Development of the Lund AMS System and the Evaluation of a New AMS Detection Technique

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Abstract

This thesis is based on the work performed at the Lund Pelletron accelerator facility in order to improve the accuracy and efficiency of the Lund Accelerator Mass Spectrometry (AMS) system. AMS is a highly sensitive atom counting technique, suitable for measuring the specific activity of cosmogenic radionuclides in a sample. In AMS, the number of rare radionuclides that are transmitted through the system is directly counted by a detector system, whereas the current of an abundant isotope is directly measured by a Faraday cup positioned immediately in front of the detector system, yielding the relative abundance of the isotopes in the sample in the ion source. To obtain a high accuracy, all measurements are performed relative to a standard of known activity. Charge state distributions have been obtained for a number of isotopes, ^{9}Be, ^{12}C, ^{13}C, ^{16}O, ^{19}F, ^{27}Al, ^{35}Cl, ^{48}Ti and ^{58}Ni, in order to improve the transmission through the system and to reduce the isotopic fractionation in the measurements. For carbon, charge state distributions were obtained both under foil- and gas stripping conditions. The pressure profile of the Lund Pelletron system has been calculated, both under foil- and gas stripping conditions, to be able to perform transmission calculations for a carbon beam. These results were used to design a new terminal stripper of the accelerator system. A new ion source has, during the last few years, been constructed providing a multiple sample wheel, enabling more accurate relative measurements and also provides, due to a higher beam current, more efficient measurements.

A new detection technique, suitable for AMS measurements on heavier radionuclides, such as ^{36}Cl, ^{44}Ti and ^{59}Ni, has been evaluated and detection limits for ^{59}Ni have been derived.

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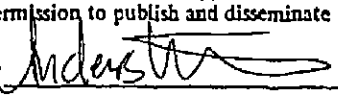
Signature  Date September 5, 1995

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To Pernilla and Julia

"A small step for mankind,
a giant leap for Anders"

Preface

This thesis is based on the following papers:

- The charge state distribution of carbon beams measured at the Lund Pelletron accelerator, presented at the Third European Conference on Applied Research and Technology (ECAART) held in Orleans (France) 31/8-4/9-93 and published in the proceedings Nucl. Instr. and Meth. B 89 (1994) 259

- The pressure profile in the Lund Pelletron accelerator, accepted for publication in Nucl. Instr. and Meth. A and is currently in press

- The transmission of a carbon beam through a tandem accelerator, accepted for publication in Nucl. Instr. and Meth. A and is currently in press

- A negative caesium-sputtering ion source for AMS investigations, will be presented at the 7th International Conference on Heavy Ion Accelerator Technology held in Canberra (Australia) 18/9-22/9-95. The proceedings will be published in Nucl. Instr. and Meth. A

- The charge state distribution of Be, O, F, Al, Cl, Ti and Ni ions, submitted for publication in Nucl. Instr. and Meth. B

- Isobar suppression in accelerator mass spectrometry by the detection of the characteristic X-rays, presented at the VII International Conference on Particle Induced X-ray Emission and its Analytical Applications. The paper is accepted for publication in the proceedings that will be published in Nucl. Instr. and Meth. B

Contents

1. Introduction

Of the more than 2300 nuclides known to man, only 265 are stable (Lederer and Shirley, 1978). The others are changing with time due to radioactive disintegration. Some of these radionuclides have existed ever since the creation of the Earth, others are constantly being produced in the atmosphere by cosmic rays but most of these radionuclides have been studied only after having been artificially produced by man (Elmore and Phillips, 1987).

In many fields of science it is important to measure low concentrations of radionuclides. Some of these fields include archaeology, art, biomedicine, climatology, cosmic-ray physics, environmental physics, glaciology, meteorites and oceanography (Kutschera, 1990).

The specific activity of a sample, i.e. the number of disintegrations per time unit, can be measured in two different ways; either by measuring the radiation emitted in the radioactive decay (decay counting) or by directly counting the number of radionuclides in the sample (atom counting). If the half-life of the radioisotope is known both of these measurements will yield the same result (Kutschera, 1993). Depending on a number of circumstances, either one of these techniques may be advantageous. If the half-life of the radioisotope is long and/or the sample size is small, atom counting is often a more efficient way to determine the specific activity in a sample. In some cases it is impossible to perform ordinary radiation detection of a specific disintegration due to a high background of other radionuclides in the sample.

Accelerator mass spectrometry (AMS), which is a highly sensitive atom counting technique, was introduced at several laboratories, independently of each other, in 1977 (Muller, 1977, Muller et al., 1977, Schwarzschild et al., 1977, Nelson et al., 1977, Purser et al., 1977 and Bennet et al., 1977). As can be guessed by its name, AMS is an extension of conventional mass spectrometry (MS). In MS an ion beam is extracted from the sample and the ions are accelerated to keV energies and are mass analysed by a dipole magnet. Interferences from abundant neighbouring isotopes and atomic- and molecular isobars are a constraint on the detection limit. In AMS, where the ions are accelerated to MeV energies using an electrostatic tandem accelerator, these interferences can often be suppressed by the use of several filters, the use of a nuclear particle detection system and also due to the fact that molecular isobars with charge state 3+ or higher are dissociated in the stripping process. AMS can yield a million-fold increase in sensitivity for isotope concentration measurements ($10^{-6} \rightarrow 10^{-12}$) compared to conventional

MS (Kutschera, 1993). See Fig. 1 for a schematic comparison between MS and AMS.

