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ScienceDirect
Nuclear Instruments and Methods in Physics Research B 259 (2007) 677–682
NIM B
Beam Interactions with Materials & Atoms
www.elsevier.com/locate/nimb

AMS measurements of ^{41}Ca and ^{55}Fe at VERA – two radionuclides of astrophysical interest

Anton Wallner [a,*], Max Bichler [b], Iris Dillmann [c,d], Robin Golser [a], Franz Käppeler [c], Walter Kutschera [a], Michael Paul [e], Alfred Priller [a], Peter Steier [a], Christof Vockenhuber [f]

[a] Vienna Environmental Research Accelerator (VERA), Fakultät für Physik, Universität Wien, Währinger Strasse 17, A-1090 Wien, Austria
[b] Atominstitut der Österreichischen Universitäten, Stadionallee 2, 1020 Wien, Austria
[c] Institut für Kernphysik, Forschungszentrum Karlsruhe, Postfach 3640, 76021 Karlsruhe, Germany
[d] Department für Physik und Astronomie, Universität Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland
[e] Racah Institute of Physics, Hebrew University, Jerusalem 91904, Israel
[f] TRIUMF, 4004 Wesbrook Mall, Vancouver, BC, Canada V6T 2A3

Available online 3 February 2007

Abstract

The potential for measuring the radionuclides ^{41}Ca and ^{55}Fe was investigated with the 3 MV tandem accelerator at VERA. Interestingly, up to now, no applications have been published for ^{55}Fe using the technique of AMS. This is in strong contrast to ^{41}Ca, which is routinely measured by medium and large tandem accelerators in various applications. Using CaF_2 samples the quantification of ^{41}Ca down to levels of a few 10^{-13} for the isotope ratio ^{41}Ca/Ca has become possible with the use of the ΔTOF technique. Both nuclides, ^{41}Ca and ^{55}Fe were found to be of interest in nuclear astrophysics. A first application of ^{41}Ca detection at VERA is the measurement of the ^{40}Ca$(n,\gamma)^{41}$Ca cross section at stellar temperatures. Similarly, of astrophysical interest is the production of ^{55}Fe via neutron capture on ^{54}Fe. To this end, different Ca and Fe blank and standard samples were investigated with the goal to establish an AMS method for ^{41}Ca and ^{55}Fe measurements. Indeed, low background levels for both radionuclides, ^{41}Ca and ^{55}Fe, were observed in these studies. The full separation power of VERA results in a very low detection limit, while still providing a high overall transmission.

PACS: 26.30.+k; 26.35.+c; 26.50.+x; 82.80.Ms

Keywords: ^{41}Ca; ^{55}Fe; AMS; ΔTOF; Nuclear astrophysics

1. Introduction

Well-established data on production rates of long-lived radionuclides, important e.g. for various nucleosynthesis processes, are highly desired [1]. Lack of information exists for a list of nuclides as pointed out by nuclear data requests. The relevant stellar scenarios and their production mechanisms are an essential key for understanding celestial evolution [1,2]. A powerful and sensitive technique to experimentally determine production rates of interest of longer-lived radionuclides in nucleosynthesis processes is obtained by combining the activation method with a subsequent offline measurement of the production of those radionuclides via AMS [3]. One goal of research at VERA are the measurements of various neutron-capture reactions leading to such long-lived radionuclides. To this end, proper target materials are irradiated with neutrons at Forschungszentrum Karlsruhe (FZK) [4]. Neutrons are generated using the ^{7}Li$(p,n)^{7}$Be reaction by bombarding a Li target with protons with an energy near the threshold of this reaction. Proper settings allows one to tailor a quasi-stellar Maxwellian neutron spectrum of $kT = 25$ keV (which corresponds to a temperature of 3×10^8 K).

* Corresponding author. Tel.: +43 1 4277 51711; fax: +43 1 4277 9517.
E-mail address: anton.wallner@univie.ac.at (A. Wallner).

0168-583X/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.nimb.2007.01.207

Within this context, first applications are the $^{40}Ca(n,\gamma)^{41}Ca$ and the $^{54}Fe(n,\gamma)^{55}Fe$ neutron capture reactions. Applying the activation technique, isotope ratios of the order of 10^{-15}– 10^{-11} can be produced at FZK. The ratio obviously scales directly with the specific neutron cross-section value. Clearly, depending on the isotope of interest, such isotope ratios often represent a challenge for smaller tandem accelerators, if ''non-standard'' isotopes (like ^{14}C or ^{26}Al) are investigated. In order to meet these requirements for a 3 MV tandem accelerator, various technical developments and systematic investigations for detecting those radionuclides were necessary. The actual potential for detecting ^{55}Fe and ^{41}Ca at VERA is discussed in the following sections.

