TRIUMF - EEC SUBMISSION

Draft Submission

Progress Report

Exp. No.

S1141

Date Created:

2007-05-17 11:49:30

Title of Experiment:

Towards the Detection of Supernova-produced Sm-146 on Earth -- Proof of Principle of a New Measuring Concept

Name of group:

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Beam Shifts Used:

Beam Shifts Remaining:

New Beam Requests:

Basic Information:

Date Created: 2007-05-17 11:49:30

Date Ready: 2007-12-31

Summary: The purpose of this proposal is to test a new concept for detecting supernova-produced ^{146}Sm on Earth. The main interference in such a measurement is the stable isobar ^{146}Nd, which cannot be separated by conventional methods yet. This new concept is based on full stripping of ^{146}Sm to q = 62 and a subsequent m/q analysis which allows are clear separation from ^{146}Nd as its maximum charge state is q = 60. The TITAN facility with the EBIT for full stripping and a Wien-filter for m/q analysis provides the essential components. First, a proof-of-principle experiment is proposed for the separation of ^{41}Ca and ^{41}K.

Plain Text Summary: The purpose of this proposal is to test a new concept for detecting supernova-produced Sm-146 on Earth. The TITAN facility with the EBIT allows full stripping and thus a separation of the interfering Nd-146.

Primary Beam Line: isac2a

ISAC Beamline/Facility

ISAC: TITAN

ISAC-I:

ISAC-II Beamline:

ISAC-II Facility:

Secondary Beam

Isotope: 41Ca

Energy:

Energy Units:

Energy Width:

Angular Divergence:

Spot Size:

Intensity:

Beam Purity:

ISAC Target:

Special Characteristics:

Production Sources

ISAC Ion Source: OLIS

TRIUMF Support (Resources Needed): Standard TRIUMF support for OLIS and TITAN.

NSERC:

Other Funding:

Safety Issues: Standard safety procedures for OLIS and TITAN will be observed.

Publications Christof Vockenhuber

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1 Motivation

The detection of supernova-produced radionuclides on Earth has been intensively discussed for more than ten years [1]. Indeed, a clear signal of ^{60}Fe ($t_{1/2} = 1.5 \times 10^6$ yr) was found in a deep sea FeMn crust using accelerator mass spectrometry (AMS), a very sensitive method for detection of tiny amounts of long-lived radionuclides [2] (Fig.1). The most likely explanation is that a supernova exploded about 2.8 million years ago in the vicinity (within a few hundred light years) of the Earth since ^{60}Fe is expected to be ejected in significant quantities [3]. A small fraction of the ejected ^{60}Fe is expected to be deposited on Earth as the shock front from the supernova wave passes and can be detected in suitable archives with a sufficiently sensitive method [4].

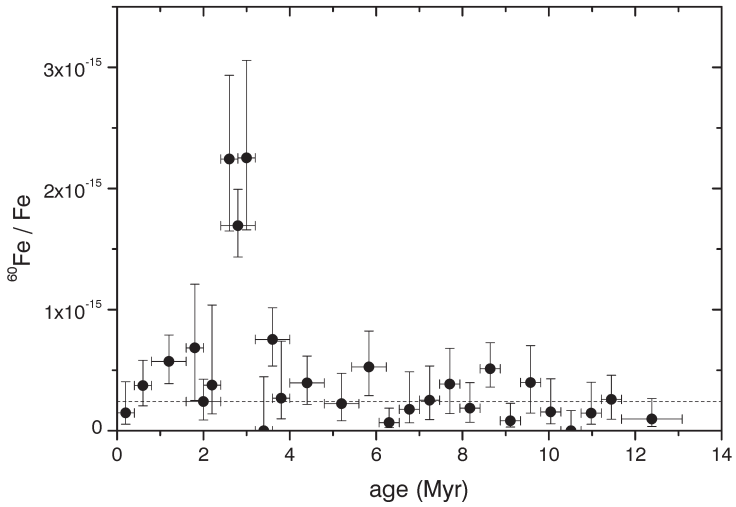

Figure 1: Measured ^{60}Fe/Fe ratio as a function of age of the layer in the deep-sea FeMn crust. The peak at 2.8 Myr is interpreted as a signal from a nearby supernova explosion. Note, the extremely low isotopic ratio, the peak contains only 69 counts taken in several weeks of beam time, indicating the tremendous challenge of these kind of measurements. Figure taken from [2].