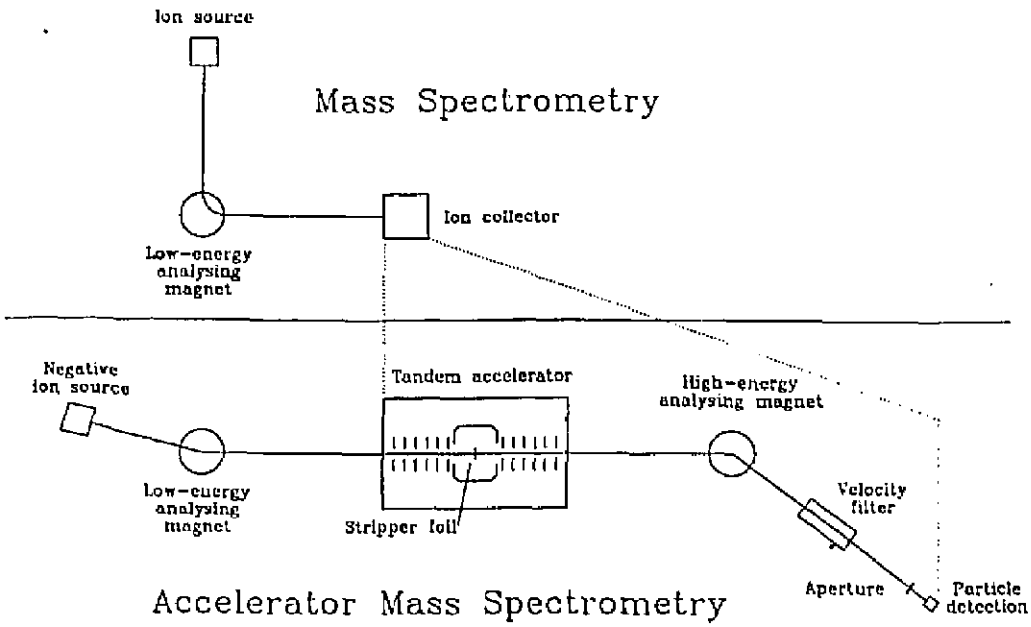

Fig. 1. A schematic comparison between accelerator mass spectrometry (AMS) and conventional mass spectrometry (MS)

After more than 15 years of development, AMS has proven to be a highly versatile tool for research, with a detection limit sufficient for detecting natural abundancies of long-lived cosmogenic radioisotopes, such as ^{10}Be, ^{14}C, ^{26}Al, ^{32}Si, ^{36}Cl and ^{41}Ca. None of these isotopes has been directly observed in concentrations occurring in nature by ordinary MS (Wölfli, 1984).

So far, ^{14}C has received most attention due to its well known biological history and also due to its other favourable properties compared to other radionuclides, see section 2.2. Radiocarbon datings have now been performed for more than ten years, using the AMS technique. To perform a ^{14}C-dating measurement only a few milligrams of carbon are needed, which is approximately one thousandth of the amount of carbon needed to measure a contemporary sample using a β-scintillation detector (Gillespie, 1984). This does not only make the AMS measurements virtually non-destructive but also opens new fields in which dating measurements can be performed, for

instance, a seed extracted from a piece of ancient pottery can be sufficient to date the object, paintings can be dated using only a tiny piece of canvas and so on.

In AMS, the whole accelerator system must be highly stable and have a high precision in order to produce accurate results and should also provide a high efficiency, which is important when working on small samples. To achieve this, the accelerator system must, among many other parameters, provide high beam current and high beam quality, good voltage stability and accurate reproducibility. Furthermore, the detection system should have high sensitivity and high discrimination for isobar separation.

The objectives of the work described in this thesis are:

(i) To optimise the AMS set-up at the Lund Pelletron accelerator in order to be able to perform more accurate and more efficient measurements.

(ii) To examine the possibility to perform AMS measurements on heavier radionuclides, such as ^{59}Ni, using small tandem accelerators.

2. Long-lived radioisotopes

2.1 Production

The radioisotopes that exist in nature can be divided in two categories; natural and artificial, where the natural radioisotopes either have existed since the creation of the Earth (e.g. ^{235}U and ^{238}U) or are constantly being produced by the interaction of the cosmic-ray flux with the Earth. The artificial radioisotopes are nuclei that have been produced by human activities.

