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Opportunities and Limits of AMS with 3-MV Tandem Accelerators

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Abstract

With the Vienna Environmental Research Accelerator (VERA, based on a 3-MV tandem accelerator) we have systematically explored the limitations due to terminal voltage, both by modelling and by experiments. If no stable atomic isobars exist, e.g. for ^{236}U or ^{244}Pu, the same detection limits as for large machines have been achieved. In cases where isobar separation is required, the achievable energy is the limitation.

To study isobar separation experimentally, we use a new method called ΔTOF: the different energy loss in a foil is measured with a time-of-flight (TOF) detector. Separation of ^{36}Cl from ^{36}S is possible. With Diamond Like Carbon (DLC) stripper foils (0.6 μg/cm^2) and by pushing our Pelletron tandem to 3.6 MV, we achieved a significant increase in energy compared to gas stripping at 3 MV: e.g., ^{36}Cl^{7+} at 28.8 MeV instead of ^{36}Cl^{5+} at 18 MeV can be used.

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1. Introduction

About half of the existing AMS facilities are based on 3-MV tandem accelerators. These "work horses" of AMS allow for routine measurements of ^{10}Be, ^{14}C, ^{26}Al, and ^{129}I, with a large number of successful applications. However, many more interesting, long-lived natural radionuclides exist which presently can only be investigated at larger tandems with higher terminal voltage. These nuclides are either very heavy (especially ^{236}U [1,2] and ^{244}Pu [3,4,5]) or there is a stable isobar which is not suppressed during negative ion formation (e.g. ^{36}Cl – ^{36}S, ^{41}Ca – ^{41}K, ^{60}Fe – ^{60}Ni [6]). 3-MV tandems can so far achieve sufficient isobar suppression only for the lightest long-lived radionuclide, ^{10}Be (^{10}Be – ^{10}B). The advantages of larger accelerators and thus higher ion energies are a direct result of the underlying physical processes.

To estimate stripping yields, energy straggling and small angle scattering, we used existing computer codes to model the AMS measurement process. Very helpful in this context was a software package described by Weller [7] implemented in Mathematica (Wolfram Research). The package, originally intended for RBS (Rutherford Back Scattering) purposes, uses Ziegler's formalism [8] and the tables of TRIM (Transport of Ions in Matter) to calculate stopping powers. The energy straggling is implemented following [9], the small angle scattering uses a semiempirical formula from [10]. The charge state distributions were estimated following the formalism of [11].

A new kind of foil has emerged as a valuable "tool" for AMS: silicon nitride membranes of 30 to 1000 nm thickness (Silson, UK). These foils show superior homogeneity [12, 13], which was crucial for our experiments.

2. Actinides

Energy straggling and angular scattering occur whenever the ion beam passes through matter. For nuclides where stable isobar suppression is not required, this happens only in the terminal stripper. For the energy range below 1 MeV/amu, our simulations predict that the energy straggling ΔE grows with $E^{0.5}$. However, since the interesting quantity for ion separation is ($\Delta E/E$), which is proportional to $E^{-0.5}$, higher energies are advantageous too. The same holds for small-angle scattering, where the computer codes predict a behavior with E^{-1}. Fortunately, even in the case of the heaviest nuclides, straggling and scattering are small: e.g. for 0.2 μg/cm^2 oxygen gas the expected energy straggling of ^{236}U at 3 MeV is ~ 3 keV (FWHM), and the angular scattering is 5 mrad (FWHM).

Acceleration reduces the beam emittance with $E^{-0.5}$ (both in horizontal and vertical direction). Thus at given dimensions of magnetic and electric separators, lower energy leads to lower achievable mass resolution. This is a disadvantage especially for very heavy isotopes, where the relative mass difference is small. The straightforward solution is to use larger analyzing elements. This was realized at VERA (Fig. 1) where an analyzing magnet with a radius of 1270 mm and an electrostatic separator with a radius of 2000 mm are used [14]. Together, they form a mass spectrometer which is operated at a mass resolution of M/FWHM(M) ~500.

