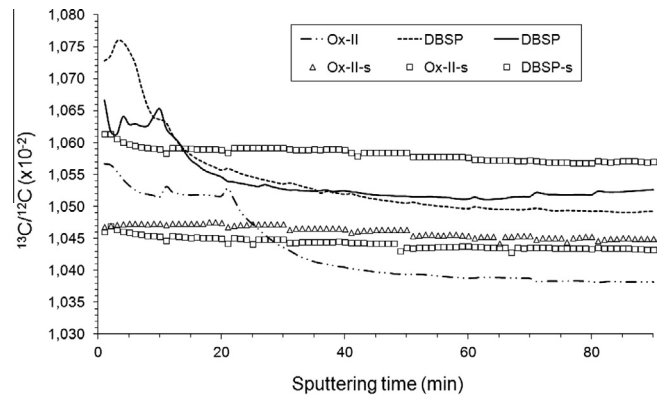

Fig. 6. Graphic showing the time evolution of the $^{13}C/^{12}C$ atom ratio after 90 min of sputtering for three cathodes with the short (-s) and the standard configuration, whose current evolution is displayed in Fig. 5.

of the targets were measured as long as the behavior was stable (i.e. until the extracted currents were less than half the currents extracted after 90 min, what happened after more than 200 min, at least). The usual measurement time for radiocarbon samples at CNA is about 60 min.

The most interesting results of the experiment are the evolution of both ^{12}C current and $^{13}C/^{12}C$ ratio over time. Figs. 5 and 6 show

the evolution of these two parameters during the first 90 min of the measurement. It is clear that each configuration behaves in a different way. In the standard configuration, ^{12}C current is about a factor of two higher than in the short configuration during the first minutes, but then decreases abruptly. On the other hand, the ^{12}C current in the short configuration keeps extremely stable

during the whole 90 min, and the current of the three targets is very close. In a very similar way, the $^{13}C/^{12}C$ ratio keeps stable within 0.5% in the short configuration for each individual cathode, while the standard configuration shows a variability of about 2%. $^{13}C/^{12}C$ ratios usually show a dependency on the extracted current. This is also true for the data presented, and the $^{13}C/^{12}C$ ratios fit linearly quite well with the extracted current in the case of the standard configuration ($R^2 > 0.84$). Results show a weaker dependency in the case of the short configuration targets ($R^2 < 0.65$). With such stable currents for the short configuration, the importance of this dependency diminishes. These results let us prefer the short configuration, although the extracted currents are lower during the usual measuring time. Assuming an overall transmission through the system of 43%, and taking 60 min as the reference measuring time, the efficiency varies from 1.2% to 1.7% for the standard configuration and from 0.9% to 1.2% for the short one. This may indicate that the efficiency is also cathode dependent, and that the surface of the graphite, the pressing and maybe other factors do have an effect in the sputtering process. Besides, subsequent measurements in the system showed that the results of this experiment for the short configuration are actually almost ideal from the current stability point of view, as the source is very sensitive to the ionizer position. To date, the short configuration is used on a routine basis at our facility for all nuclides.

4. AMS measuring technique at the CNA

In February 2006, the 1 MV AMS system at the CNA passed the acceptance tests for ^{10}Be, ^{14}C and ^{26}Al [3]. Since then, we have worked on the improvement of the technique for these radionuclides and on the measurement of other ones. In Table 1, details of the conventional measurement techniques and information about the standards and blanks used in every case are displayed. The most remarkable advances are discussed next.

4.1. ^{10}Be, ^{14}C and ^{26}Al

For ^{10}Be measurements, no remarkable improvements have been achieved since the status of the technique was reported in 2010 [14]. In the context of that work, it is worth to mention the studies focusing on the ^{10}B suppression using passive absorbers with different thicknesses and selecting different ions from the ion source (i.e. the BeF$^-$ method). Currently, routine measurements are performed using the beryllium oxide method and a 135 nm silicon nitride foil as a passive absorber. The transmission through the accelerator at a terminal voltage of 1 MV is 58% for charge state 1+. Selecting the 2+ charge state after the absorber, the overall transmission is 8%. The achieved background figure for a commercially available beryllium salt from Merck is about 4×10^{-14}. The primary ^{10}Be standard coded ^{10}Be-01-5-1, with a nominal $^{10}Be/^9Be$ ratio of 2.709×10^{-11}, is used for routine measurements [15].