The isotopes called cosmogenic radioisotopes are produced, directly or indirectly, by the cosmic-ray flux to which the Earth is exposed. The atmosphere surrounding the Earth acts as an effective shield against cosmic-rays and, hence, the production of cosmogenic radioisotopes will vary with the cosmic-ray flux and therefore with the altitude above sea level (Kutschera, 1990). The main components of the atmosphere are nitrogen (78%) and oxygen (21%). The only long-lived radioisotopes that are formed from these two elements, are (excluding ^{3}H which is only moderately long-lived) ^{10}Be and ^{14}C (Raisbeck and Yiou, 1984). The next most abundant component in the atmosphere is Ar (0.9%) from which four long-lived radioisotopes are produced (^{26}Al, ^{32}Si, ^{36}Cl and ^{39}Ar) (Raisbeck and Yiou, 1984). Heavier elements in the atmosphere are extremely rare. Some production of heavier, long-lived radionuclides, such as ^{41}Ca, takes place at sea level even though the cosmic-ray flux is attenuated by a factor of ~1000 (Raisbeck and Yiou, 1984).

Some of these long-lived radionuclides, and others, are also produced by human activities. In addition to the production of moderately long-lived radionuclides, especially ^{85}Kr, ^{90}Sr and ^{137}Cs, and the production of very long-lived radionuclides, such as ^{239}Pu and ^{129}I, that takes place in the fuel in nuclear power plants, also ^{14}C is produced through various nuclear reactions in the fuel, the moderator and the coolant, and in structural materials (Davies, 1979). In the 1950's and 60's, nuclear bomb tests in the atmosphere caused the production and release into the atmosphere of large quantities of ^{14}C in addition to radioactive fallout products due to the fission process. Some of these radionuclides are also produced industrially for various applications in medicine, science and industry.

2.2 Applications

As previously mentioned, the AMS technique is especially suited for measuring concentrations of long-lived radioisotopes in a sample. The reason for this is that during a normal measuring time, say one hour, many more of the long-lived radioisotopes in a sample survive than decay, i.e. when the half-life of the radioisotope is much longer than the measuring time, atom counting is more efficient than decay counting. The radioisotopes that have been studied in AMS experiments by 1990 is indicated in Fig. 2 (Kutschera, 1990). In addition to these isotopes, AMS has also been demonstrated for: ^{3}H, ^{32}Si, ^{59}Ni and ^{60}Fe (Paul, 1993 and Elmore and Phillips, 1987).