A few other long-lived radionuclides are produced and ejected as well, but only those with a half-life much shorter than the age of the solar system (the so-called "extinct" nuclides with half-lives of 1000 yr $< t_{1/2} <$ 100 Myr) can be used as a supernova indicator. Figure 2 shows some expected surface densities for five nuclides based on the ^{60}Fe signal. Among these nuclides, the lighter ones are also produced in significant quantities mainly by spallation reactions by cosmic rays in the atmosphere and by infall of previously exposed meteoritic material. Thus, only the heavier candidates have no significant background which would obscure the supernova signal. In particular ^{146}Sm (1.03×10^8 yr), ^{182}Hf (8.9×10^6 yr), ^{244}Pu (8.0×10^7 yr), and also ^{247}Cm (1.56×10^7 yr) are produced and ejected as well, although in smaller quantities, and could be detected in a similar way [5][1]. In fact, a confirmation of the supernova origin of the ^{60}Fe signal can be provided by a finding of at least one of these other nuclides. From the astrophysical point of view ^{182}Hf, ^{244}Pu and ^{247}Cm would confirm for the first time that supernovae are the site of the r process, whereas ^{146}Sm is produced by the p process [6], a process which is responsible for the production of a few proton-rich nuclides and which is not well

[1] In Ref. [5] ^{146}Sm was discussed misleadingly as an r-process nuclide. But, lying on the proton rich side of the valley of stability and well separated from stable nuclides, it can only be produced by the p process (Fig. 3). Also, background from fission does not have any relevance because of the neutron-rich nature of the fission fragments and subsequent β^- decays to ^{146}Sm are blocked by the stable nuclide ^{146}Nd.

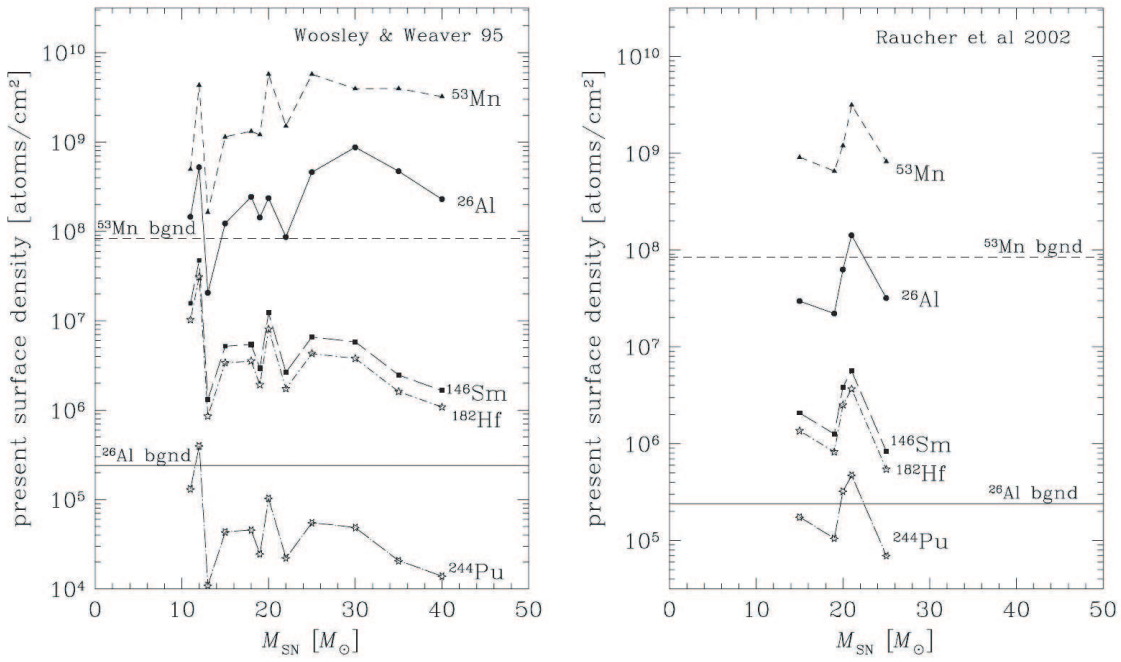

Figure 2: Expected terrestrial surface densities of a few relevant nuclides (^{26}Al, ^{53}Mn, ^{146}Sm, ^{182}Hf and ^{244}Pu) as a function of supernova progenitor mass using two different supernova models. The calculation is based on the measured ^{60}Fe signal of 2×10^9 atoms cm^{-2} [2] and is meant to show the order of magnitude of an expected signal. See Ref. [5] for details, from which the figure is taken.

understood yet. Evidence that these nuclides are indeed produced in stellar environments comes from their abundance in the early solar system inferred from isotopic anomalies of the stable daughter nuclide in meteorites (see e.g. [7, 8] for a general review). In particular, ^{146}Sm is a p-process isotope that α-decays to stable ^{142}Nd (Fig. 3). From anomalies in the ^{142}Nd/^{144}Nd ratio an early solar system ratio ^{146}Sm/^{144}Sm ≈ 0.01 [9] can be inferred, which also has cosmochronologic implications [10].

