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Development of isobar separation for ^{182}Hf AMS measurements of astrophysical interest

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Abstract

The detection of ^{182}Hf by Accelerator Mass Spectrometry (AMS) is greatly complicated because of the stable and common isobar ^{182}W. Although significant W reduction can be achieved during negative ion formation using HfF_5^-, additional separation is necessary to achieve detection sensitivities sufficient for astrophysical applications, i.e. detection of potential supernova-produced ^{182}Hf on Earth. In this paper, we present a new development of isobar separation using the ΔTOF detection method at the Munich MP Tandem accelerator (TV = 14 MV), where ion energies for ^{182}Hf of about 1 MeV/amu can be achieved. Particular attention is drawn on specific energy loss and energy loss straggling measurements in various materials, the basis for our method of isobar separation.

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Keywords: ^{182}Hf; Isobar separation; Specific energy loss; Energy loss straggling

1. Introduction

Isobar separation is one of the main challenges in Accelerator Mass Spectrometry (AMS). Only a few, nevertheless very important, radionuclides have the advantage that potential interfering stable isobars do not exist (e.g. for the actinides) or do not form negative ions (e.g. ^{14}N for ^{14}C detection). Thus an AMS facility based on a tandem accelerator is used in most cases. The remaining candidates require isobar separation at the detector, with the interfering stable isobar often several orders of magnitude more intense than the radionuclide. Only the lighter radionuclides (e.g. ^{10}Be, ^{36}Cl, ^{41}Ca) are routinely measured at small or medium-sized accelerators up to 5 MV terminal voltage (TV) providing energies around 1 MeV/amu. In the medium mass range (e.g. ^{60}Fe, ^{63}Ni) large tandem accelerators (up to TV = 14 MV) are necessary to provide a high enough energy (around 3 MeV/amu) for isobar separation [1]. For even higher masses (e.g. ^{182}Hf) up to now no method for isobar separation at the level of several orders of magnitude difference in abundance exist.

Most separation methods are based on a difference in specific energy loss depending on the nuclear charge Z. Fig. 1 shows calculated differences in stopping power for three ion energies. It clearly illustrates the reduction in stopping power difference with increasing Z. For ^{182}Hf the expected difference to ^{182}W is only around 2%, even with a difference in Z by two. Due to this small difference in specific energy loss between ^{182}Hf ($Z = 72$) and ^{182}W ($Z = 74$), even at 200 MeV, conventional methods like an ionization chamber (e.g. see Fig. 2) or a gas-filled magnet are not applicable. In this paper we present our status of a new separation method at the Munich 14 MV tandem accelerator which is based on passive absorption and time-of-flight measurement (called ΔTOF [2]), initially

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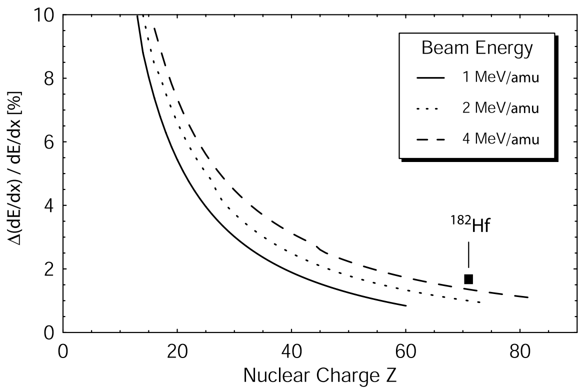

Fig. 1. Differences in stopping power in isobutane (C_4H_{10}) between two neighboring isobars depending on nuclear charge Z for three ion energies. Stopping powers are calculated as outlined in Section 4. For each element the stable (or long-lived) isotope with the highest mass is used. The difference is calculated from the next lower element of the same mass. The lines are truncated below a difference of 1% because of artifacts in the calculation. The position for ^{182}Hf at 1 MeV/amu is indicated, however, here the difference in Z to ^{182}W is two.