Background ions, which should be completely suppressed by these large separators, can interact with residual gas along the beam line and reach the final rare isotope detector on arbitrary paths. The most likely interaction process is charge exchange. The corresponding cross sections are not well measured and are expected to go with E^{-x}, with the exponent $x > 0$ depending on the process (single electron loss, electron capture, multiple electron loss,…) and on the model [15]. This type of background was clearly observed in our first measurements on ^{236}U with a time-of-flight setup [16]. However, since these ions undergo charge exchange, they are either before or after the main mass spectrometer in the wrong charge state and thus can be separated by additional ion optical filters of comparable low resolution. This is accomplished at VERA with a small Wien filter and the switcher magnet [14].

In a recent measurement on natural ^{236}U a bolometric cryo detector [17] was used, which provides higher efficiency than our usual TOF setup. Even though the detector could not distinguish ^{236}U and ^{235}U of the same magnetic rigidity an upper limit of ^{236}U/U < $6 \cdot 10^{-12}$ was determined for uranium separated from a well in Bad Gastein, Austria. This is the first ^{236}U measurement on uranium which does not originate from uranium ore, and the low ^{236}U/U ratio is consistent with expectations [18]. This measurement proofs that 3-MV tandems can offset the disadvantage of lower terminal voltage up to the heaviest masses, if heavy isobar suppression is not required. Also the sensitivity is similar, since the ion optical losses are kept very small at VERA.

3. Energy vs. terminal voltage

Since higher charge states are populated at higher terminal voltages, the achievable energy grows faster than linear with the terminal voltage. For quantitative estimates, an approximation is needed. We select the commonly used negative ion (e.g. ^{36}Cl^{-} or ^{10}Be^{16}O^{-}) and calculate its energy at the accelerator terminal. The charge state distribution is estimated by the formula from [11]. The decision for a certain charge state is a crucial part of every AMS experiment, and a general rule cannot be given. For our investigations we simply select the highest charge state with a yield of at least 5% (Figure 2). The achievable energies can be approximated in the form

$$E \propto U_{\mathrm{term}}^{a} \tag{1}$$

Besides the case of ^{10}Be, where full stripping is achieved for higher terminal voltages, the exponents a obtained for ^{36}Cl, ^{60}Fe, ^{182}Hf, and ^{236}U are close to 1.3 both for gas and foil stripping. Thus, we can expect that a 5-MV tandem achieves twice the energy than a 3-MV machine, and a 15-MV tandem the 8-fold energy. Foils populate higher charge states, and the resultant energy gain is about 50% near 3-MV terminal voltage. Considering formula (1), this energy gain would require a terminal voltage increase from 3.0 to 4.2 MV.

4. Separation of isobars

Stable isobars are not critical in AMS if they do not form negative ions. If they do, separation of isobars is usually based on the different energy loss in matter. This is done either by an active measurement of the energy loss in an ionization chamber or by measuring the residual energy loss after a passive absorber. The principal limitation is the energy straggling competing with the separation. Additional technical limitations are inhomogeneities of foils and detector windows, causing energy spread and low energy tails, and detector properties like electronics noise and incomplete charge collection.

Figure 3 models this for ^{36}Cl and ^{36}S with an initial energy of 18 MeV. As a measure for the suppression we use the ratio

$$S_{\mathrm{A,B}} = \frac{\left|E_{\mathrm{A}}(x) - E_{\mathrm{B}}(x)\right|}{\sqrt{\frac{1}{2}[\sigma_{\mathrm{A}}^{2}(x) + \sigma_{\mathrm{B}}^{2}(x)]}} \tag{1.1}$$

with $E_{\mathrm{A}}(x)$ and $E_{\mathrm{B}}(x)$ the residual energies of ion A and B, respectively, after passing through a layer of silicon nitride (see below) of thickness x. $\sigma_{\mathrm{A}}(x)$ and $\sigma_{\mathrm{B}}(x)$ are the distribution widths caused by energy straggling. The best separation is achieved at $x \approx 750$ µg/cm^2. Figure 4 shows the best separation for ^{36}Cl/^{36}S for different terminal voltages. Higher energies are clearly advantageous for isotope separation.

Passive absorber foils have been successfully used to separate ^{10}Be from ^{10}B [19] and were investigated to separate ^{32}Si from ^{32}S [20]. We have used Time-of-Flight detectors to measure the residual energy. With this method, which we call ΔTOF [21], the energy resolution can be made arbitrarily high by increasing the flight path. This allows studying the physical limitations due to energy straggling independently from detector properties. For the separation of ^{36}Cl/^{36}S, the ^{36}Cl ions have lower residual energy than the ^{36}S ions after the absorber. Thus, the ^{36}Cl is easily buried in the low energy tail of ^{36}S (Figure 2 of [21]). The almost complete absence of low energy tails was crucial for our measurements.