2. ^{55}Fe measurements at VERA

2.1. ^{55}Fe – an AMS nuclide

Iron represents an essential and nearly omnipresent element, with ^{55}Fe ($t_{1/2} = 2.75$ years) [5] having a great potential for various bio-analytical or technological applications. The natural abundance of stable Fe isotopes is 91.75% for ^{56}Fe and 5.85% for ^{54}Fe (the remaining isotopes are ^{57}Fe (2.1%) and ^{58}Fe (0.28%), respectively). ^{55}Fe has not attracted much attention for AMS measurements [6], although it is widely used as an X-ray calibration source. It is the requirement of the application as a calibration source, which recently resulted in very consistent half-life measurements [5]. ^{55}Fe almost exclusively disintegrates by electron capture to the ground state of stable ^{55}Mn, while emitting a low-energy K_α X-ray of 5.9 keV (and K_β of 6.5 keV). Of astrophysical interest is the production of ^{55}Fe via neutron-capture on ^{54}Fe. Iron represents the seed element for nucleosynthesis starting into the higher-mass region. It is this application, which triggered the development of ^{55}Fe AMS measurements at VERA. Interestingly, so far no AMS applications of ^{55}Fe detection have been published at all. The great potential of AMS for measuring ^{55}Fe, however, has been reported some 15 years ago by Korschinek et al. [6]. With the use of a ^{55}Fe standard material they could estimate a sensitivity for $^{55}Fe/Fe$ of about 8×10^{-13} using the 14 MV tandem accelerator at Munich. In this paper we present first measurements of ^{55}Fe using a smaller tandem accelerator with the goal to demonstrate the capability of measuring the stellar neutron-capture cross section for $^{54}Fe(n,\gamma)^{55}Fe$ in a temperature range relevant for nucleosynthesis processes.

2.2. ^{55}Fe blank samples measured with our standard AMS setup

Two different types of commercially available Fe powder in metallic form (Merck and Alfa-Aesar) with natural isotope abundances were used for our first approach towards Fe measurements at VERA (an enrichment in ^{54}Fe may be required for the quantification of some specific low-yield reaction cross sections on ^{54}Fe). The monoisotope ^{55}Mn is the only stable isobar to ^{55}Fe. Fortunately, Mn^- was not detected in AMS measurements. It has been demonstrated by Korschinek et al. [6] that Mn^- is too short-lived or does not form negative ions at all (i.e. they bind too weakly in order to survive and to be detected with an AMS particle detector). Therefore, it is advantageous to extract Fe^- from the ion source instead of the more prolific FeO. Currents for $^{56}Fe^-$ were found to be in the range between 500 nA and 1 μA.

Fig. 1 shows the yield for various charge states using gas stripping at VERA (currents were measured after our analyzing magnet). The abscissa represents the stripper gas pressure in arbitrary units. We have investigated first the 4^+ charge state because of higher particle energies compared to charge state 3^+ while still dealing with an acceptably high charge state yield. Later on we switched to the 3^+ charge state, which gives the highest yield.

In a first attempt we counted ^{55}Fe ions at the position right after the high-energy electrostatic analyzer (see Fig. 2, position A), which is usually used at VERA for ^{14}C and ^{26}Al measurements. The advantage of this position is that no beam losses were observed along the beamline between analyzing magnet (position of our Faraday cups used for the fast sequencing mode) and the detector position. Fig. 3(a) shows some spectra obtained with a Silicon surface barrier detector. Peaks labeled with ^{56}Fe and ^{54}Fe, respectively, represent the overlay of 2 consecutive runs with attenuated ^{56}Fe and ^{54}Fe beams. To this end the beam was first tuned for ^{56}Fe to collect ^{56}Fe signals. Then the setup was tuned for ^{54}Fe particles while leaving the magnetic rigidity constant (i.e. adjusting only the electrostatic components to account for their different masses). These setups have then been scaled for mass 55. ^{55}Fe signals are expected to appear in between these two peaks of ^{56}Fe and ^{54}Fe. Indeed some counts were registered when the machine was tuned for mass 55. The filled area in