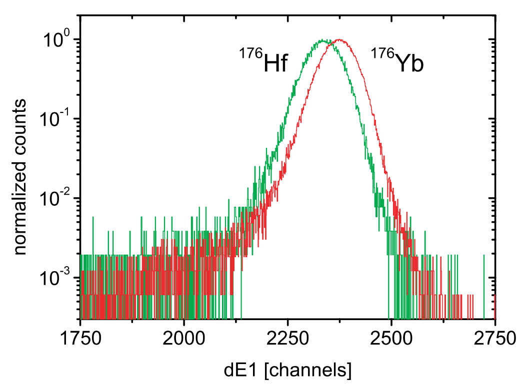

Fig. 2. Separation of isobars (^{176}Hf, $Z = 72$ and ^{176}Yb, $Z = 70$) at 200 MeV in a segmented ionization chamber. The energy loss signal in the first segment (dE1) is shown. Only a small shift of the peaks is visible. Isobar separation is not possible here.

developed at VERA for ^{36}Cl and ^{41}Ca at a 3 MV tandem accelerator (see also [3,4]).

2. Astrophysical motivation for AMS measurements of ^{182}Hf

The relevance of the extinct radionuclide ^{182}Hf (half-life of 8.90 ± 0.09 million years [5]) for various applications in geo- and astrophysics is summarized in [6]. The main motivation for detection of ^{182}Hf by AMS is because live ^{182}Hf can be expected to be found on Earth today from recent and nearby nucleosynthesis events (e.g. supernovae). A positive signal would help to confirm the ^{60}Fe signal recently found in deep-sea Fe–Mn crusts [7] and to test nucleosynthesis models.

A first attempt to find live ^{182}Hf in deep-sea sediments using low-energy AMS based on a 3 MV tandem accelerator resulted only in an upper limit, mainly due to the high

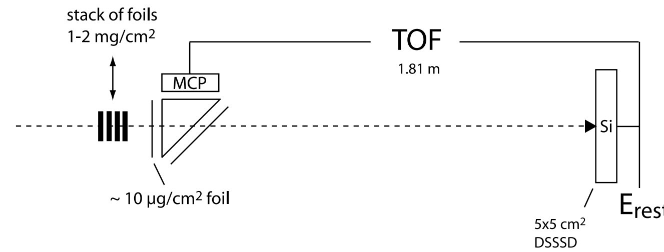

Fig. 3. Schematic setup of the ΔTOF detector.

background of the stable isobar ^{182}W [8]. Although suppressed by chemistry, and in the ion source by using $^{182}HfF_5^-$ by a factor of 6000 [9], isobar separation at the final detector is necessary to reach the desired detection limit, which is only possible at high ion energies.

3. Principle detector design

Fig. 3 shows the principle design of the ΔTOF detector, which comprises a stack of energy degrader foils and a subsequent high resolution TOF setup. Isobars are accelerated to the same velocity but lose different energy in the energy degrader foils due to their different nuclear charge Z which leads to a difference in velocity and time of flight.

The start detector is based on secondary electrons emitted from a thin C-foil and detected by a fast micro channel plate (MCP). As a stop detector we use a double-sided silicon strip detector (DSSSD, Micron Semiconductor Ltd., Lancing, UK) with a size of 5×5 cm^2. The segmentation of 16 front and 16 back strips allows to correct for position dependent signals. For a fast timing signal, preamplifiers with integrated shaper and timing filter (Mesytec, Putzbrunn, Germany) are used. With bias voltages at the DSSSD up to 150 V a time resolution of less than a nanosecond could be achieved. The corresponding energy resolution depends on the respective energy of the ions, but is typically around 1%. The DSSSD also provides information of the residual energy E_{rest} which is useful for discrimination against E/q ambiguities during rare isotope AMS measurements.

Up to three different stacks of energy degrader foils can be mounted on the moveable foil ladder in front of the start detector. Different materials were tested: silicon nitride membranes of high uniformity (here simply called SiN) and two different sets of carbon foils optimized for high homogeneity.[1] The total thickness of the stack is 1–2 mg/cm^2.