Figure 5 shows the spectrum obtained with 2 × 1000 nm silicon nitride foils for a material with ^{36}Cl/Cl ~ 10^{-10}. A separation of $S = 4.5\ \sigma$ was achieved. The suppression of ^{36}S in the region of interest is ~10^{3}. The results of a dilution series with ^{36}Cl samples in the range from 10^{-10} to 10^{-13} showed a detection limit of about 10^{-12}, mainly because of a high ^{36}S count rate. The detector efficiency was only 0.14% with this setup. Ref. [21] discusses the reasons for this low efficiency and possible improvements of the setup. This paper also presents first tests on the separation of ^{41}Ca – ^{41}K and ^{60}Fe – ^{60}Ni.

Additional to ΔTOF, we investigated the separation of the different residual energies with our high-resolution ESA (Electrostatic Analyzer). With a 500 nm silicon nitride foil mounted at it's object slits, a suppression factor of 170 for ^{36}Cl - ^{36}S was achieved. Also for this method, angular scattering limits the detection efficiency: the ion optical transmission achieved was only 3%. However, since the dimensions of the ESA are given, an improvement in efficiency is hard to realize with the current setup.

5. Diamond-Like Carbon (DLC) stripper foils

Although it provides higher energy, foil stripping is rarely employed in 3-MV tandems. Usually, less than 0.5 µg/cm^{2} are sufficient to reach charge state equilibrium, and gas strippers are operated at these areal densities. Conventional stripper foils cannot be made thinner than ~2 µg/cm^{2}, and thus are significantly thicker than required. The additional areal density does not further increase the charge state yield but leads to increased straggling in angle and energy. DLC (Diamond Like Carbon) foils can be produced down to a thickness of ~0.6 µg/cm^{2} at the Kurchatov Institute in Moscow [22]. Their good performance in timing detectors [16] motivated us to test them in the terminal stripper also.

The foils are mounted on a supporting copper mesh with a spacing of ~10 µm and a transmission of 80 to 90%. During a test on ground potential, the mesh started to glow at a current of 50 nA ^{12}C (11 MeV) and broke after a few minutes at ~ 1µA. This current limitation has to be kept in mind especially for beam tuning. For an AMS measurement, the average current on the foil is small. The stable isobars needed for normalization can be injected with a very low duty cycle, which results in low average beam loads.

We studied this by performing a routine ^{14}C measurement with DLC foil stripping instead of gas stripping. The transmission degraded with a constant slope, different from self supporting foils which break suddenly. Inspection after the measurement revealed that the supporting grid was still intact, and that just the windows at the highest beam intensity were broken. The decline in transmission was accompanied by a reduction of the quality of the acquired data, which we attribute to the resulting inhomogeniety of the foil. However, with the finally chosen DC current load of 1-2 nA and a new foil the measurement precision was the same as for gas stripping, and the transmission was reduced by about 2% within 12 hours.

For ^{36}Cl^{5+} at 3 MV the measured ^{36}Cl^{5+}/^{36}Cl^{-} ratio is about 19% with DLC foil stripping compared to about 6% for gas stripping.

6. Probing the terminal voltage limit of a nominally 3-MV tandem accelerator

Although VERA is nominally a 3-MV tandem, the highest terminal voltage where stable operation is possible maybe significantly higher (the acceptance test required 3.3 MV). To probe this limit, we used our automatic accelerator conditioning program which can work unattended. The condition program ramps up the terminal voltage slowly as long as the spark rate stays below a given limit. After a spark, the terminal voltage is automatically restored, and the measurement can be resumed within less than one minute. From our experience with sparks over the years, venturing for higher terminal voltages seemed possible at an acceptable risk. Within five days and a maximum spark rate of 0.5 h^{-1} the program reached a terminal voltage of 3.7 MV. The accelerator did not de-condition even after the accelerator was operated for several days at lower voltages. Recently, we have decided to start the conditioning program automatically whenever the accelerator is idle.