^{14}C measurements are currently performed on a Micadas system that was setup at the CNA in 2012. Recently, both systems have demonstrated to produce similar results in terms of precision and background levels [16]. Therefore, the 1 MV one is used to support the ^{14}C dating service during long-term technical stops or maintenance operations that directly affect the Micadas performance. The achieved blank levels for the so-called IAEA-C1, Carrara marble [17], and for the carbonate sample Doublespar from Iceland, kindly provided by Prof. J. Heinemeier, range from 0.002 to 0.004 F^{14}C (F stands for fraction of modern carbon as defined in [18]), which is equivalent to approximately 50,000–45,000 years BP or a $^{14}C/^{12}C$ ratio of $\sim 3 \times 10^{-15}$. The laboratory also uses different organic materials as blanks, with similar results. The so-called Ox-2 reference material, with a 1.3407 ± 0.005 F^{14}C ($\delta 13C$: -25.0%), is used for normalization purposes.

^{26}Al is routinely measured on the 1+ charge state, featuring the best transmission through the accelerator of about 23% as reported in [4]. In the last years, the 3+ charge state has been also investigated. Similar background levels for the $^{26}Al/^{27}Al$ atom ratio, of about 3×10^{-14}, have been achieved in both cases, without an obvious role of the stripper gas thickness. For normalization purposes, the primary standard coded ^{26}Al-01-4-1, with a nominal $^{26}Al/^{27}Al$ ratio of 7.444×10^{-11}, is used [19].

4.2. ^{129}I, ^{236}U and plutonium isotopes

Details about the ^{129}I measuring technique can be found in [8]. Briefly, ^{129}I is extracted as I$^-$, stripped to I^{3+} at 1 MV terminal voltage with 10% of overall transmission, and counted from the 2-anode signal provided by the GIC. The best obtained $^{129}I/^{127}I$ background ratio for the Woodward Iodine (Woodward Iodine Corp., United States) is $(1.887 \pm 0.017) \times 10^{-13}$. For normalization purposes, we use a secondary standard named Q1 with a $^{129}I/^{127}I$

Table 1
Details of the measurement technique on the 1 MV AMS system at the CNA for the radionuclides that are analyzed routinely (^{10}Be, ^{14}C, ^{26}Al, ^{129}I, Pu isotopes and ^{236}U), and other ones which are in phase of study (^{41}Ca) or seldom measured (^{237}Np and $^{241,243}Am$). Ar gas is used as a stripper. The blank levels are expressed in atom ratios apart from Pu, Np and Am isotopes, where the number of atoms is given. P stands for primary standard; S for secondary standard.

Radionuclide	^{10}Be	^{14}C	^{26}Al	^{41}Ca	^{129}I	^{236}U	$^{239,240}Pu$, ^{237}Np, $^{241,243}Am$
I_{LE} (μA) (ion)	0.6 ($^9Be^{16}O$-) 0.1 ($^9Be^{19}F$-)	25 ($^{12}C^-$)	0.08 ($^{27}Al^-$)	0.001 ($^{40}Ca^{19}F_3^-$)	1–5 ($^{127}I^-$)	15 pA of $^{238}U^{3+}$ in FC3/μg ^{238}U	60 cps in GIC/pg (Pu, Np, Am)
Terminal voltage (MV)	1	1	0.7	1	1	0.65	0.65
Charge state	1 + (2 + after absorber)	2+	1+	3+	3+	3+	3+
Transmission (%)	8 (58 after accelerator)	43	23	6	10	11	NM
Blank ($\times 10^{-15}$)	40	2–4	30	10^4	200	5×10^4	1E6 atoms
Material blank	^9Be carrier from Merck	C1, SIRI-K, organic materials	^{27}Al carrier from Merck	–	Woodward iodine ($^{129}I/^{127}I < 5 \times 10^{-14}$)	Using "^{239}Pu"/^{238}U from Vienna-KkU as a proxy	Fe_2O_3 + Nb (1:3 mass ratio)
Stda used (P/ S, name) ratio	(P, ^{10}Be-01-5-1) $^{10}Be/^9Be$ = 2.709×10^{-11}	(P, Ox-2) (1.3407 ± 0.005) F^{14}C	(P, ^{26}Al-01-4-1) $^{26}Al/^{27}Al$ = 7.444×10^{-11}	(P, set of ERM-AE701)	(S, Q1) $^{129}I/^{127}I$ = 4.664×10^{-11}	(S, CNA-U236) $^{236}U/^{238}U$ = 3.5×10^{-8} $^{236}U/^{233}U$ = 0.71	(S, CNA-Pu) $^{239}Pu/^{242}Pu$ = 0.534 $^{240}Pu/^{242}Pu$ = 0.281
Refs.	[14]	[16]	[4]	–	[8]	[12]	[9]