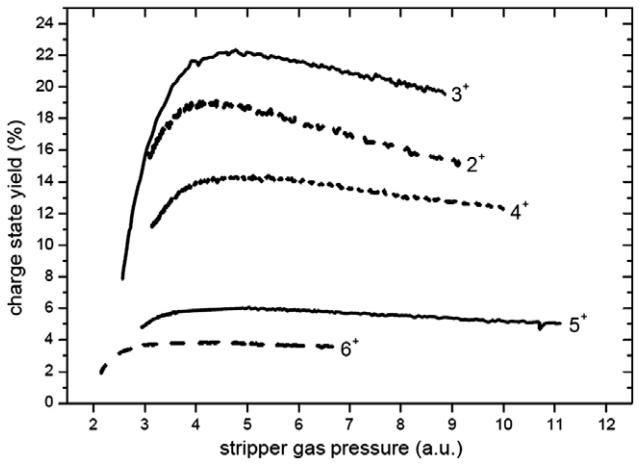

Fig. 1. Charge state yield for ^{56}Fe measured with a terminal voltage of 3.1 MV using a O_2 gas stripper. The current is measured after the analyzing magnet. The pressure reading (abscissa) in arbitrary units is proportional to the gas stripper pressure.

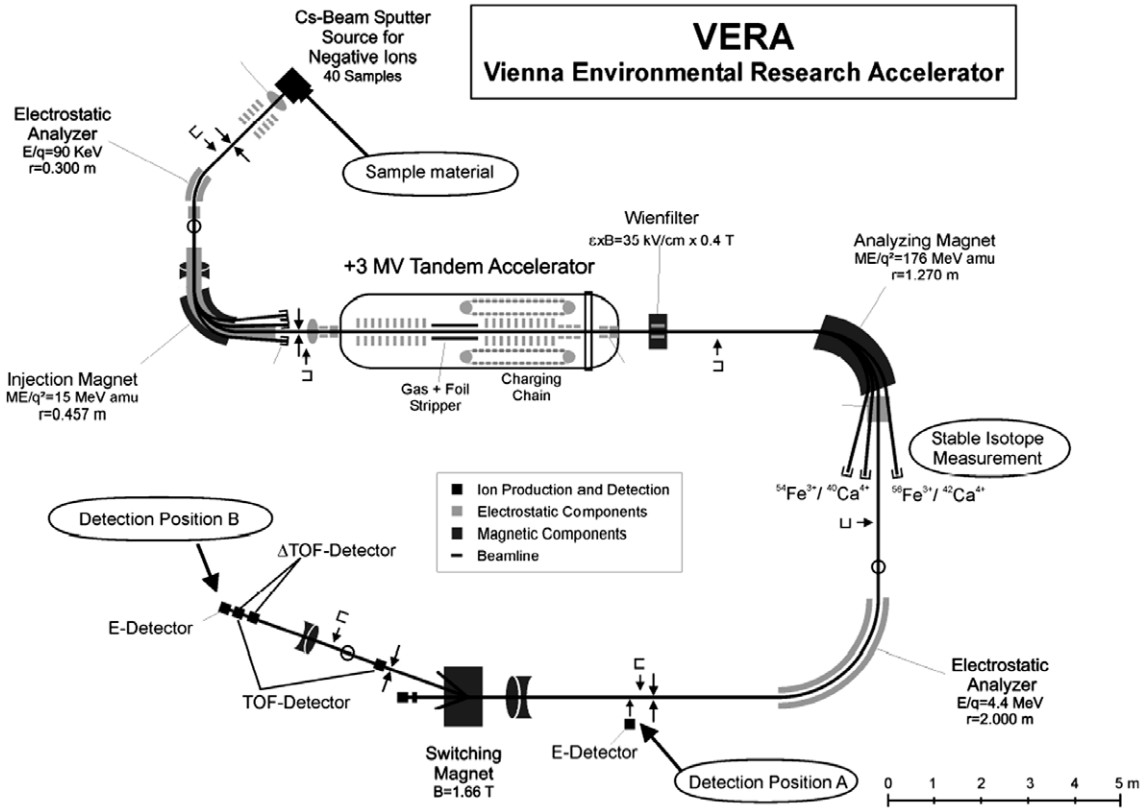

Fig. 2. Schematic layout of the VERA facility. The energy detectors used for this work are shown at position A (surface barrier detector) and in the heavy-ion beamline at position B (TOF and ionization chamber).