First measurements were performed at 3.6 MV terminal voltage with DLC foil stripping. For ^{36}Cl, we were able to use the 7+ charge state at 28.9 MeV (instead of the 5+ at 18.1 MeV) with a measured stripping yield of 9%. A repetition of the dilution series measurement was carried out, however not with sufficient statistics to derive an improved detection limit. Also for ^{10}Be, where routine measurements are performed at VERA, an improvement can be achieved: the measured yield for the 3+ charge state is ~ 9%, which is twice as high as for gas stripping at 3.0 MV. For heavy ions the operation at 3.6 MV is expected to result in a significant improvement of the stripping yields.

7. Summary and outlook

From experience with 129I at a tandem operated at 2 MV, Purser et al. suggested a universal 3-MV tandem AMS system with the ability to detect rare actinides at natural levels and below [18]. This is realized with the new analyzer beam line of VERA [14]. We have demonstrated that 3-MV tandems can offset the disadvantages of lower terminal voltage even for the heaviest isotopes as long as stable isobar separation is not required.

Higher energy has significant advantages in cases where stable isobars must be suppressed. However, even under "routine" conditions (3 MV, gas stripping) a separation of ^{36}Cl – ^{36}S is possible with the new method of ΔTOF. The sensitivity limits are still two orders of magnitude worse than achieved at larger accelerators. This is partially caused by the large sulfur count rate. No special chemical treatment was performed to reduce the sulfur content of the materials, and significant improvements can be expected in this context. An optimized design of the ΔTOF is needed to minimize scattering losses. By foil stripping and by pushing the terminal voltage of the accelerator, an energy of 28.9 MeV for ^{36}Cl^{7+} could be reached. At this energy we expect that also with an ionization chamber a separation is possible, which would result in 100% detector efficiency.

Although we have not yet done thorough investigations on the separation of ^{41}Ca – ^{41}K, this case looks even more promising. The stable isobar ^{41}K can be suppressed by using $^{41}CaH_3^-$ or $^{41}CaF_3^-$ ions, so that no large suppression in the detector is required.

Our investigations show that physics allow separation of stable isobars with 3-MV tandems also beyond ^{10}Be – ^{10}B. Our findings have convinced us that 3-MV accelerators will soon venture into fields which are presently the sole domain of much larger machines. Many technical problems still have to be solved, but work is in progress.

We hope that our new findings will be helpful not only for 3-MV tandems to separate isobars around mass ~40, but also for larger machines at even higher masses. Nuclides which are both heavy and require stable isobar suppression have not been measured at the expected natural concentrations so far (e.g ^{182}Hf – ^{182}W [23], ^{205}Pb – ^{205}Tl).

8. References

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Figure Captions

Figure 1. Schematic Layout of the VERA AMS system.

Figure 2. Calculated maximum achievable energy *E* with a tandem accelerator of terminal voltage *U* for ^{10}Be and ^{36}Cl, for both gas and carbon foil stripping. The highest charge state which gives as least 5% yield following Sayers's formula [11] is used. The steps in the plots appear when the next higher charge state is selected.

Figure 3. Calculated separation of ^{36}Cl – ^{36}S with an initial energy of 18 MeV in a silicon nitride foil. The double lines in (a) indicate the energy straggling (1 σ). For the definition of the separation $S_{\mathrm{A,B}}$ see text. The small circle in (b) is a value measured at VERA.

Figure 4. Calculated separation of ^{36}Cl – ^{36}S achievable for different terminal voltage with silicon nitride foils. The optimum foil thickness is selected for each energy.

Figure 5. Measured TOF-spectra of a standard with ^{36}Cl/Cl = 10^{-10} and a blank after ~530 μg/cm^2 of silicon nitride. Lower channel numbers correspond to longer TOF and thus lower energy. The shaded area is the integration region rejecting half of all ^{36}Cl events, but resulting in a background suppression of 10^3.

