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^{41}Ca measurements on the 1 MV AMS facility at the Centro Nacional de Aceleradores (CNA, Spain)

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ABSTRACT

We present a detailed study of the performance parameters for ^{41}Ca measurements at the 1 MV AMS system at the Centro Nacional de Aceleradores (CNA) in Seville.

Mixing CaF_2 with Ag, we get stable $(^{40}CaF_3)^-$ currents between 50 and 100 nA. Transmission of the 2+ state in the He stripper is 41%, while optical transmission in the HE sector is typically higher than 95%. At low energies we cannot separate ^{41}Ca from the isobar ^{41}K at the detector, but the interference is reduced by using $(^{41}CaF_3)^-$ ions. The remaining contribution is corrected by measurement of the other stable isotope of potassium, ^{39}K, to estimate this interference (K-correction). After this correction, we reach a ^{41}Ca/^{40}Ca background level between 5×10^{-12} and 8×10^{-12}, making possible measurements of ^{41}Ca/^{40}Ca ratios higher than 4×10^{-11}.

Intercomparisons with the 0.6 MV AMS system at ETH Zurich show a quite good correlation.

1. Introduction

The main interest of ^{41}Ca accelerator mass spectrometry (AMS) measurements comes from its use as a tracer of the calcium metabolism [1–3]. The most recent value of its half-life, $(9.94 \pm 0.15) \times 10^4$ years [4], makes ^{41}Ca a perfect candidate for this. A much lower radioactive dose is received (below natural background) and it allows larger tracing time in comparison to the short-lived isotopes ^{45}Ca and ^{47}Ca, and it has a negligible natural abundance in comparison with stable isotopes of calcium.

The main interference in ^{41}Ca measurements is its stable isobar, ^{41}K (with a natural isotopic abundance of 6.73%). In the past, CaH_2 and extraction of $(CaH_3)^-$ were used to reduce the presence of ^{41}K in the beam. Since $(KH_3)^-$ ion is not stable, the only ^{41}K contribution would come from the extraction of $(^{41}K^1H^2H)^-$. The ^{41}K/^{40}Ca interference is on the order of 10^{-13} [5], leading to a final ^{41}Ca/^{40}Ca background $< 10^{-15}$ thanks to the discrimination in the detector. However, the chemical preparation of CaH_2 and its handling are problematic, because of its hygroscopic character, making it not suitable for large-scale applications.

In contrast, CaF_2 sample material can be easily prepared followed by an extraction of $(CaF_3)^-$, but the ^{41}K/^{40}Ca interference is increased to 10^{-12}–10^{-11}, and currents are lower [6]. For biomedical applications of ^{41}Ca, which do not require very low background, and usually a large number of samples have to be handled, the use of CaF_2 instead of CaH_2 means a great advantage. Because of that, simple ^{41}Ca radiochemical procedures for CaF_2 production were established for biological samples [7–9].

At larger accelerator AMS facilities (terminal voltage ≥5 MV) this ^{41}K interference can be discriminated on different ways. For instance, at these energies, ^{41}Ca and ^{41}K signals in a multianode ionization chambers can be distinguished, and final ^{41}Ca/^{40}Ca background levels in the order of 10^{-15} can be achieved [10]. Even in 3 MV systems, the use of silicon nitride detector windows is enough to provide a very good identification of both isobars, reaching ^{41}Ca/^{40}Ca background levels on the order of 10^{-14} [11].