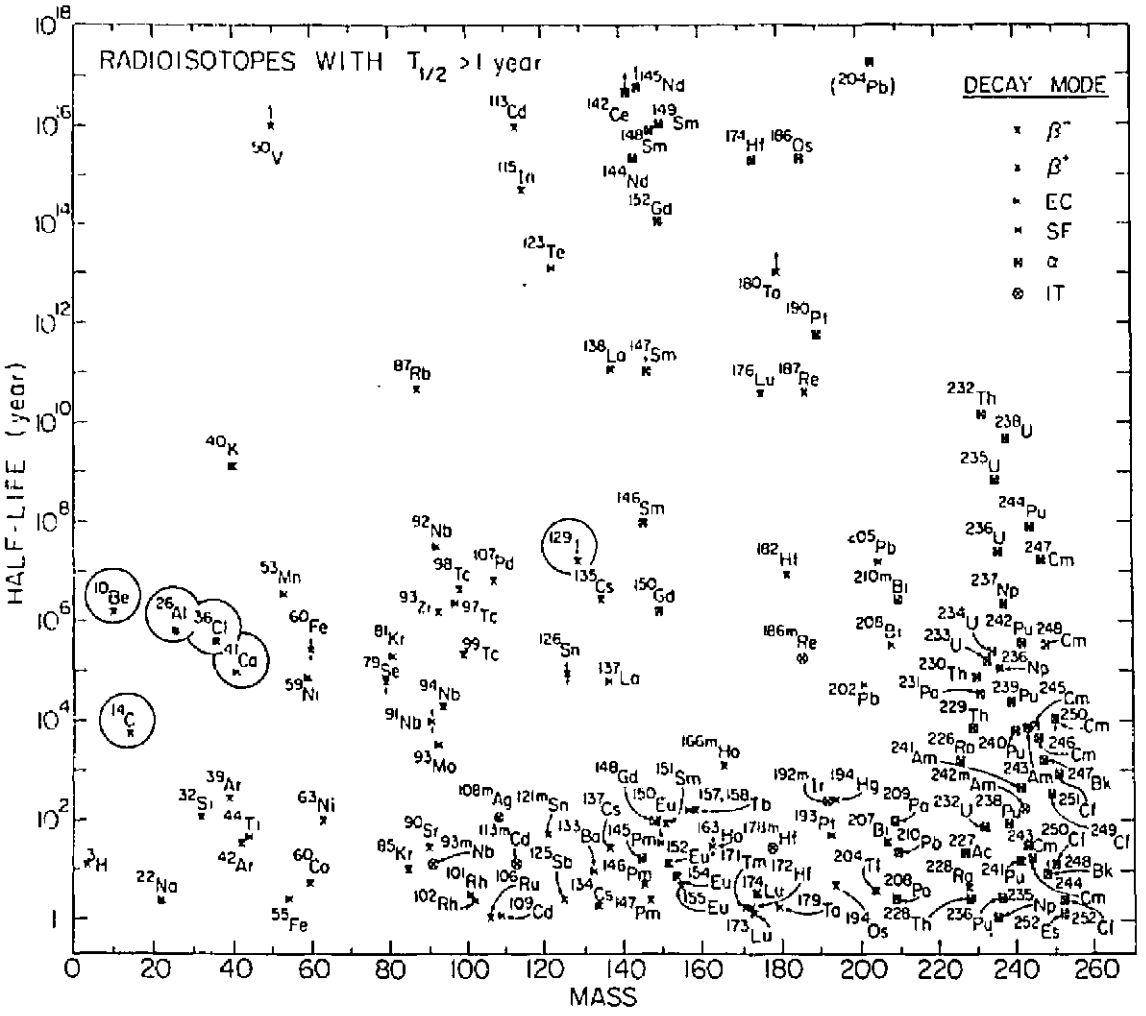

Fig. 2 Radioisotopes with half-lives longer than one year. The six isotopes used in AMS studies by 1990 are indicated by circles (After Kutschera, 1990).

Since the invention of AMS in 1977, by far the most measurements have been performed on ^{14}C. There are a number of reasons for this. First of all, ordinary β-decay measurements on ^{14}C have been performed since the early fifties and the technique has over the decades been developed into a precise and widely used dating tool. ^{14}C has a useful half-life ($t_{½}$=5730 years) for dating back to about 50 000 years and has been calibrated with tree rings back to about 9000 years BC (Long et al., 1993). The distribution of ^{14}C over the surface of the Earth is very uniform, giving a well-defined $^{14}C/^{12}C$ ratio (Elmore and Phillips, 1987). ^{14}C is very important from a biological point of view as carbon is an essential element in all living matter. Moreover, ^{14}C is especially suited for AMS measurements with a tandem accelerator, due to the fact that there exists no stable, negatively charged atomic isobar, thus simplifying the detection. There are a numerous of applications where the ^{14}C activity of a sample is studied. A short and incomplete list of these fields include: archaeology, quaternary geology, oceanography, art and biomedicine (Kutschera, 1993).

Of the other cosmogenical, long-lived radioisotopes, ^{10}Be and ^{26}Al have received much attention. ^{10}Be has a very long half-life ($t_{½}$=1.6×10^{6} years) and has been used as a sensitive monitor of the cosmic-ray flux and can therefore be used as a climate monitor ranging several 100 000's years back in time (Sumayalulu, 1977). For ^{26}Al, ($t_{½}$=7.2×10^{5} years) the potential application is geoscience and cosmology but it has also been used as a tracer element to study the correlation between aluminum intake and Alzheimers disease (Barker et al., 1990).

^{36}Cl is an ideal isotope for dating and tracing old ground water (Elmore and Phillips, 1987). It has for this application a suitable half-life of 3×10^{5} years, simple geochemistry, a high solubility in water and a very low sticking probability to solid surfaces (Kutschera, 1993).

^{59}Ni is a long-lived ($t_{½}$=7.6×10^{4} years) radioisotope that is produced in extraterrestrial matter by the interaction of cosmic rays (Paul et al., 1993) but is also produced in shielding material in nuclear power plants by neutron activation (Wagner et al., 1994 and Artigalas et al., 1993). For the final disposal of nuclear waste, activity measurements must be performed (SKB Report, 1993). Because of its long half-life and since ^{59}Ni decays only via electron capture (EC) AMS can be a suitable technique to perform these measurements.

3. The AMS system

3.1 General

An outline of the AMS system at the Lund University is shown in Fig. 3. It consists of a NEC Pelletron tandem accelerator with a maximum terminal voltage of 3 MV. On the low-energy side of the accelerator an ion source, attached to a 15° dipole magnet, creates the beam that is injected into the accelerator. The high-energy side of the accelerator consists of a dipole magnet, where the 36° exit tube is used when performing AMS investigations, a velocity filter and a particle detection system. The number of radioisotopes transmitted through the system is counted by the detector system, whereas the current of an abundant isotope is directly measured by a Faraday cup positioned just in front of the detector, thus registering the relative concentration of the isotopes in the sample in the ion source. By normalising the results obtained to the result obtained for a standard of known activity, the specific activity of the sample in the ion source can be deduced. The negative ion beam, which is required by the tandem accelerator, is created in the ion source by sputtering solid samples by a Cs^+ beam. Depending on the electron affinity, either elementary ions or molecular ions are extracted. The ion beam is mass selected by the first dipole magnet and focused onto an aperture before being injected into the accelerator. The negative ion beam is accelerated to high velocities by the potential gap between ground and the positively-charged terminal of the accelerator. In the terminal the ion beam traverses a stripping medium, by which a number of electrons are stripped from the ions, yielding an ion beam of multiply-charged, positive ions. The beam is then further accelerated down to ground potential. The energy of the ions, in MeV, emerging from the accelerator is given by $E=T_a \cdot e(1+q)+T_{is} \cdot e$, where T_a is the terminal voltage in MV, T_{is} is the extraction potential of the ion source in MV, e is the elementary charge and q the charge state of the positive ion. On the high-energy side of the accelerator the ion beam is focused onto a pair of vertical slits behind the 36° dipole magnet by which the beam is momentum- and charge state analysed. On the experimental beam line the beam is velocity-analysed by the use of a Wien filter and focused onto an aperture just in front of the Faraday cup and detector system. The ratio between the currents of the radioisotope and the abundant isotope can be measured reproducibly by changing the settings of the AMS system.