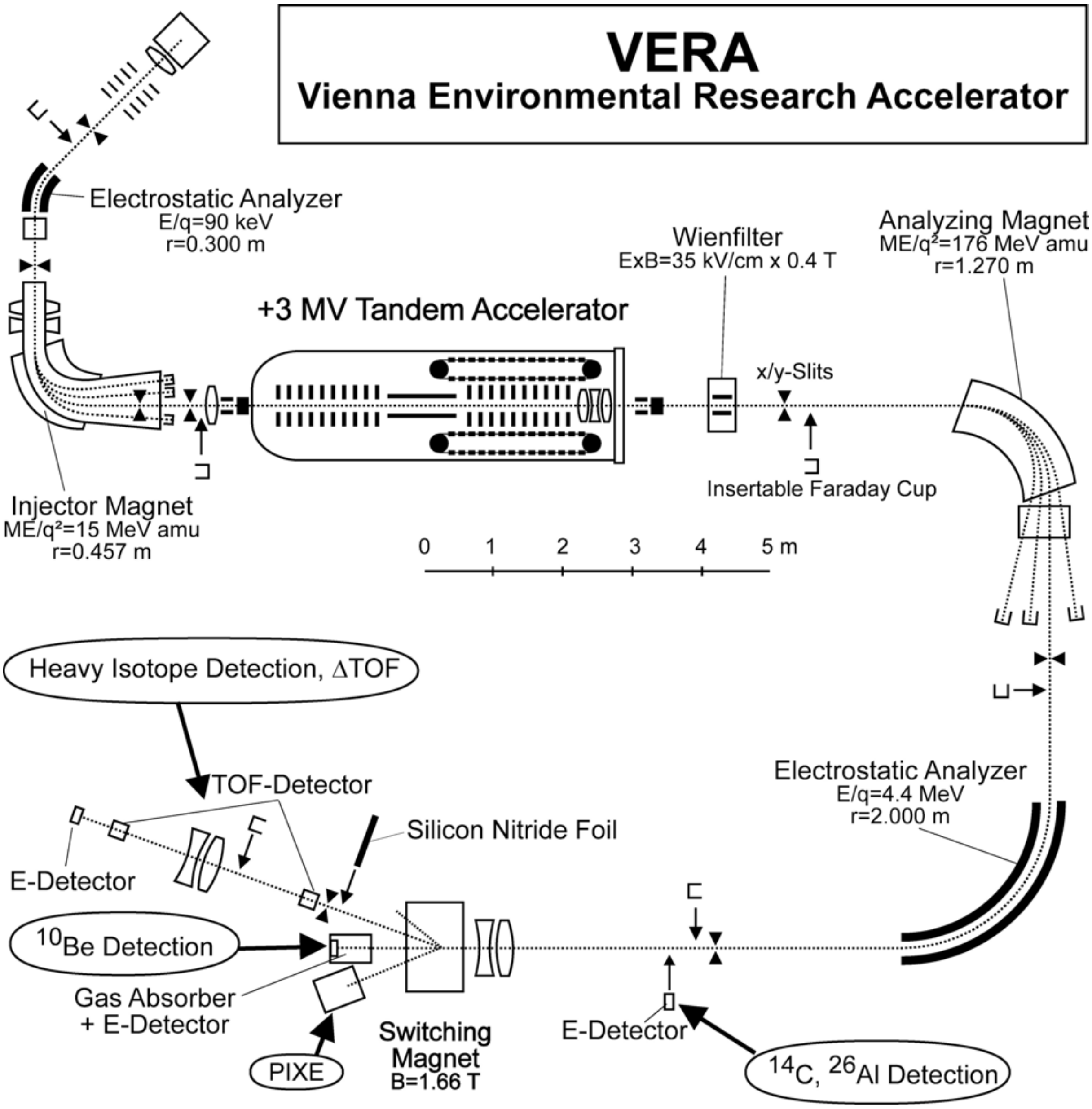
VERA
Vienna Environmental Research Accelerator
Electrostatic Analyzer
E/q=90 keV
r=0.300 m
Wienfilter
ExB=35 kV/cm x 0.4 T
Analyzing Magnet
ME/q²=176 MeV amu
r=1.270 m
+3 MV Tandem Accelerator
x/y-Slits
Insertable Faraday Cup
Injector Magnet
ME/q²=15 MeV amu
r=0.457 m
0 1 2 3 4 5 m
Heavy Isotope Detection, ΔTOF
TOF-Detector
Silicon Nitride Foil
E-Detector
Electrostatic Analyzer
E/q=4.4 MeV
r=2.000 m
10Be Detection
Gas Absorber
+ E-Detector
PIXE
Switching
Magnet
B=1.66 T
E-Detector
14C, 26Al Detection