Since biomedical studies can be designed so that ^{41}Ca/^{40}Ca ratios are higher than 10^{-10}, compact AMS facilities (terminal voltages ≤1 MV) are able to make this kind of ^{41}Ca measurements without

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relatively large background, even when no ^{41}K-^{41}Ca separation on the detector signal can be reached [12]. The trace amount of potassium may differ from one sample to another, and may depend on the chemical preparation, so treating ^{41}K/^{40}Ca contribution as part of the background would be incorrect. However, since ^{41}K and ^{39}K are both natural isotopes of potassium, measuring ^{39}K/^{40}Ca together with ^{41}Ca/^{40}Ca let us correct the interference due to ^{41}K [13]. The ^{41}K/^{39}K measured ratio is not exactly the natural one because of the slightly different transmissions in the stripper and the high energy (HE) sector depending on the mass. This experimental ratio can be easily measured in blanks, and then applied sequentially during the data analysis (K-correction).

The first compact AMS system reported to measure ^{41}Ca had been the 0.6 MV AMS system at ETH Zurich [12,13]. Most of the systems used to measure ^{41}Ca use terminal voltages ≥3 MV, and there is no robust published information of the performance for systems operating with voltages between 0.6 MV and 3 MV. The possibility of 1 MV AMS systems for ^{41}Ca has been discussed in previous bibliography [14,15], but no studies of their optimal conditions have been published. Moreover, all these studies were performed using argon as stripper gas. Helium has been proved to provide much higher transmissions for ^{41}Ca, among others, at low energies [16], and no studies about the performance of He stripping at 1 MV had been performed. In order to be able to optimize this kind of systems for routine measurements of ^{41}Ca, a detailed characterization is needed. In addition, the K-correction has to be implemented to get reliable results.

During the last years, we have optimized the performance parameters of the 1 MV compact AMS system at the CNA for ^{41}Ca. In the first section of this paper we present the main results of the optimization experiments for important performance parameters: ion source output, He stripper transmission, detector signal identification, background level and HE sector transmission. In the second section, we present the results of an intercomparison with the 0.6 MV AMS system at ETH Zurich. In order to do that, we measured two different standards from the ETH in-house set [17], and 40 biomedical samples previously measured at the 0.6 MV AMS system.

2. Performance of the 1 MV AMS system at CNA for ^{41}Ca

2.1. Experimental system and measurement technique

A detailed description of the 1 MV AMS system SARA (Spanish Accelerator for Radionuclide Analysis) can be found in previous publications [18,19]. However, in the last years, several changes have been made in the system. The most important are the upgrade to the new SO-110B ion source [20], the change of the stripping gas from Ar to He, and the substitution of the original detector with a miniaturized gas ionization chamber provided by the ETH Laboratory of Ion Beam Physics [21].

In Fig. 1, a schematic view of the setup for ^{41}Ca is shown. In the ion source, negative ions are produced by Cs sputtering with an energy of 35 keV. After focusing with an Einzel lens, the molecular ions $(^{40}Ca^{19}F_3)^-$, $(^{41}M^{19}F_3)^-$, and $(^{39}K^{19}F_3)^-$, are selected sequentially with an 90° bouncer magnet and injected into the tandem accelerator (^{41}M denotes ions of mass 41, i.e. ^{41}Ca and ^{41}K). The presence of an electrostatic lens (Q-Snout) in front of the low-energy acceleration tube allows focusing the beam in the stripper channel tube independently of the extraction energy from the ion source.

The quadrupole triplet (Q-pole) after the high-energy acceleration tube allows focusing the beam in the same point in the high energy sector independently of the charge state. This additional focusing from the Q-pole is more important for ^{41}Ca measurements than for other radionuclides. The focusing of the high energy acceleration tube depends on the effective charge state $\frac{M_0}{M}q$, where M_0 is the mass of the negative molecular ion selected in the LE side and M the mass of the positive ion selected in the HE side [22,23]. Therefore, the optics of the $^{41}Ca^{2+}$ beam would be equivalent to that of charge state 5+ because of the high mass difference with the initial negative ion, with a mass of 98 u. In contrast, compact AMS systems (such as the 0.6 MV AMS system TANDY at ETH Zurich) are usually designed for charge states 1+, 2 + and 3 + .