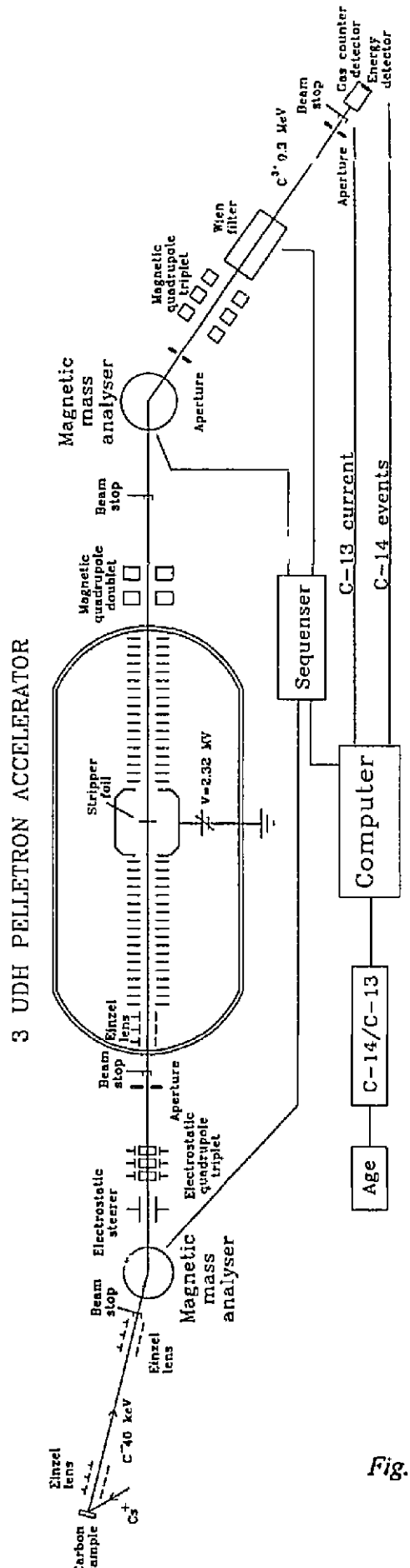

Fig. 3. A schematic diagram of the Lund AMS system

3.2. Background rejection in AMS

The background in the final detection will determine the lowest concentration of radioisotopes detectable in the sample in the ion source. The background is either due to contaminations in the sample to be analysed or to the instrumental background. The instrumental background may either be due to stable isotopes of the radioisotope of interest, often abundant in the sample, that managed to be transmitted through the accelerator system, isobars of the radioisotope or any other detected ion giving rise to a signal that is misinterpreted as a signal from a detected radioisotope. The total background of the system can be determined by performing a measurement of a blank sample (practically free of the radioisotope of interest) that has undergone the same sample preparation as the samples to be analysed.

In order to reduce the instrumental background of the system several filters are included in the AMS system, as described above. Three different kinds of filters exists, which use static electric and magnetic fields (Litherland, 1980). These are shown in Table 1. Normally a combination of two or three of these types of filters is used.

Type of analysis	Quantity selected
Magnetic field	Mass×energy/charge^2
Electrostatic field	Energy/charge
Velocity analyser	Energy/mass

Table 1. Three types of analytical steps used in AMS.

3.3 Isotope separation

3.3.1 Low-energy side mass separation

In the recently-installed, new ion source in the Lund AMS facility, the negative ion beam is created, by bombarding the sample with a 25 keV Cs^+ beam. In the sputtering process in the ion source, negative ions are created which are extracted from the sample by a 20 kV potential gap and further accelerated in a 20 kV potential gap, yielding a 40 keV ion beam. However, in the sputtering process, kinetic energy can be transferred from the Cs ions to the sputtered ions of the sample, yielding a non-monoenergetic ion beam (Litherland, 1984). These high-energy tails can be troublesome, especially if

heavier elements are analysed. Low-energetic tails are created by unstable molecules that dissociate in the extraction field, yielding an energy continuum (Suter, 1990). Electrical optical elements focus the ion beam onto a pair of vertical slits beyond the 15° dipole magnet. As previously mentioned, the inflection magnet selects E/q×M/q, but since both the energy and the charge state are the same for most ions issuing from the source, the magnet acts only as a mass analyser.