There are three critical parameters for an optimal detector design. The first one is the specific energy loss in the degrader foil responsible for isobar separation. The second one is energy loss straggling which is very critical for the achievable resolution. These two are the most critical parameter and difficult to simulate accurately. The

[1] Obtained from P. Maier-Komor.

third parameter is angular scattering in the degrader foil, which is not critical for separation and resolution, but must be considered for an efficient detector design (see Section 5).

4. Energy loss and energy loss straggling

Attempts to improve the understanding of energy loss of heavy ions in matter have been performed since many decades (see e.g. [10]), however, measured values for stopping power and energy loss straggling of heavy ions ($A \gtrsim 100$) around 1 MeV/amu are very scarce or missing entirely. In addition, measurements of energy loss straggling are often complicated by target inhomogeneities and insufficient measurement resolution.

As shown already some time ago energy loss straggling in solids is less than in gases [11]. Thus a foil is preferred over a gas absorber cell. However, in the past the use of not sufficiently homogeneous foils often covered this effect (see e.g. [12]).

Although in most cases not reliable enough for accurate predictions of differences in the 1–2% range, we performed simulations with common codes to get an idea of the basic physical behavior. Most of our simulations of the specific energy loss are based on the Ziegler formalism [13] implemented in a Mathematica package [14], which seem to give acceptable results.

We studied isobar separation using the stable isobar pair ^{176}Yb and ^{176}Hf, because they can be easier produced in the real world. ^{176}Yb has the lower Z (70) and thus represents ^{182}Hf ($Z = 72$); ^{176}Hf then corresponds to ^{182}W ($Z = 74$), respectively. Three energies were studied, 150 MeV, 175 MeV and 200 MeV. All three are possible at the Munich 14 MV tandem accelerator with reasonable stripping yields for the respective charge states. Two target materials for degrader foils were used for simulations and measurements, carbon and silicon nitride.[2] In addition to the setup described in Section 3 the high resolution spectrometer Q3D [15] at another beam line was used for measuring energy loss and energy loss straggling. The very first ΔTOF investigations were carried out using two MCP based timing detectors with a TOF resolution of ~500 ps and a long flight path of 3.5 m.

We define separation D as the distance between the peaks of ^{176}Yb and ^{176}Hf in MeV. The measured separation in energy is calculated from the TOF or the position at the focal plane detector at the Q3D, respectively. Suppression of isobars depends also critically on the peak shape which comprise the width of the peaks (W = FWHM) as well as their tails. Since in our case the separation is small, W still dominates the suppression and we use the term $S = D/W$ as a measure for separation power of foil-energy combinations.

The measured width of the peaks (W_{meas}) depends on several parameters:

$$W^2_{\text{meas}} = W^2_{\text{straggl}} + W^2_{\text{foil}} + W^2_{\text{det}} + W^2_{\text{beam}} \quad (1)$$

with W_{straggl} the energy loss straggling, W_{foil} the straggling from foil inhomogeneities, W_{det} the detector resolution and W_{beam} the beam energy spread. The last two are not considered as physical measurement limits and can be measured without a degrader foil in place. For comparison of various foil-energy combinations with setups of different detector resolution we use the corrected W

$$W^2 = W^2_{\text{meas}} - (W^2_{\text{det}} + W^2_{\text{beam}}). \quad (2)$$

The simulated width of the peaks contains only the energy loss straggling (W_{straggl}) calculated according to the empirical formula from Yang et al. [16]. It should be mentioned that this formula is only valid for "thin" targets, i.e. where the stopping power is not changing during deceleration of the ions. For thicker foils this should be taken into account by integrating over the energy loss. Additionally, the energy-loss straggling in various deeps intervals is correlated. For our case (Hf/Yb below 200 MeV) the stopping power is continuously decreasing, which means ions which lose more energy in the first half lose less energy in the second half and *vice versa*. This effect should lead to an energy focusing (as discussed in [17–19]) and thus a smaller width of the peaks. However, our simulations do not take these effects into account.

The foil thickness was determined from the energy loss of the ions and the stopping power of [14]. A good agreement between values obtained in different runs at different energies and with different number of foils was found with a scatter in the order of a few percent.