One advantage of ^{146}Sm is its half-life, which is about 70 times longer than that of ^{60}Fe, allowing a search for signatures of nearby supernova a few 100 Myr back. However, that advantage might be compensated by the fact that supernovae that produce the p-nuclides are less frequent than those producing the r-process nuclides, at least the ones with masses $A > 130$, as recently inferred from Sm and Nd isotopic compositions [11]. From that point of view, a direct finding of nuclides produced by different processes (i.e. p- and various components of the r-process) from a single supernova would provide a constraint on the site of these processes.

The direct detection of the above mentioned nuclides from nearby supernovae is often very difficult because of the extremely small amount. It requires careful sample selection and elaborate sample preparation, which will not be further discussed here (see e.g. [2] for some discussion). In addition, for ^{146}Sm and ^{182}Hf additional interference from stable isobars (^{146}Nd and ^{182}W, respectively) is a major experimental challenge in conventional AMS as it requires very high ion energies to separate them in the final detector. Isobar separation by mass is nearly impossible given the very small mass difference (e.g. for ^{146}Sm – ^{146}Nd, $\Delta M/M = 5.2 \times 10^{-7}$). The purpose of this proposal is to test a new measuring concept for ^{146}Sm at low energies using highly charged ions.

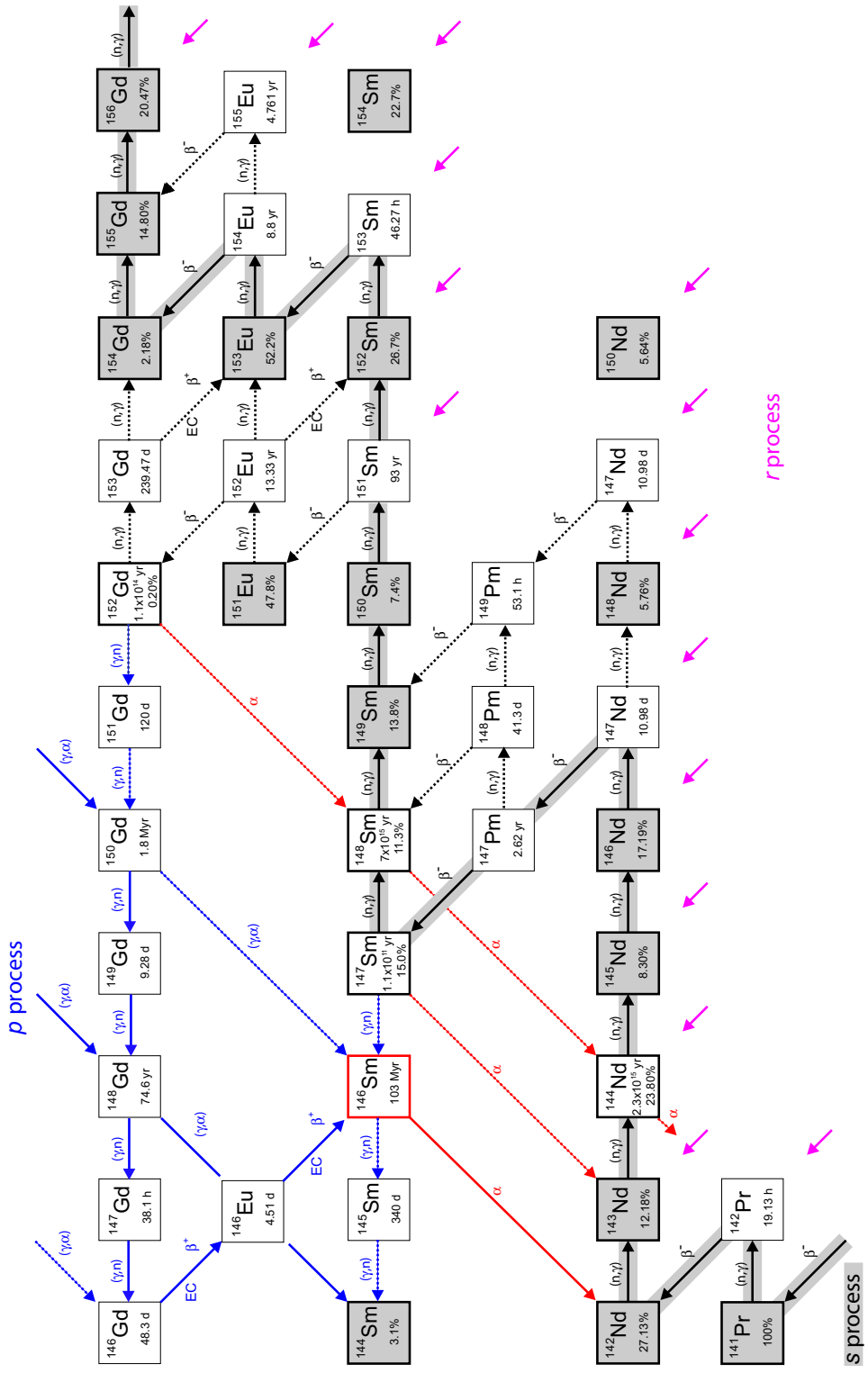

Figure 3: Nuclear chart in the region of Sm. The reaction path of the s process follows the stable isotopes (black arrows, main path underlined in grey). Neutron-rich isotopes are produced by the r process, indicated by short pink arrows from the bottom-right. ^{146}Sm is bypassed by these processes and can only be produced by the p process. A possible production scenario by a series of photodisintegration processes on heavy seed nuclei as discussed in [6] is indicated by blue arrows. After the nucleosynthesis event long-lived isotopes decay to stable isotopes shown in red. ^{146}Sm decays to ^{142}Nd by α decay. The abundance in the early solar system can be inferred by isotopic anomalies in Nd. Stable nuclei are indicated by grey squares, radioactive nuclei by white boxes with either a bold border for long-lived nuclei with primordial abundance or a thin border for short-lived nuclei. Weak paths are shown by a dotted line.

2 New measurement concept

Before proposing the new concept, we briefly describe conventional AMS and its limitations for the measurement of ^{146}Sm.