The 90° analyzing magnet and the 120° electrostatic deflector (ESA) perform the selection of charge state, mass and energy; this system is designed to provide an achromatic transport to the beam. ^{41}Ca and ^{39}K are finally measured with the two anode gas ionization chamber, using isobutane as counting gas.

The sequential injection of ^{39}K and ^{41}M in the detector is done by using different voltages at the accelerator terminal and the electrostatic deflector, as it is done in plutonium measurements [24]. ^{40}Ca is measured in the movable Faraday cup (FC-3) with the same terminal voltage as the one used to inject ^{39}K (around 970 kV).

2.2. Extraction of $(CaF_3)^-$ from CaF_2 with the SO-110B ion source

First tests of ^{41}Ca with the 1 MV system were done mixing the CaF_2 material with Nb powder in a CaF_2:Nb mass ratio of 1:3. Typical currents of $(^{40}CaF_3)^-$ were lower than 50 nA. In order to improve this, a test was performed with different blank targets, mixing CaF_2 with Nb or Ag and in 4 different mass ratios: 1:1, 1:2, 1:4 and 1:9.

Results, which are presented in Fig. 2, show a high increase of the current when Ag is used instead of Nb, and it also appears to be more stable. Optimal behavior is reached when the CaF_2:Ag mass ratio is 1:4, with stable $(^{40}CaF_3)^-$ currents higher than 75 nA. This mass ratio is the one closer to the density relationship between Ag and CaF_2, which is 3.3, meaning similar volumes of fluoride and silver. If we suppose that ionization efficiency is linearly dependent on the silver amount, this configuration with the same volume of CaF_2 and Ag would be that one with the highest total ion extraction yield.

2.3. He stripper performance for calcium at 1 MV

Preliminary tests of calcium measurements with the 1 MV AMS system were performed before the stripper gas was changed to He. In those tests with Ar, transmission for $^{40}Ca^{3+}$ was only 6% [20]. After changing the stripper gas to He, transmission for $^{40}Ca^{3+}$ increased to 28%.

However, using charge state 3+ would not allow us to measure ^{39}K because of the m/q interference of $^{13}C^+$. Since transmission for 4+ state is too low at the available energies, state 2+ has to be used, so special care with possible molecular interferences has to be taken. Transmission of $^{40}Ca^{2+}$ at 972 kV terminal voltage (stripping energy of 415 keV) for different He gas pressures is presented in Fig. 3. Measurement was performed with a blank sample. Since pressure is only measured at the center of the stripper channel, we use a simple calculation to estimate He mass thickness ρ_A: assuming a triangular distribution of the gas pressure in the stripper channel, we use the half of the average measured pressure and a temperature of 294.14 K to calculate the stripper gas density. This approximation is not trustworthy at low pressures, so only data with mass thicknesses above 0.015 μg·cm^{-2} is used to get the fitting curve.

In order to get the charge state fraction, we have applied the fitting found in Ref. [25]. This fitting gets the parameters Φ_{2+}, σ_p and σ_m for a transmission curve $T(\rho_A) = \Phi_{2+}[1 - \exp(-\sigma_p{\cdot}\rho_A)](1 - \sigma_m{\cdot}\rho_A)$. The factor $[1 - \exp(-\sigma_p{\cdot}\rho_A)]$ represents the production of Ca^{2+} ions; $(1 - \sigma_m{\cdot}\rho_A)$, the losses because of scattering. Φ_{2+} = 53% is the 2+ charge state fraction for a stripping energy of 415 keV, very close to the one for 223 keV, which is a 60% [16].

Fig. 3 also shows the dependence between ^{41}M/^{40}Ca ratio and He mass thickness in a blank sample. Exponential fitting lets us estimate the molecular destruction cross section as 255 cm^2·μg^{-1}. A maximum transmission of 45% is found for stripper pressures around 0.015 mbar, meaning a mass thickness of 0.03–0.04 μg·cm^{-2}; nevertheless, since