Fig. 4 summarizes the results of both simulation and measurement. In Fig. 4(a) the simulated separation in MeV between the two isobars is plotted against foil thickness (in μg/cm^2). The maximum separation peaks at slightly larger foil thickness for SiN foil compared to C-foils. This is as expected because of the higher Z of the target atoms. Fig. 4(b) shows the simulated energy loss straggling (FWHM) in MeV and Fig. 4(c) the separation power $S = D/W$. From these simulations a higher isobar separation can be expected with SiN at slightly larger foil thickness compared to C.

The measured data (Fig. 4, right column) are plotted in the same way as the simulations (Fig. 4, left column) for direct comparison. The measured separation between ^{176}Yb and ^{176}Hf (Fig. 4(d)) is about a factor two larger than in the simulations, underlining the problem of accuracy of the simulations. Energy loss straggling on the other hand is reproduced by the Yang formula reasonably well (Fig. 4(e)). This indicates that the foils used in this work are very homogenous and have minor influence on the peak width. However, energy tails are significantly larger with

[2] For the simulations the stoichiometric correct form of Si_3N_4 was used, although most available SiN foils have a composition of $Si_3N_{3.1}$ in order to get them "stress-free". However, a check of both compositions gave very similar results in the simulations and the small differences does not change any conclusions.