Accelerator mass spectrometry (AMS) started about 30 years ago in nuclear physics laboratories with large tandem accelerators [12]. Since then, it evolved into a mature method for measuring long-lived radionuclides at very low concentrations; isotopic ratios ranging from 10^{-10} down to 10^{-16} are routinely measured. There are essentially two prerequisites for measurements of extremely low isotopic ratios. First, a separation of the radionuclide of interest from other isotopes and (molecular and atomic) isobars is required. The challenge here is the much higher abundance of these interferences by many orders of magnitude. Second, high efficiency of the measuring system together with high beam currents from the ion source is needed. As an example, considering an isotopic ratio of 10^{-14}, a beam of 100 nA of the stable isotope ($= 6.24 \times 10^{11}$ ions per second) leads to a count rate of 22 counts per hour of the radionuclide (assuming 100% detection efficiency).

The great success of AMS is provided by the fact that isotopic interference is, to a large extent, reduced by two mass spectrometers, one acting at the low energy side, the other at the high energy side of the tandem accelerator. Interference from molecular isobars is removed by the stripping process at the high voltage terminal of the tandem accelerator. Atomic isobars are, in certain cases, suppressed in the ion source if they don't form stable negative ions. The accelerator also provides an important advantage of AMS over conventional mass spectrometry, the virtual absence of dark counts because of the high ion energies at the final detector, which enables measurements of extremely low count rates.

However, despite its great success, there are several disadvantages to AMS, as it is practiced at present: First, while it is essential for the successful measurement of several nuclides like ^{14}C, ^{26}Al and ^{129}I, the requirement of negative ions for injection into the tandem accelerator is one limitation, which makes the overall detection for many relevant nuclides less efficient or even, as for instance for the noble gases, impossible. Second, if not provided by the ion source, isobar separation requires high ion energies for a separation in the final detector. While it is possible to measure light radionuclides with isobaric interference at facilities based on small accelerators (e.g. ^{10}Be, ^{36}Cl at the 3-MV facility VERA [13]), and radionuclides in the medium mass range at large accelerators (e.g. ^{60}Fe, ^{63}Ni at the 14-MV tandem accelerator of the Maier-Leibnitz-Labor in Munich [14]), no method exist right now for the heaviest radionuclides with isobaric interference at the required low concentrations (e.g. ^{146}Sm, ^{182}Hf).[2]