Figure 1

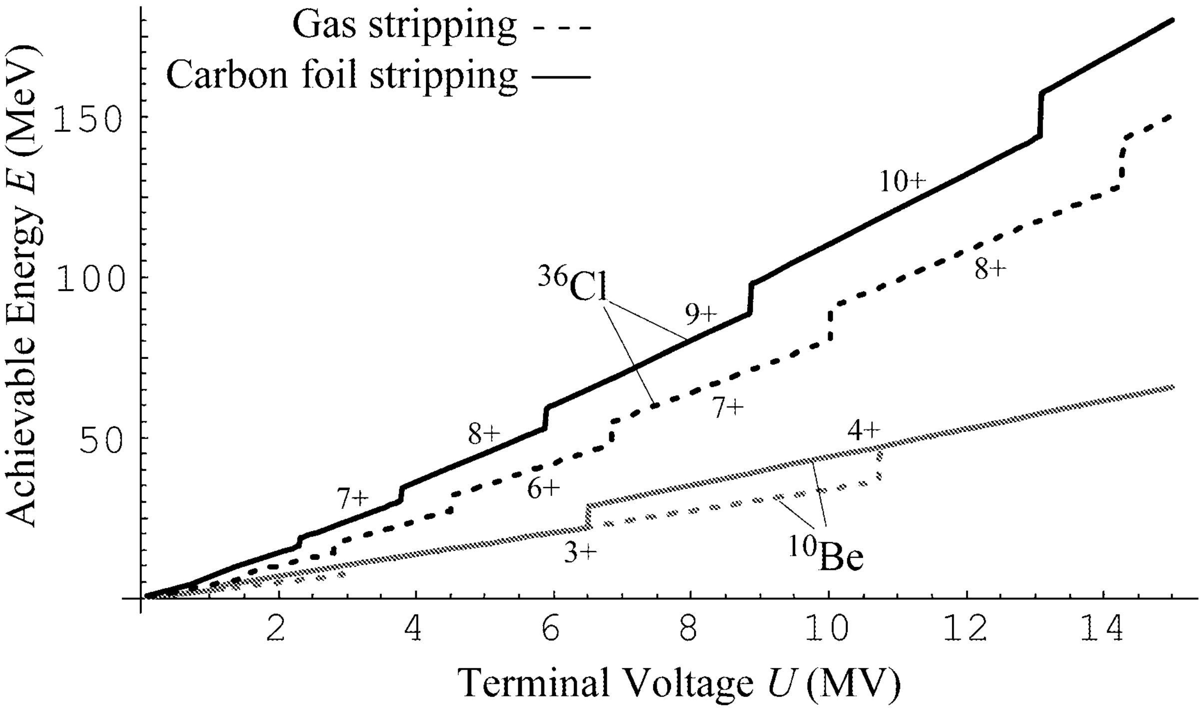
Gas stripping - - -
Carbon foil stripping ——
Achievable Energy E (MeV)
Terminal Voltage U (MV)
50
100
150
2
4
6
8
10
12
14
^{36}Cl
^{10}Be
7+
8+
9+
10+
6+
7+
8+
3+
4+