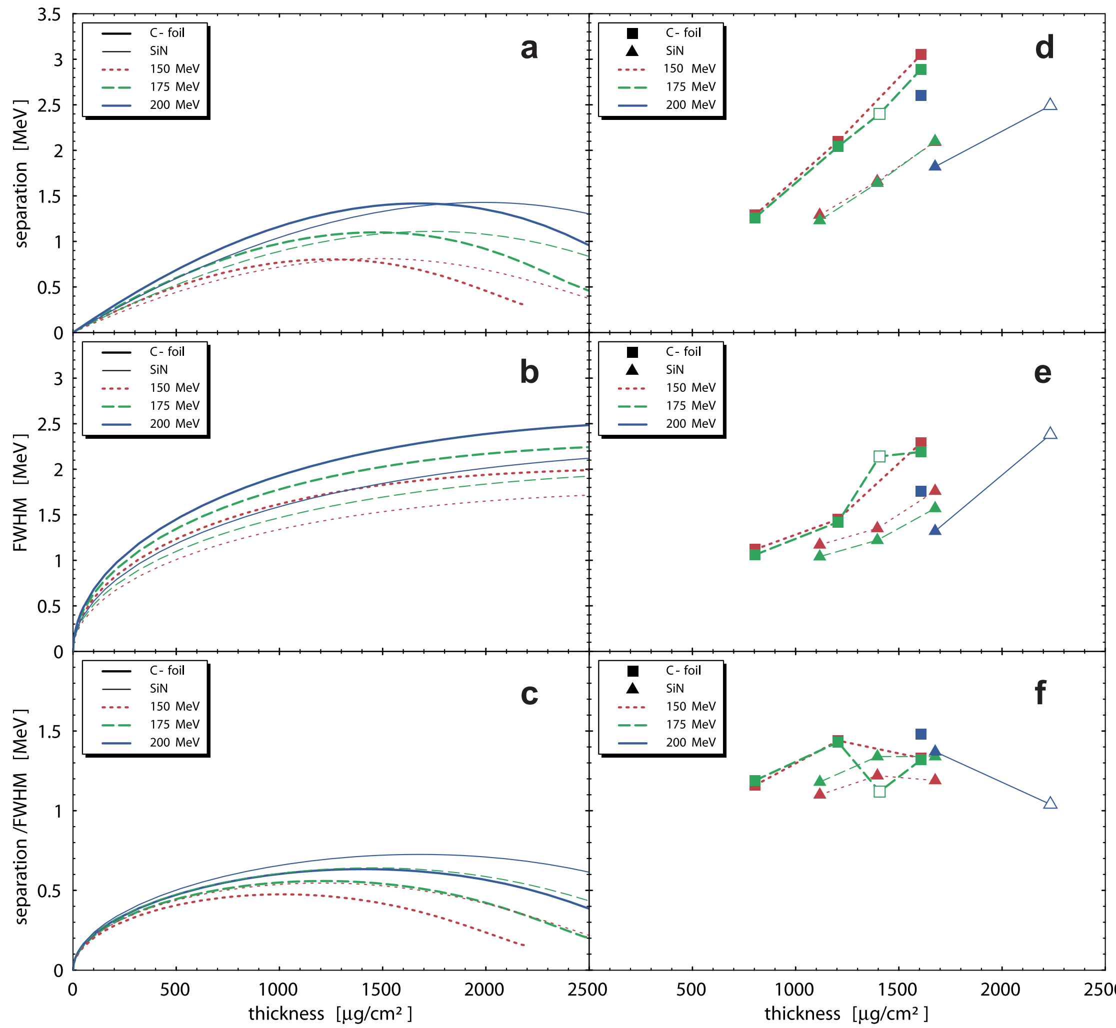

Fig. 4. Summary of simulated (left column) and measured (right column) separation between ^{176}Yb and ^{176}Hf. Filled symbols denote measurements at the Q3D spectrometer, open symbols using ΔTOF.

C-foils compared to the SiN foils. The measured separation power $S = D/W$ (Fig. 4(f)) is greater than expected from the simulations, although the expected trend depending on foil thickness is not clearly visible because of some small random uncertainties between different measurement runs.

5. Angular scattering and detector efficiency

Although our focus was mainly the optimization of separation, we can have a look at the angular scattering and detector efficiency. As shown in [2] for scattering of Cl ions on SiN reliable simulations of angular scattering could be made with TRIM98 and later SRIM2003 [20]. Fig. 5 shows simulated distributions of 200 MeV ^{182}Hf ions scattered on C and SiN. As expected SiN scatters more compared to C-foils because of the higher Z of the target atoms.

These distributions can be compared to actual detector efficiencies, measured as the count rate at the DSSSD divided by the count rate at the start detector during our separation studies. Measured efficiencies are close to the values expected from the SRIM simulations, e.g. for 200 MeV ions between 20% and 30%.

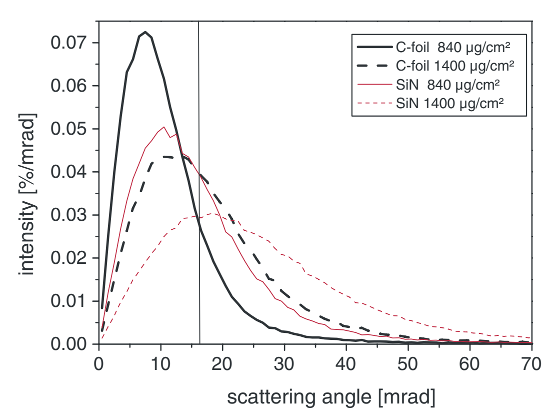

Fig. 5. SRIM simulations of scattering of 200 MeV ^{182}Hf ions on carbon and SiN foils of various thickness. The acceptance of the described ΔTOF setup is indicated as the vertical line.

6. Isobar suppression factor

The relevant quantity for the AMS measurement is the isobar suppression factor, defined as the counts in the main background peak divided by the counts in the cut for the isobar of interest. A reasonable compromise between efficiency and background suppression is achieved by cutting at the maximum of the distribution of the isobar of interest. This reduces their number by a factor of ~2 which must be included in the total efficiency calculation.

With the Q3D spectrometer energy loss and peak width (FWHM) can be measured very precisely, however, a constant background at the 10^{-2} level prevents higher suppression factors than 100 (Fig. 6). This background stems most likely from energy loss tails from other charge states or image aberrations from ions scattered to very large angles.