The basic idea of the new concept is the following: Some of the radionuclides of interest in AMS have a higher nuclear charge than their interfering stable isobar. In principle, they can be unambiguously identified if they are electromagnetically analyzed as bare nuclides, since the radionuclide can reach a higher charge state and thus a lower mass-to-charge ratio. This has been done for relatively light ions such as ^{36}Cl at high ion energies by the method of full stripping using a thin carbon foil [18]. Very high energies of 3.6 GeV had to be employed in a full-stripping AMS experiment of ^{81}Kr [19] which renders this method impractical for even heavier ions. However, there exists the possibility to produce highly charged ions in a so-called Electron Beam Ion Trap (EBIT) by the use of an intense electron beam with sufficient energy. If such a device is coupled to two mass spectrometers in a similar way as the tandem accelerator in conventional AMS, one can apply the method of full stripping at much lower energies and thus at a much smaller facility (Fig. 4). Many advantages of

[2] Some progress has been recently reported using the ATLAS facility for separation of ^{146}Sm and ^{146}Nd at the gas-filled magnet [15], and for ^{182}Hf at VERA [16] and at the Maier-Leibnitz-Labor in Munich using the ΔTOF technique [17]; however, the achievable sensitivity has not reached the required level yet.

the conventional AMS are also applicable: Molecular interferences are completely destroyed in the intense electron beam. Depending on the charge state and the extraction voltage, energies in the MeV range can be achieved making the final detection easier. One difference is that positively charged ions can be used, which can provide high ionization efficiencies and also the possibility of measuring long-lived noble gas nuclides. Theoretically, such a device can be used even for isobars in the high mass range (e.g. ^{146}Sm), provided the electron energy is high enough for efficient full stripping.

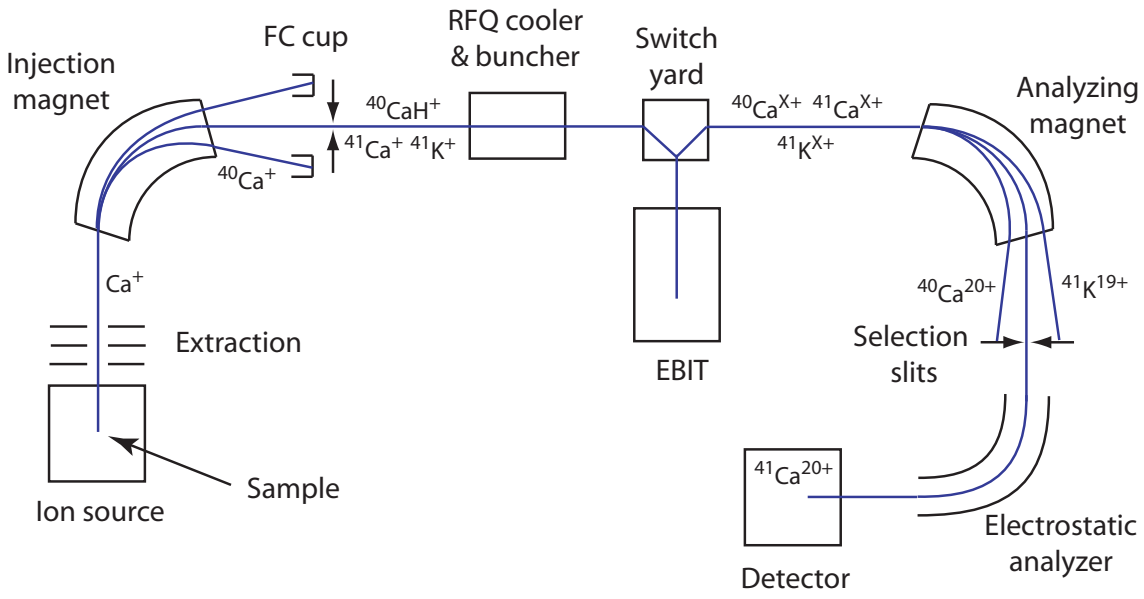

Figure 4: Schematic setup of an EBIT based Mass Spectrometry facility. As an example, the separation of ^{41}Ca from isotopes (^{40}Ca) and isobars (^{40}CaH,^{41}K) is shown. The combination of OLIS, injection magnet and TITAN is very similar to the proposed setup, except the analyzing magnet and electrostatic analyzer are replaced by a Wien-filter.