3.3.2 High-energy side mass separation

One of the problems associated with MS measurements is interferences from molecular isobars. In the case of ^{14}C, the molecular isobars present in the beam are not only $^{12}CH_2$ and ^{13}CH but also $^{7}Li_2$. The use of a tandem accelerator in AMS provides the possibility, to reduce these interferences to a very high degree. At the terminal of the tandem accelerator the ions are charge-exchanged into positive ions in the stripper medium. All molecules with charge state 3+ or higher dissociate rapidly and have not been observed emerging from a tandem accelerator (Litherland, 1980). This feature of tandem accelerators eliminates the need for very high resolution mass spectrometers to resolve molecular isobars, if charge state 3+ or higher is selected by the analysing magnet. However, fragments from dissociated molecules might still be present in the momentum-analysed beam if the ions have, for instance, undergone charge exchange processes in the accelerator tubes or scattering processes. It is therefore crucial to remove these tails. This can be done either by using an electrostatic analyser or, as in the case of the AMS system in Lund, by using a velocity analyser. The velocity analyser applies an electric field at right angles to the beam and magnetic field orthogonal to both the beam and the electric field. The fields give rise to two opposite forces, $\vec{F}_E$ and $\vec{F}_B$. For the selected ions the forces are balanced, $\vec{F}_E = \vec{F}_B$ or $qE = qvB$, and the ions will pass straight through, while the unwanted ions will be bent off. The advantage of a velocity analyser, or Wien filter, compared to an electrostatic analyser, is that the Wien filter can be positioned in a straight section of the beamline.

3.4 Isobar separation

3.4.1 General

Isobars, i.e. particles having the same mass as the radioisotope of interest, are either atomic or molecular. As previously mentioned, molecular isobars can be greatly suppressed by the use of a tandem accelerator. In order to suppress atomic isobars the first suppression can in many cases be performed in the sample preparation, by chemical separation. A practical limit of the relative concentration of impurities present in the samples is 10^{-6} to 10^{-8} (Elmore and Phillips, 1987). It is often unnecessary to further improve the reduction of isobars due to significant contributions from contaminations at this level in the ion source. Additional isobar separation is necessary to reach the 10^{-12} to 10^{-15} level.

3.4.2 Isobar separation on the low-energy side

In some cases, the use of a tandem accelerator can also reduce the intensity of atomic isobars, due to the fact that the tandem accelerator requires negative ions to be injected. In the case of ^{14}C, the atomic isobar ^{14}N does not form stable negative ions. In fact, this was one of the reasons, together with the dissociation of the isobaric molecules in the stripping process, that the potential of AMS, using a tandem accelerator, was first realised (Purser et al., 1977). The atomic isobars, ^{26}Mg, ^{36}Ar and ^{129}Xe, are also greatly suppressed compared to the long-lived radioisotopes ^{26}Al, ^{36}Cl and ^{129}I (Suter, 1990). Sometimes the use of a negative molecular beam can suppress the isobaric background, as in the case of $^{41}CaH_3^-$ where the K background is reduced by several orders of magnitude, compared to extracting negative atomic beams (Raisbeck et al., 1981).

3.4.3 Isobar separation on the high-energy side

One of the great advantages of AMS compared to ordinary MS is that the use of a tandem accelerator yields much higher beam energies. In some cases this provides the possibility to perform isobar separation by analysing fully stripped ions, thereby removing isobars with a lower atomic number from the beam. For example, the isobars ^{7}Li and ^{26}Mg can be removed when measurements on ^{7}Be and ^{26}Al, respectively, are performed. However,

complete stripping of heavier elements, such as Al, requires very large accelerators.

Another advantage in accelerating ions to high energies is the possibility to use nuclear particle detection devises for particle identification and, hence, for isobar separation. A detection system extensively used in AMS measurements of lighter ions, such as Be and C, is the ΔE-E gas counter telescope. The basic principle of this kind of detector is that the energy loss (dE/dx) of an ion in matter is proportional to the square of the nuclear charge of the ion (Ziegler et al., 1985). There are some varieties of this type of detector arrangement, but in principle most detectors are similar to the one used at the AMS facility in Lund, shown in Fig. 4.

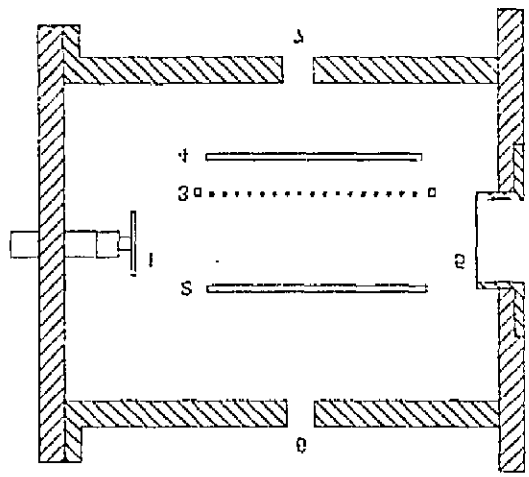

Fig. 4. Schematic sketch of the ΔE-E gas counter telescope used in Lund. The numbers correspond to: energy detector (1), cathode (2), grid (3), anode (4), entrance window (5), gas inlet (6) and gas outlet (7).

It consists of a gas cell, filled with gas (90% CH_4 + 10% Ar) to a pressure of ~100 mbar, and a solid state Si detector. The energy loss of the incoming projectile is registered by the ionisation detector, and the residual energy is measured by the Si detector. A typical, three-dimensional representation of ^{14}C data measured with the ΔE-E gas counter telescope is shown in Fig. 5.