Figure 2

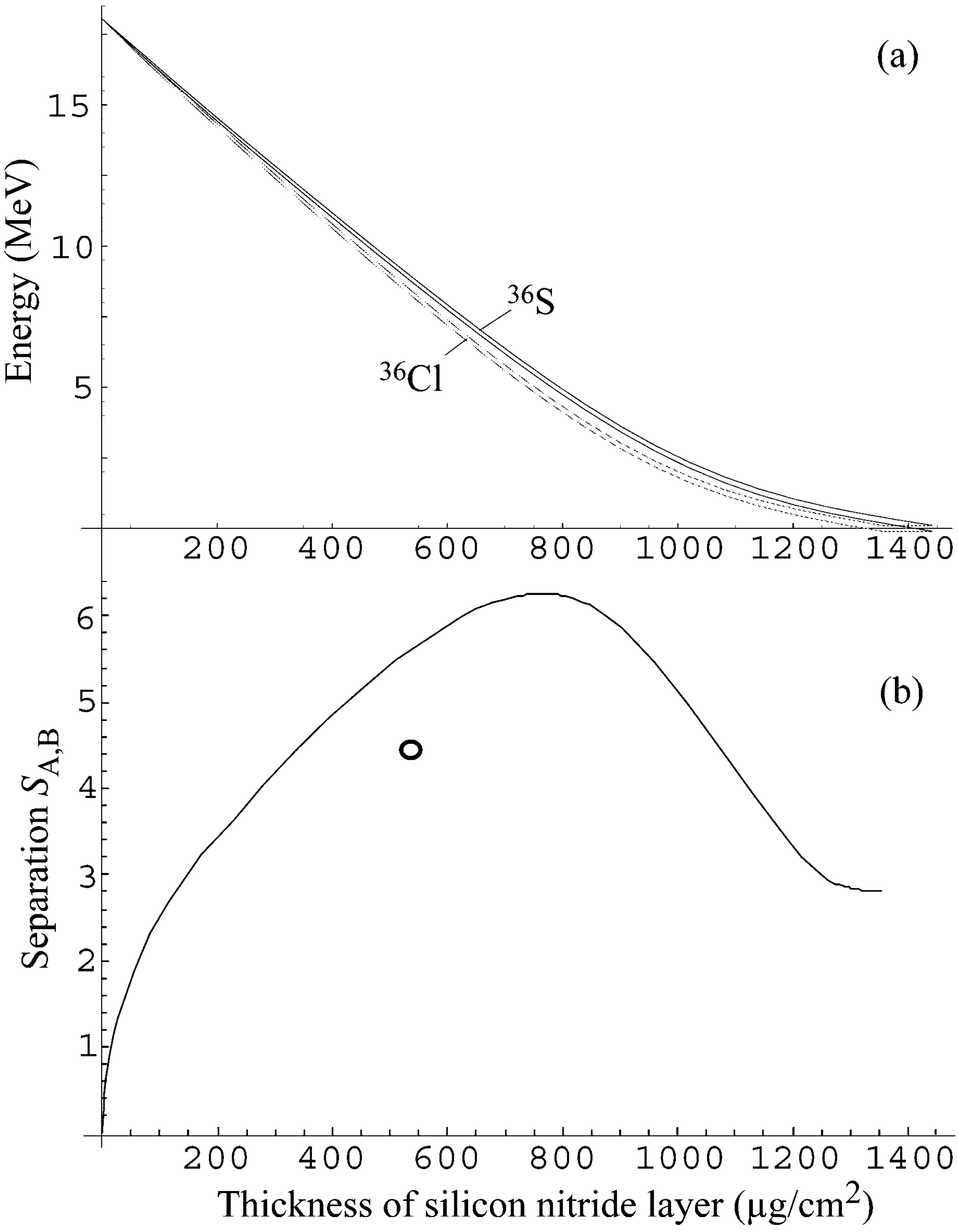
(a)
Energy (MeV)
15
10
5
^{36}S
^{36}Cl
200
400
600
800
1000
1200
1400
(b)
Separation $S_{A,B}$
6
5
4
3
2
1
200
400
600
800
1000
1200
1400
Thickness of silicon nitride layer (μg/cm^2)