Fig. 3(a), noted as "^{55}Fe", represents a measurement on a blank sample, which is supposed to be free of ^{55}Fe. If we compare the peak maximum obtained from this blank sample, its position fits well with that found for the ^{54}Fe-setup. Very likely those counts obtained for the blank sample originate from ^{54}Fe-ions which were injected as $^{54}FeH^-$ into the tandem. A background ratio for $^{55}Fe/^{56}Fe$ in the range of 1×10^{-14}– 5×10^{-14} was obtained in runs on Fe blank samples. This value could be lowered by adding an aperture in front of the particle detector. It should be noted that this background level of a few 10^{-14} already fulfills the requirements needed for measuring the $^{54}Fe(n,\gamma)^{55}Fe$ cross section for typical stellar temperatures.

2.3. ^{55}Fe blank samples measured after passing an additional magnet

In order to get rid of those background events, the beam was transported to the heavy ion beamline after passing a switching magnet (see Fig. 2). Time-of-flight (TOF)-signals and finally also the energy signal from a Bragg-type ionization chamber were recorded (position B, see Fig. 2). Because at that time no ^{55}Fe standard material was available, particular attention was paid to a proper estimation of the absolutely measured isotope ratios. The transmission through the whole system was checked by switching to the stable isotopes, ^{54}Fe and ^{56}Fe, and by checking their measured isotope ratio (nominal ratio $^{56}Fe:^{54}Fe = 15.7:1$). Count-rates and the counting efficiency of the TOF-system were also checked with attenuated beams by verifying the measured count-rates of ^{56}Fe and ^{54}Fe with their natural abundance. In addition, a comparison of the count-rates at position B using attenuated ^{56}Fe and ^{54}Fe beams with that obtained from the surface barrier detector (see above, at position A) gave consistent results. No significant beam losses were found between the beam current measurement position and the detector at position A. Therefore, the measured absolute $^{55}Fe/Fe$ ratios using this detector at position A are to first order absolute ratios.

The measurements have shown that adding an additional magnet strongly reduces background signals. Measurements of blank samples gave no counts at all during two independent series of runs at position B, corresponding to an upper limit for the $^{55}Fe/^{56}Fe$ background ratio for ^{55}Fe-blank samples of 2×10^{-15}.

2.4. ^{55}Fe measurements at VERA using neutron activated samples

In a next step, a series of Fe powder samples were irradiated in a reactor with thermal neutrons to produce ^{55}Fe via the $^{54}Fe(n,\gamma)$ reaction. Therewith, reference materials with $^{55}Fe/^{56}Fe$ isotope ratios between 10^{-10} and 10^{-12} were obtained. These Fe standards were measured together with