atom ratio of $(4.664 \pm 0.020) \times 10^{-11}$. It was prepared by diluting the primary Standard Reference Material coded SRM-3231 supplied by the National Institute of Standards and Technology (NIST, United States), and calibrated at the ETH against other primary standards.

The plutonium and uranium isotopes measurement techniques are explained in detail in [9,12], respectively. Plutonium is an anthropogenic element without stable isotopes. The isotopes of interest, usually [239]Pu and [240]Pu, are normalized against an artificially added spike, usually [242]Pu. The three masses are counted in the detector in 5–25 s periods after the adjustment of the AMS system settings by the SSI software. The same technique is applied for [236]U studies, where usually an internal spike is also added to the samples (usually, [233]U). However, [238]U, the major isotope, is measured as a current in FC3. In both cases, monoxide ions are selected from the ion source (e.g. [236]U[16]O). Charge state 3+ is selected on the high-energy side with an overall transmission of 10% (e.g. [236]U[3+]). The detector gas pressure is adjusted to stop the ions below the first anode. The total energy signal so obtained is used to separate the 1+ and 2+ molecular fragments.

Both uranium and plutonium measurements benefit from the new cathode-holder configuration as discussed before, due to the gain in current stability that is especially critical for the SSI method. In fact, in the case of plutonium measurements, the instrumental precision of the atomic ratios (i.e. standard deviation of the mean of the Pu standard samples used in a measurement sequence) has been improved by a factor of 2, from 2% to 1%, by using the short configuration instead of the standard one. Secondary standards have been prepared in both cases for normalization purposes. In the case of plutonium isotopes, three independent [239]Pu, [240]Pu and [242]Pu reference solutions provided by National Physical Laboratories (NPL, England) were mixed to prepare a secondary standard solution (CNA-Pu) that was calibrated at the ETH Zürich. The final [239]Pu/[242]Pu and [240]Pu/[242]Pu ratios are 0.534 ± 0.011 and 0.281 ± 0.005, respectively [20]. As for [236]U measurements, a secondary standard solution (CNA-U236) was prepared from a [236]U standard solution supplied by the Institute for Reference Materials and Methods (IRMM-075/2), by adding [233]U and diluting the mixture with natural uranium. The resulting solution was also calibrated at the ETH Zürich. The final [236]U/[238]U and [236]U/[233]U atom ratios are, respectively, $(3.50 \pm 0.07) \times 10^{-8}$ and 0.71 ± 0.02. Background values of about 10^6 atoms have been obtained for plutonium isotopes, as explained in [9]. As for [236]U, abundance sensitivity for the [236]U/[238]U ratio of 5×10^{-11} has been estimated using the "[239]Pu"/[238]U atom ratio produced by a uranium mineral as a proxy, as discussed in [12]. No truly blanks for the [236]U/[238]U ratio are available at our laboratory.