The dE/dx technique can in some cases be employed successfully for isobar separation by simply using a passive absorber, a gas cell or a foil, in combination with a Si detector which registers the residual energy of the ions. For light elements for which the unwanted isobar has a higher nuclear charge than the radioisotope, passive absorbers can be used to completely stop the isobar and let the radioisotope pass through the absorber. This has

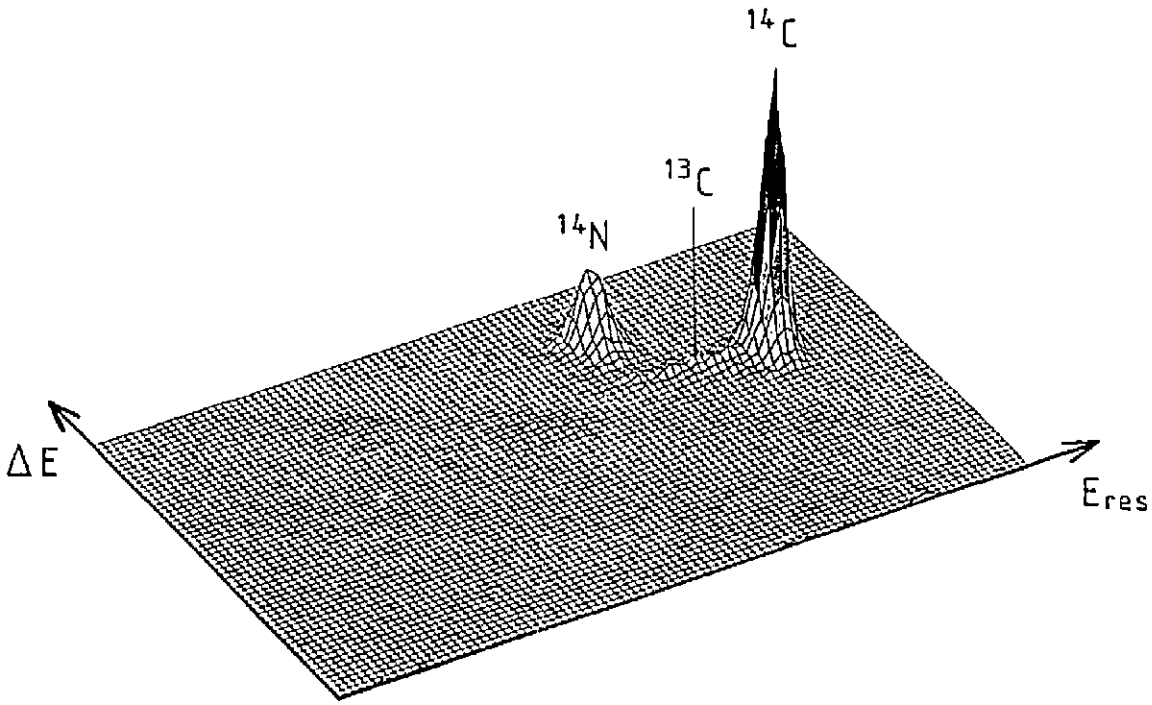

Fig. 5. Three-dimension pulse-height distribution obtained with an activated ^{14}C sample in the ion source, with the inflection magnet tuned towards mass 15 in order to obtain a ^{14}N peak. (^{14}N is injected as $^{14}NH^-$.)

the advantage that counting rates in the detector remain low. Another way of utilising the dE/dx technique is by using a gas cell sufficiently thick to completely stop of the incoming ions and by measuring the successive energy loss with an array of ionisation detectors in the same gas volume (Paul et al., 1993). Gas filled magnets in combination with time-of-flight detection systems have also been successively used when performing AMS measurements. These systems, however, require high beam energies for best operation (Suter, 1990). In gas filled magnets the different isobars will be deflected differently due to the fact that the average charge state of an ion beam will differ depending on the nuclear charge of the ions. The gas filled magnet detector offers little suppression of the isotopic background and, hence, it is often used in combination with a time-of-flight detector which is used to distinguish ions with different mass numbers (Paul, 1993).

The limited beam energy available at small AMS facilities, such as the Lund Pelletron accelerator, is not sufficient for isobar separation of heavier radionuclides, such as ^{41}Ca, and ^{59}Ni, using conventional detection techniques. However, a new detection technique has recently been presented

where the different isobars in the beam are distinguished by their characteristic X-rays that are produced when the beam is stopped by a target (Artigalas et al., 1993). This detection technique greatly expands the region of radioisotopes that can be measured at smaller AMS facilities. The detection efficiency is, however, relatively low when smaller tandem accelerators are used and approximately one of every thousand incoming projectiles is registered (Artigalas et al., 1994).

4. Measuring procedure

For some years, the AMS system at the Lund Pelletron accelerator facility has been undergoing development and ^{14}C measurements are now being performed (Skog et al., 1992). In order to produce a stable beam current, the material to be analysed must be converted to elementary carbon. This is done in the graphitization system (Stenström et al., 1994). The carbon obtained from the graphitization system is mixed with iron or copper powder (to increase the thermal conductivity, which in turn increases the ion beam current obtained from the sample (Suter, 1990)) and pressed into a copper probe that is placed in the ion source.