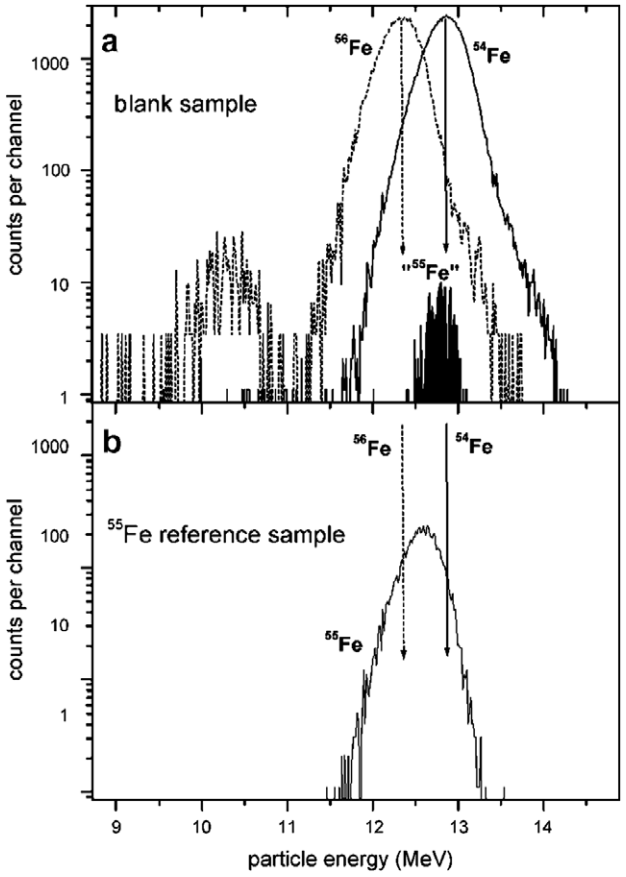

Fig. 3. Spectra obtained with a surface barrier detector at the detection position A. (a) Plotted are the peaks of attenuated ^{56}Fe and ^{54}Fe beams (overlay of different runs) with the same magnetic rigidity. The filled area depicts counts registered when the machine was set to mass 55. (b) ^{55}Fe counts obtained from an activated reference sample ($^{55}Fe/^{56}Fe = 2.8 \times 10^{-11}$).

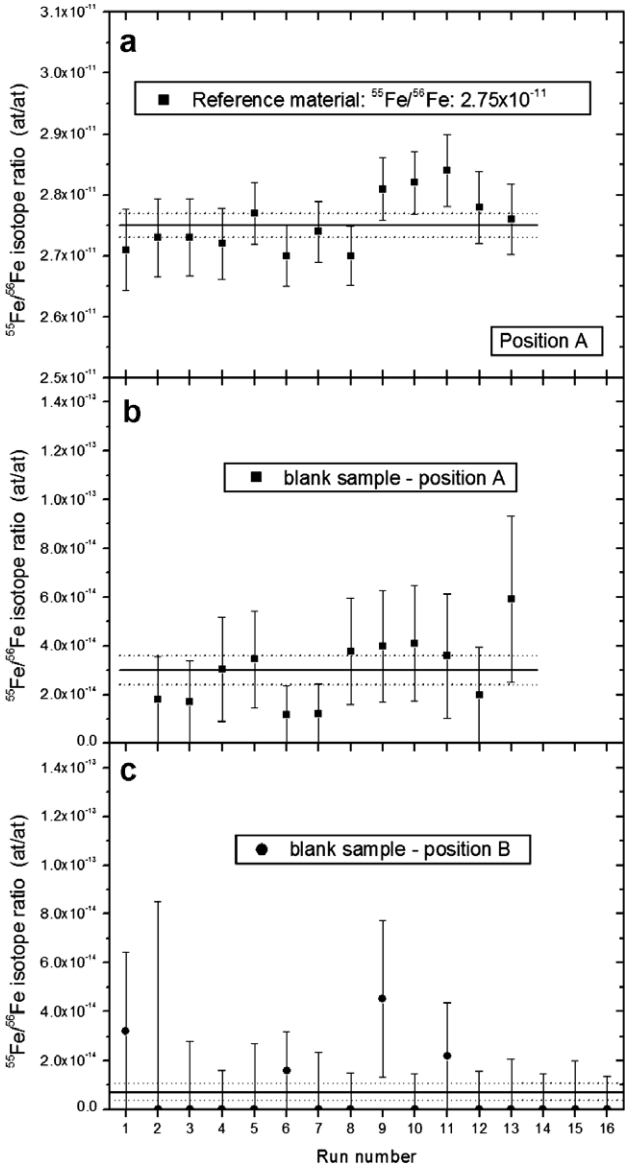

Fig. 4. ^{55}Fe/^{56}Fe isotope ratios for a reference (a) and a blank sample (b) as measured using the surface barrier detector at position A. Results for blanks samples using the additional switching magnet are shown in (c). The residual counts are attributed to cross-contamination from highly activated reference samples.

blank samples using the SNICS ion source, which contains a 40-samples target wheel. Both detection systems, the surface barrier detector (after the electrostatic analyzer at position A) and the ionization chamber using the heavy ion beamline (after the switching magnet, position B) were utilized. Fig. 3(b) shows a spectrum obtained at position A from a reference material.

The performance of Fe measurements is displayed in Fig. 4. Plotted are isotope ratios for a measurement series on an ^{55}Fe standard ($^{55}Fe/^{56}Fe \approx 2.8 \times 10^{-11}$) (Fig. 4(a)) and measurements on a non-irradiated Fe blank sample (Fig. 4(b)). Those data were obtained using the surface barrier detector (position A) including fast switching between ^{56}Fe, ^{54}Fe and ^{55}Fe. At this position, a mean value for the $^{55}Fe/^{56}Fe$ isotope ratio of $(2.75 \pm 0.03) \times 10^{-11}$ has been obtained. These data reflect the potential for measuring ^{55}Fe with high precision on the level of 1%. The blank sample gives a mean background ratio of $(3 \pm 1) \times 10^{-14}$.