The TITAN facility [20] (Fig. 5) at ISAC has essentially all important components to perform a proof-of-principle experiment of the aforementioned concept.

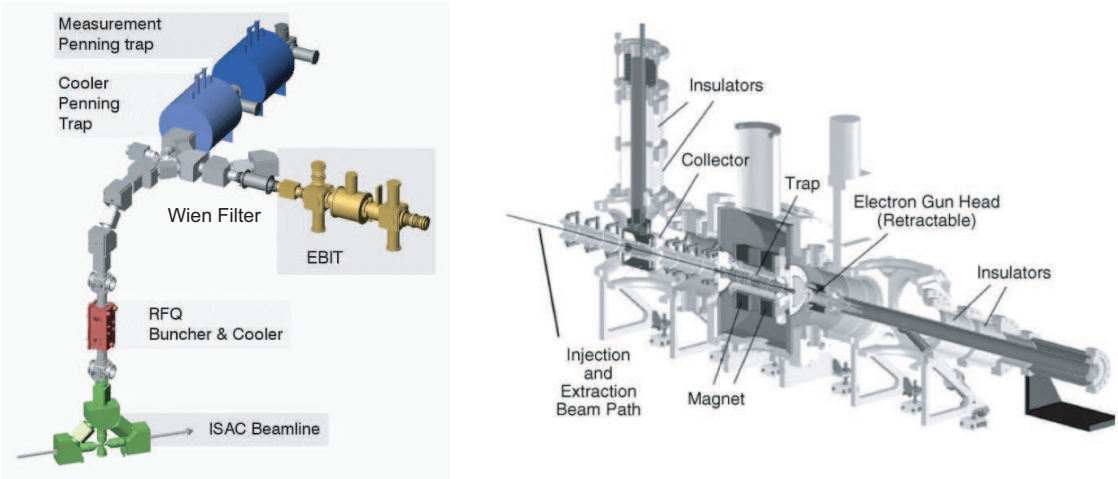

Figure 5: Schematic setup of the TITAN facility (left) and the EBIT (right).

3 Detection of ^{41}Ca – proof-of-principle experiment

In order to test this new concept, we propose to start with a light nuclide, ^{41}Ca ($t_{1/2} = 1.03 \times 10^5$ yr). It is one of the classical AMS nuclides with many applications from biology to astrophysics [21], which will not be further discussed here. The isotopic ratios relevant for many applications are very low: ^{41}Ca/^{40}Ca ratios ranging from 10^{-10} down to 10^{-16}. The nuclear charge Z of ^{41}Ca is 20; the interfering stable isobar is ^{41}K with a Z of 19. Thus, by full stripping of ^{41}Ca to charge state 20+ and a further m/q analysis it can be unambiguously identified because $^{41}K^{20+}$ does not exist. Note that molecular isobars like ^{40}CaH are completely destroyed if stripped to high charge states due to coulomb dissociation.

We will start with a sample material containing a relatively high ^{41}Ca/^{40}Ca ratio of 10^{-10} (AMS standard material). The OLIS surface ion source will be loaded with a few mg of that material and $^{41}Ca^+$ ions will be extracted. Interference from doubly charged $^{82}Kr^{++}$ is unlikely because of the high second ionization potential (> 30 eV). ^{40}Ca is separated by the dipole magnet in the extraction beam line. Ions of mass 41 (depending on the source condition probably mainly $^{40}CaH^+$, and only very little $^{41}Ca^+$ and $^{41}K^+$) will be transported to the TITAN facility, where they are first cooled and bunched in an RFQ cooler and then injected into the EBIT where they are trapped. An intense electron beam provides the charge breeding. After a few 10's of milliseconds the highly charged ions are released from the trap and extracted by high voltage (Fig. 6). The energy depends on the charge state and is calculated as $E_{\text{ext}} = qU_{\text{ext}}$ with $U_{\text{ext}} = 5$ kV. The highly charged ions are then separated by their mass-to-charge ratios in a Wien-filter (with a nominal resolution of 300).[3] By selecting ions with a mass-to-charge ratio of 41/20, only ^{41}Ca ions should arrive at the final detector (Fig. 4). ^{41}K ions can reach only a $m/q = 41/19$ and thus can be separated by selection slits. In a similar way, ^{40}Ca ions entering the EBIT as ^{40}CaH ions are separated by a different $m/q = 40/20$.