4.3. New radionuclides: [41]Ca, [237]Np and [241,243]Am

The main challenge for the AMS measurement of [41]Ca is the suppression of the interfering isobar [41]K (6.73% isotopic abundance). Three different and complementary methods have been reported to reduce and/or correct this interference at low energies: (i) the chemistry improvement, (ii) the analysis of CaF_3^- from the Cs-sputter ion source, due to the lower ionization yield of KF_3^- and, (iii) the inspection of the bidimensional spectra to find qualitative evidences of the presence of the isobar [21]. In a first approach to the technique, a set of [41]Ca reference materials supplied by the European Reference Materials, with [41]Ca/[40]Ca ratios ranging from 10^{-11} to 10^{-8} (ERM-AE701), and a commercially available background material (i.e. high-purity $CaCO_3$ from Merck), were studied on the 1 MV facility at the CNA. They were adapted to a CaF_2 matrix following the sample preparation protocol given in [21]. CaF_3^- ions were analyzed on the low energy side, stripped to 3+ at 1 MV terminal voltage with 6% transmission, and finally
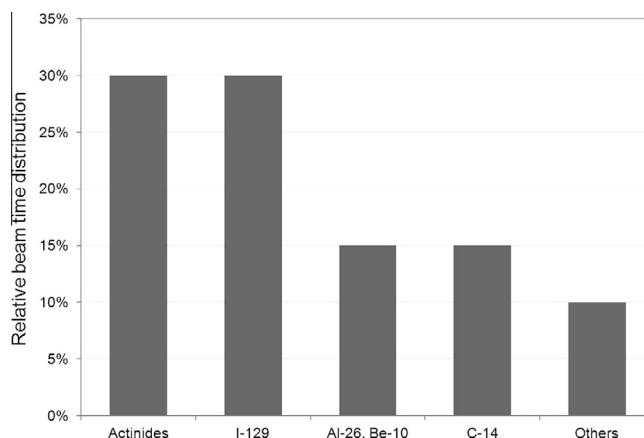

Fig. 7. Relative beam time distribution for the different radionuclides measured at the CNA in 2013.

counted from the bidimensional signal provided by the GIC. No sample-dependent effect was observed on the spectra. A very promising [41]Ca/[40]Ca background level of 10^{-11} has been achieved that way. This is only a factor of 2 higher than reported from the ETH for the 600 kV Tandy AMS system [20,21].

As for [237]Np and [241,243]Am, a similar measurement technique as for uranium and plutonium isotopes has been applied. For [237]Np, [233]U has been used as an internal spike for the AMS measurement. From the study of an in-house standard, for uranium a factor of two lower counting efficiency has been obtained compared to neptunium and plutonium. Background levels of about 10^6 atoms have been also obtained for [237]Np and Am isotopes, using iron oxide mixed with Nb as blank material and counting times of about 1 h, following a similar procedure as for plutonium isotopes [9].

5. Performance of the AMS facility

In order to discuss the functionality of the SARA AMS facility, 2013 can be considered as a reference year. During that year, no long-term maintenance service was needed, and the manpower featured with a stable situation. On average, 20% of the working days were dedicated to the maintenance of the system, and about 45% to the measurement of radionuclides. Regarding the studied radionuclides, about 30% of the beam time was dedicated to actinides and also to [129]I, which are, in addition to ^{14}C, the most demanded radionuclides from our laboratory (Fig. 7). With the Micadas system coming on the scene in 2012, only 15% of the 1 MV system beam time was dedicated to [14]C. An additional 15% was devoted to [26]Al and [10]Be measurements. The remaining 10% was allocated to the testing of new radionuclides.

Another interesting data is the number of measured samples per year, considering only the samples for external users. In 2013, the figures for every radionuclide were the followings: 180 samples for [129]I determinations; 100 samples for Pu isotopes studies; 30 for [236]U determinations; and 30 samples for [26]Al and 70 for [10]Be studies. Concerning the ^{14}C dating service, more than 1600 samples has been measured since its official implementation in 2007. The sample preparation is performed at the CNA laboratories.

6. Conclusions and future prospects

During the more than 8 years the 1 MV AMS system at the CNA has been in operation, its performance for most of the AMS radionuclides that can be measured at low energies has been studied. To date, [10]Be, ^{14}C, [26]Al, [129]I, [236]U and Pu isotopes can be measured on a routine basis with very competitive blank levels.

Although not mentioned on the paper, this includes the sample preparation techniques. The improvements on the performance of the ion source and on the so-called CGPD have been incorporated into the routine operation of the system. Our future prospects are the implementation of the He gas as stripper and the setup of a miniaturised GIC developed by the ETH Zurich group.

Acknowledgments

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