When using the heavy-ion beamline with the additional switching magnet (position B) similar results were obtained. However, due to beam losses in the switcher magnet the reproducibility of those measurements was about a factor of 3 lower compared to position A; i.e. typical values were of the order of 3%. Blank samples give very few counts (see Fig. 4(c)). Data for a blank sample, shown in the lower plot of Fig. 4, correspond to a $^{55}Fe/^{56}Fe$ background ratio of $(7 \pm 3) \times 10^{-15}$. Interestingly, blank samples, which were originally found to be free of ^{55}Fe signals using this detection beamline, now showed up true ^{55}Fe counts when measured together with standard samples within the same sample wheel. Isotope ratios for blank samples measured under such conditions were found in the range of a few times 10^{-15} and up to 10^{-14}. These measurements indicate a cross contamination for Fe in our ion source at a level of approximately 10^{-4}. To summarize, using the detection setup A allows high precision measurements with a background level in the range of a few

10^{-14}. In contrast, using the heavy ion beamline (setup B) allows ^{55}Fe measurements for very low isotope ratios. Its background level was likely dominated by cross contamination from strong ^{55}Fe reference samples in the ion source. However, when using this setup B a slightly lower precision is obtained.

As already mentioned above, Fe^- currents up to 1 μA were extracted from the ion source. These measurements show that ^{55}Fe may be considered as a useful candidate for various other applications, too. Such applications were already previously identified [6], e.g. in medicine, biology, and metallurgy. The combination of a mass spectrometric technique and the high overall transmission of an AMS facility make such applications favorable.

The next step at VERA will be the application of ^{55}Fe detection for measuring the $^{54}Fe(n,\gamma)^{55}Fe$ cross sections at stellar temperatures. To this end, Fe samples have already been irradiated with neutrons at FZK with a 25 keV quasi-Maxwellian spectrum. The reaction rate in this energy range is of interest in the nucleosynthesis flow to heavier nuclei via the s- and r-process, which starts at the Fe-mass region. An accurate measurement of the cross section is highly desired.

3. ^{41}Ca measurements at VERA

Until recently, the long-lived medium-mass isotopes ^{41}Ca, was not accessible to AMS measurements using tandem accelerators with terminal voltages of 3 MV or lower. The discrimination of the isobaric interference ^{41}K represents a difficult task at low particle energies. However, the successful development of the so-called Δ TOF technique [7,8] has opened a new possibility. First investigations on Ca measurements at VERA have been presented in a previous publication [9]. The measurement of ^{41}Ca at VERA was also driven by an ongoing study of the $^{40}Ca(n,\gamma)^{41}Ca$ reaction cross-section at astrophysical energies: Ca-samples were irradiated with a quasi-stellar neutron spectrum of $kT = 25$ keV at FZK ([4], see Section 1). After the irradiation, part of the sample material was converted into CaF_2 and the number of produced ^{41}Ca nuclei is measured offline by AMS. Several CaF_2 standards were at our disposal, kindly supplied by K. Nishiizumi [10] and by G. Korschinek (GAMS-group, TU Munich), which encompass $^{41}Ca/^{40}Ca$ isotope ratios from 1.16×10^{-10} down to 5.9×10^{-13}. In addition, CaF_2 blank material was prepared from commercial $CaCO_3$ powder.

For samples with a $^{41}Ca/Ca$ isotope ratio of 10^{-13} and higher, the detection of ^{41}Ca by means of CaF_2 samples becomes possible. For our studies we prepared CaF_2 samples for ^{41}Ca detection at VERA. When extracting CaF_3^-, although KF_3^- is suppressed, it is not eliminated totally from the beam [11,12]. We utilized the ΔTOF technique to discriminate ^{41}K from ^{41}Ca. The basic principle of this method is to measure the difference of the energy loss when passing an absorber foil. A difference in the residual energy is reflected also in a difference of the flight time, which is measured applying a TOF setup (see below). The discrimination between the isobars, ^{41}K and ^{41}Ca, increases with higher particle energies.