A micro-channel plate (MCP) detector will be used to count the ions. The fast timing of the MCP together with the pulsed beam from the EBIT allows for a time-of-flight (TOF) measurement, which can provide additional separation. In principle, a Si detector or a small ionization chamber could be also used to count the ions and measure their energies to identify lighter molecular break-up products[4]. However, the energy of the Ca ions $E = 20 \cdot 5 = 100$ keV would be too small. To boost the detection energy, the trap in the EBIT could be floated to higher voltages (up to 60 kV). Alternatively, the detector could be kept at high voltage (e.g. 100 kV). The final detection energy is then calculated as $E_{\text{det}} = q\ (U_{\text{ext}} + U_{\text{det}})$. For higher masses (e.g. fully stripped ^{146}Sm with $q = 62$) we can reach energies of a few 100 keV. It should be noted that the TITAN Penning trap used for high precision mass measurements can also act as an excellent detector as it provides a very high mass resolution up to 10^8, enough to separate certain isobars by its mass difference.

In order to be independent of the ionization efficiency in the ion source, the isotopic ratio to a stable isotope will be measured.[5] This will be done by selecting mass 40 and 41 alternately: The majority of the time mass 41 will be selected for ^{41}Ca counting, whereas the stable isotope ^{40}Ca will be measured as a current in a Faraday cup after the first bending magnet from time to time.

[3] A Wien-filter combines a magnetic and an electrostatic field and selects velocity or $v = \sqrt{2E/m}$; since in our case $E = qU_{\text{ext}}$, we get a selection by $\sqrt{2qU_{\text{ext}}/m}$, i.e depending only on m/q.

[4] In principle, an optimized ion chamber is preferred over a Si detector because it would have a better energy resolution at low energies. However, any gas leaking through the window would introduce problems from charge changing reactions of the highly charged ions.

[5] Mass fractionation effects between isotopes should be small and thus should not play a significant role at our precision level; but it could be investigated by measuring other stable isotopes; in particular ^{42}Ca, ^{43}Ca and ^{44}Ca are free of stable isobars.

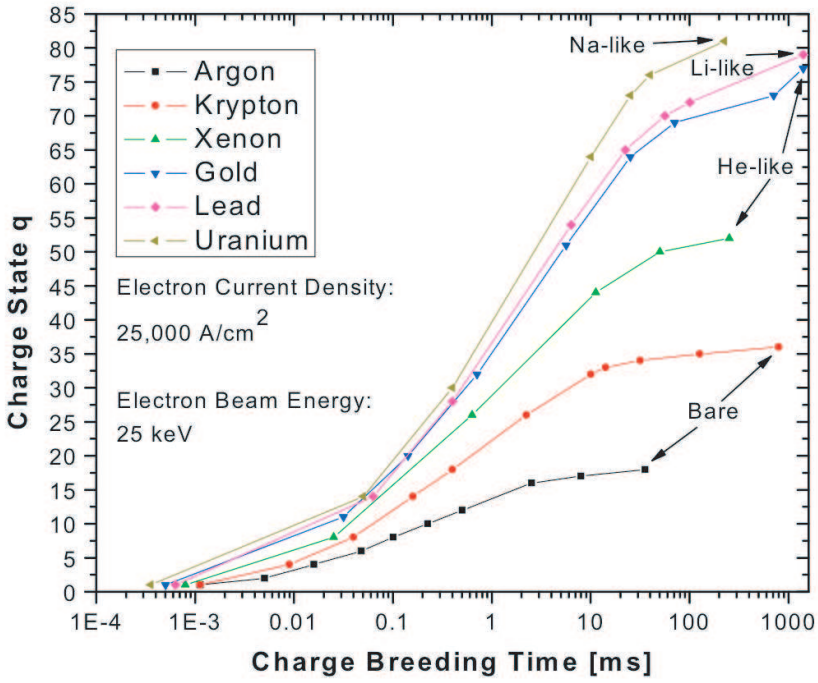

Figure 6: Expected charge breeding time at the TITAN EBIT for several elements. Bare nuclei in mass 40 region can be reached within a few 10 ms. Figure taken from [22].

This sequence will be repeated through the entire measurement time until enough ^{41}Ca statistics are collected or the sample material is used up in the ion source. Compared to a dedicated AMS facility with fast sequencing, the slow sequencing allows only for a moderate precision. However, in this proof-of-principle experiment we don't aim for a high precision, and we don't plan to compete with a dedicated AMS facility in the first place.