Figure 3

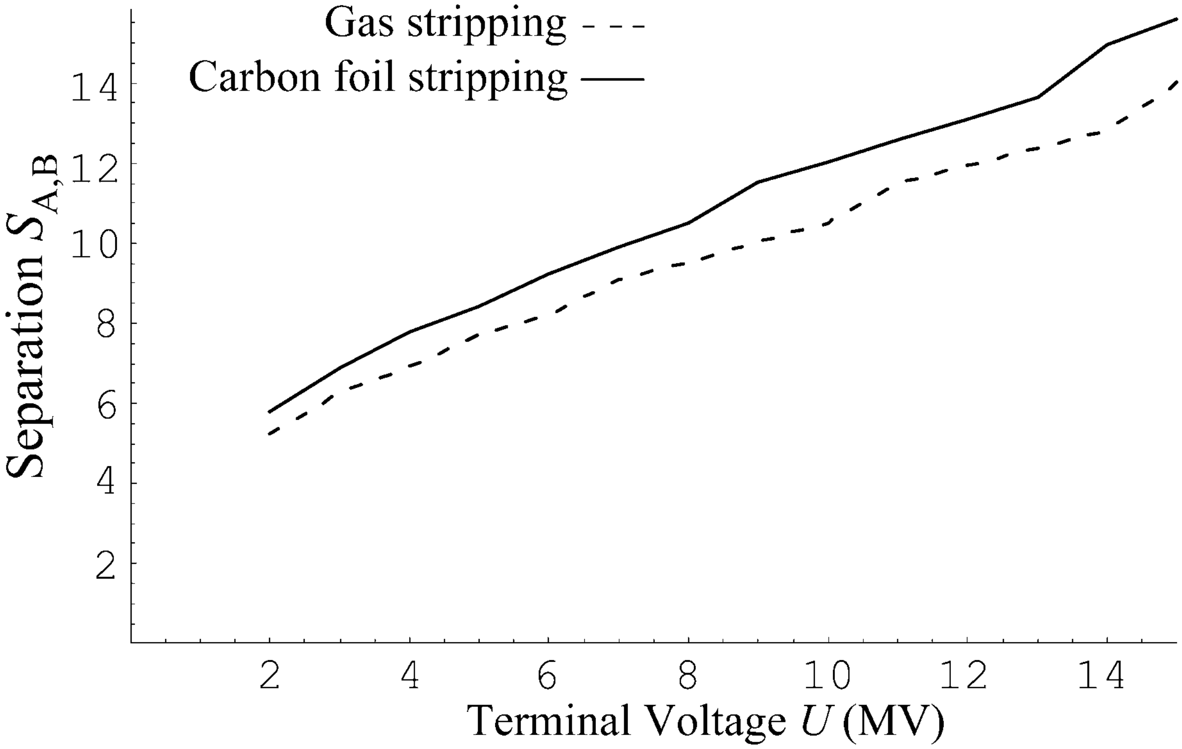
Gas stripping - - -
Carbon foil stripping ——
Separation $S_{A,B}$
2
4
6
8
10
12
14
Terminal Voltage U (MV)
2
4
6
8
10
12
14

Figure 4

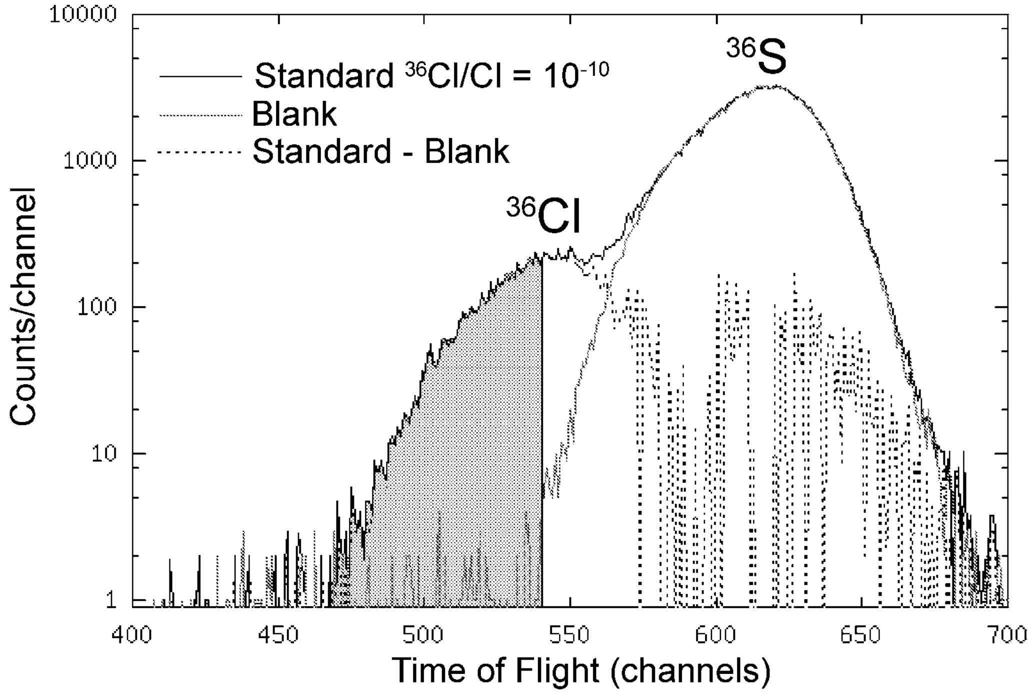
Standard ^{36}Cl/Cl = 10^{-10}
Blank
Standard - Blank
^{36}S
^{36}Cl
Counts/channel
Time of Flight (channels)
10000
1000
100
10
1
400
450
500
550
600
650
700

Figure 5