CaF_3^- ions were injected into the tandem, which is operated typically at 3.3 MV (this 10% increase over the nominal terminal voltage of 3 MV can safely be used at VERA [7]). Selecting the 4^+ charge state yields particle energies of 14.6 MeV. Approximately 12% of the injected ions are stripped to the 4^+ charge state by using gas stripping in the terminal. After passing a 90° electrostatic analyzer and an additional switching magnet, the beam is transported to the heavy ion beamline, which includes the ΔTOF detection system (see Fig. 2). The ΔTOF detector consists of two micro-channel plates. Right after the first channel plate (start detector) stacks of Silicon nitride (SiN) foils can be inserted with different thickness-combinations to provide the Z-dependent energy degradation for transmitted ions. The second micro-channel plate (TOF-stop detector) follows in 0.64-m distance to the start detector. After the TOF-path the particles finally enter a Bragg-type ionization chamber for the measurement of the residual energy. The different energy losses of ^{41}Ca and ^{41}K in the silicon nitride degrader foils leads to the separation of the remaining ^{41}K counts from the ^{41}Ca peak. It is the exceptional homogeneity of the SiN foils which allows to resolve the different residual energies of ^{41}K and ^{41}Ca by a TOF measurement.

The measured ratios of well-known ^{41}Ca standards were compared with their reported nominal ratios. SiN stacks spanning a thickness range between 600 and 1600 nm (200–544 μg cm^{-2}) have been investigated to quantify the separation power for Ca and K and to assess the transmission of the ΔTOF beamline. Particle transmissions between approx. 55% of the 4^+-particles were measured for the 600 nm foils and approx. 7% for the 1600 nm foils, respectively. Both setups reproduce the nominal isotope ratios very well if the different transmission is properly corrected. Blank samples showed isotope ratios of the order of 10^{-14} and 10^{-15}, respectively, for these setups. When corrected for their different losses along the Δ TOF beamline, these ratios scale to a ^{41}K induced background of the order of 10^{-13}. From these results it can be deduced that isotope ratios in the low 10^{-13} range can be quantified.

In a later measurement series, a ^{41}K background countrate about a factor of 10 higher was observed. This enhanced K background was found to originate from the ion source itself: new ceramic beads used to insulate the ionizer heating were identified to contain potassium at a high concentration level. When replacing those ceramic parts, the K background decreased to its previous level. Very helpful in this context are new refinements in our ion source [13], which allow scanning across and beyond the sputter target surface by moving the sample relative to the Cs beam.

These tests have shown that the ΔTOF method is capable of quantifying $^{41}Ca/Ca$ isotope ratios down to a few 10^{-13}. We have now started to measure the Ca samples

irradiated at FZK (see above) to determine the neutron capture cross-section for a typical stellar environment. The expected isotope ratio of this sample is of the order of 10^{-11}, well above the background level. These measurements will represent the first ^{41}Ca AMS runs on a real sample using a 3 MV tandem accelerator. A detailed description of Ca measurements at VERA will be published in a separate paper [14].

4. Summary

AMS has evolved from nuclear physics techniques. A strong relationship between AMS and nuclear physics is therefore natural. In particular, nuclear astrophysics experiments represent a prominent application of AMS, e.g. the search for extraterrestrial radioactivity on Earth, measurements of long-lived radionuclides from the Moon or from meteorites, and the understanding of our solar system abundances. All those are research fields with important contributions from AMS. The combination of activation techniques with AMS has proven to be a powerful tool for the investigation of nucleosynthesis processes. At VERA systematic studies on various isotopes are being performed in this context. The development of the ΔTOF technique makes it now possible to detect ^{41}Ca with 3 MV tandems using CaF_2. It could be demonstrated that ^{41}Ca/Ca ratios at levels of a few 10^{-13} can be quantified. The advantage of an isobar-free AMS isotope like ^{55}Fe makes smaller AMS facilities a proper tool, especially when combined with an effective isotope suppression. This fact is reflected in an upper detection limit of ^{55}Fe/^{56}Fe of 2×10^{-15}. ^{55}Fe detection benefits from the efficient background suppression at VERA, enabling us to investigate very low reaction cross-sections. For both isotopes, ^{41}Ca and ^{55}Fe, a measurement program is underway for studying their production rates in stellar nucleosynthesis processes via the measurement of neutron capture cross-sections simulating such stellar conditions.

Acknowledgements

We gratefully acknowledge Kunihiko Nishiizumi and Gunther Korschinek for supplying some of the Ca standard material for our measurement program. I.D. is supported by the Swiss National Science Foundation Grants Nos. 2024-067428.01 and 2000-105328.

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