The efficiency of the various steps are critical for the ultimate sensitivity and the measurement precision. The ion source is, in principle, independent from the detection setup and affects also the stable isotope ^{40}Ca whereas the efficiency of the detection setup is decisive only for the radionuclide ^{41}Ca. Thus, the two parts can be evaluated individually. We will concentrate in this proposal on the efficiency of the detection setup. The first component is the RFQ cooler and buncher with a measured efficiency of 68% for ^{133}Xe. The most critical part is the charge breeding process in the EBIT. Depending on the breeding time, an efficiency of 20% can be estimated for a fully stripped ^{41}Ca ion. Beam transport accounts for losses of 50%. This results in a total detection efficiency of about 7%.

Assuming we have a sample with a ^{41}Ca/^{40}Ca ratio of 10^{-10} and a stable ^{40}Ca beam from the ion source of 100 nA, we can expect 62 ^{41}Ca per second enter the detection setup. With the total detection efficiency of 7% we expect a count rate of about 260 ^{41}Ca per minute at the final detector.

The question of background is hard to predict without experimental experience. In principle, ions should be well separated after the Wien-filter according to their m/q. However, due to the high isotopic ratios, small background effects, which are usually negligible, become the main limitation. In conventional AMS for instance, charge changing processes of intense molecular break-up products (e.g. ^{40}Ca from the ^{40}CaH molecule) are a significant source of background [23]. In order to reduce that background, a sequence of several alternating analyzing elements are used. In our case, charge changing could be a major problem because of the highly charged ions. Thus, the vacuum in the

beam line should be as good as possible. The vacuum in the TITAN beam line is in the range of 10^{-9} Torr. Eventually, a sample with a lower ^{41}Ca/^{40}Ca ratio or a blank sample will be investigated to demonstrate the feasibility of the method to measure very low isotopic ratios.

3.1 Experimental Equipment

The following equipment will be used: OLIS surface ion source, bending magnet for mass separation; TITAN facility: RFQ cooler and buncher, EBIT, Wien-filter, MCP detector, (Penning trap).

3.2 Readiness

All the equipment will be ready as soon as the EBIT is fully implemented in the TITAN beam line.

3.3 Beam Time required

We anticipate 10 12-h shifts for the proof-of-principle experiment which includes investigation of the mass separation after the ion source, setting up the RFQ cooler and buncher, beam transport, EBIT and detector, study the isobar separation ^{41}Ca – ^{41}K and background from ^{40}Ca. Efficiencies of each component will be determined with stable ^{40}Ca.

3.4 Data Analysis

Data analysis will be done online.

4 Prospects for ^{146}Sm detection

Although it is not planned to detect ^{146}Sm at the early stage of this experiment, we discuss briefly the conditions for ^{146}Sm – ^{146}Nd separation at the TITAN facility.

The nuclear charge of ^{146}Sm is $Z = 62$ and for the interfering stable isobar ^{146}Nd it is $Z = 60$. Thus, in principle H-like ($q = 61$) Sm could be used as well as since the maximal charge state of Nd is $q = 60$. The binding energy of the s-electron in Sm is 46.834 keV [24]. Thus, an electron beam with an energy of $E > 50$ keV is required to fully ionize ^{146}Sm, which is within the range of the TITAN EBIT with 60 keV maximal electron energy. The resolution in m/q required to separate the isobar is 73 or 146 for bare and H-like ^{146}Sm, respectively, which is also well within the resolution of the TITAN Wien-filter. In order to test the isobar separation, the naturally occurring long-lived/stable isobar pair ^{148}Sm/^{148}Nd can be used. The introduction of a sample containing a high concentration of ^{146}Sm should be avoided in order not to jeopardize the supernova search with a contamination of the ion source.

A critical point is the charge breeding efficiency. A high value is desirable. The electron energy could be raised to 80 keV. The maximal electron current density is 40,000 A/cm^2 among the highest values for existing EBITs. The charge breeding time could be extended to a few 100 ms increasing the efficiency for the highest charge states.

The production and ionization efficiency of Sm in the ion source could be investigated independently. The current setup is not optimized for such a kind of measurements as it usually doesn't run on small samples. Improvements of the ion source might be necessary.

To this end, the possibility of a detection of supernova produced ^{146}Sm seems to be, at least in principle, within reach. However, first the concept of this new measuring method has to be demonstrated by this proposed proof-of-principle experiment for ^{41}Ca. If successful a dedicated experiment for ^{146}Sm will be proposed.

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