The ΔTOF detector has the advantage to be insensitive to scattering, and different charge states are not distinquished. Fig. 7 demonstrates a clear separation of isobars with the ΔTOF detector equipped with SiN foils. Besides the significant separation between the peaks of ^{176}Yb and ^{176}Hf we have to point out the almost perfect Gaussian shape of the peaks, a result of the perfectly homogeneous SiN membranes. Only low energy tails at a 10^{-3} level are visible and no high energy tails. This is particularly important for ^{182}Hf detection since it is expected at the high energy side of the interfering ^{182}W peak. Suppression factors are up to three orders of magnitude.

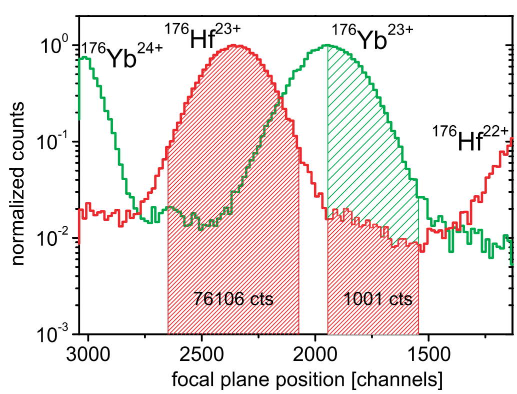

Fig. 6. Separation of isobars (^{176}Hf and ^{176}Yb) at 175 MeV with the Q3D spectrometer with SiN foils of 1.68 mg/cm^2 thickness. The focal plan position represents E/q and is reversed to show low E/q values to the left. The isobar suppression factor is 76,106/1001 = 76.

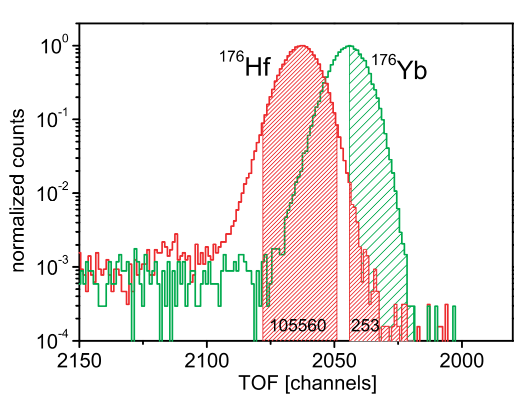

Fig. 7. Separation of isobars (^{176}Hf and ^{176}Yb) at 200 MeV with the ΔTOF detector equipped with SiN foils of 1.4 mg/cm^2 thickness. The TOF scale is reversed to show low energies to the left. The spectra were taken with the ΔTOF setup consisting of two MCP based timing detectors. The suppression factor here is 105,560/253 = 417.

7. Summary and outlook

Although not fully optimized at the moment, we could achieve significant progress in isobar separation of heavy ions. Highly homogeneous degrader foils made of carbon and SiN in combination with a high resolution TOF setup result in separation of isobars close to the physical limit of energy loss straggling. Since the separation is still dominated by the width of the peaks, small improvements in resolution (e.g. time resolution of the DSSSD) and separation can result in a significant improvement in isobar separation. A reasonable detector efficiency could be achieved, however, flight path and stop detector size can be further optimized for highest possible detection efficiency.

Our best achieved isobar suppression was a factor of 940 with an effective efficiency of 13%. Applying the ΔTOF technique for ^{182}Hf detection it should be possible to suppress the stable isobar ^{182}W up to three orders of magnitude with reasonable efficiency of >10%.

Besides isobar separation, additional work is necessary for successful AMS measurements of ^{182}Hf. Particularly, the improvement of low transmission through the tandem accelerator, since Coulomb explosion of HfF_5^- seems to increase the emittance of the beam after the terminal stripper foil significantly. Experience with ^{182}Hf gained at other large tandem accelerators (i.e. at the 14 MV Pelletron in Rehovot/Israel [21], the HI-13 AMS System at China Institute of Atomic Energy (CIAE) in Beijing/China [22] and ANU 14UD tandem in Canberra/Australia [23]) might be useful. Furthermore, the developments of isobar separation using projectile X-ray emission AMS (PXAMS) and a similar approach using degrader foils followed by an ionization chamber by [23,24] bears great potential.

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