Accurate AMS measurements can only be performed by doing relative measurements (Kutschera, 1993). Therefore all measurements are normalised to the value measured on a standard with a known activity (NBS oxalic acid standard (Stuvier and Polach, 1977)). To obtain the instrumental background and the background of the sample preparation system, measurements are always performed on a ^{14}C-free anthracite sample (old charcoal), prepared in the same way as the samples to be analysed.

In order to exclude interferences from the molecular isobars $^{12}CH_2$ and ^{13}CH, charge state 3+ is analysed by the high-energy side dipole magnet and to minimise the isotopic fractionation between $^{13}C^{3+}$ and $^{14}C^{3+}$, the ions are stripped at a terminal voltage of 2.44 MV, (see section 6.1). To minimise the effects of drift in the AMS system, the settings of the AMS are alternated several times during a measurement between mass 13 and mass 14. To correct for possible long-time drifts in the AMS system, the standard and the background sample are analysed several times during a series of measurements.

A pulsing electrostatic deflector has quite recently been installed on the low-energy side of the accelerator giving the possibility to inject ^{12}C into the system without loading the high-voltage terminal of the machine. ^{12}C is injected in 0.1 ms long pulses with a frequency of 100 Hz. By also measuring the ^{13}C to ^{12}C ratio the isotopic fractionation, either natural in the sample or instrumental, can be deduced. Up to now, $\delta^{13}C$ of the samples have been measured using conventional MS.

Computer control of the dipole magnets and the Wien filter has been installed, enabling accurate and efficient alternation between the different settings of the system during each measuring cycle.

5. Accuracy and efficiency

5.1 General

The overall efficiency of an AMS system is defined as the ratio of the number of detected (radioactive) atoms to the number of sputtered (radioactive) atoms from the sample. The efficiency is equal to the product of the efficiency of the formation of negative ions in the ion source, the stripping yield of the particular charge state selected, the transmission through the accelerator system and analysing equipment and the efficiency of the detector system. A high total efficiency is necessary in order to achieve a low statistical uncertainty when only very small samples are used. To perform acceptable archaeological datings, the accuracy of the measurements must fall within ±1% to compete with classical radiocarbon datings (Wölfli, 1984). In order to be able to perform highly sensitive, accurate and efficient measurements the accelerator system must provide, a high beam current and high beam quality, low instrumental background, good stability and reproducibility, good vacuum conditions, low isotopic fractionation and also the possibility to measure the samples repeatedly.

5.2 The ion source

5.2.1 General

Most ion sources used for AMS measurements are of a Cs-sputter type, based on a discovery by Krohn (1962) who found that the yield of negative ions from solid samples increases significantly if the sputter surface is continuously overlaid by Cs.

The efficiency of a Cs-sputtering ion source is a function of the Cs ion energy and the coverage of the sample by Cs atoms. Experiments have shown that the optimum yield of negative ions is achieved at a coverage of the sputter target of close to one-half mono-layer (Middleton, 1983). Cs-sputter ion sources suitable for AMS measurements should incorporate the following properties, a high beam current and stability, low beam emittance and energy spread, low cross-talk between samples, low background, minimum of maintenance and also the possibility to use the sputter targets repeatedly and to change the sputter target with only a short interruption. The old Cs-sputter ion source in Lund did not fulfil some of these requirements and therefore a

new source was developed. The principle of the construction of the new ion source, which was recently installed, is very similar to a type that has been commonly used at various laboratories for several years (e.g. Åström and Possnert, 1983, Elmore, 1981 and Purser et al., 1981).

Preliminary measurements show that the ion source is capable of delivering an analysed negative carbon beam of approximately 15 μA. The stability of the source over several days is better than 10% and short term variations are less than 1%.

5.2.2 Function

A drawing of the new ion source used for AMS measurements in Lund is shown in Fig. 6. The ion source is a Cs-gun, multiple sample ion source for solid targets with a target wheel holding 20 samples The mechanical construction consists of four sections, the caesium gun, the chamber with the lens system, the multiple sample holder and the vacuum system. The chamber is mounted on an adjustable stand and is connected to the ceiling via insulators. The chamber forms the base for the other sections of the ion source. The ion source is equipped with a mechanism to automatically crack the glass ampoule containing the caesium. This mechanism consists of a bellows, containing helium gas, that expands as the volume outside the bellows is evacuated and cracks the glass ampoule containing the caesium by a lever.

The Cs gun consists of a Cs reservoir, which is heated to approximately 200 °C. The Cs vapour diffuses through the tungsten crystal, heated to approximately 1200 °C, from which Cs^+ ions are extracted by a 5 kV potential gap. The Cs^+ beam is focused by the two Einzel lenses and by the non-uniform electrical field within the hemisphere and further accelerated, by a 20 kV potential gap. The central segment of the second Einzel lens is split into four sections, which provides a possibility to steer the Cs beam. From the target negative ions are sputtered by the Cs^+, accelerated to an energy of 20 keV, focused by the hemisphere and by an Einzel lens and steered by the electrostatic steerer. Since the whole ion source assembly is elevated to a negative potential of 20 kV, the ion beam will emerge from the source with an energy of 40 keV.

The multiple sample holder makes it possible to change the samples easily without letting air into the chamber. The sample head is electrically insulated from the chamber potential by 2 rotating glass rods. These glass rods also

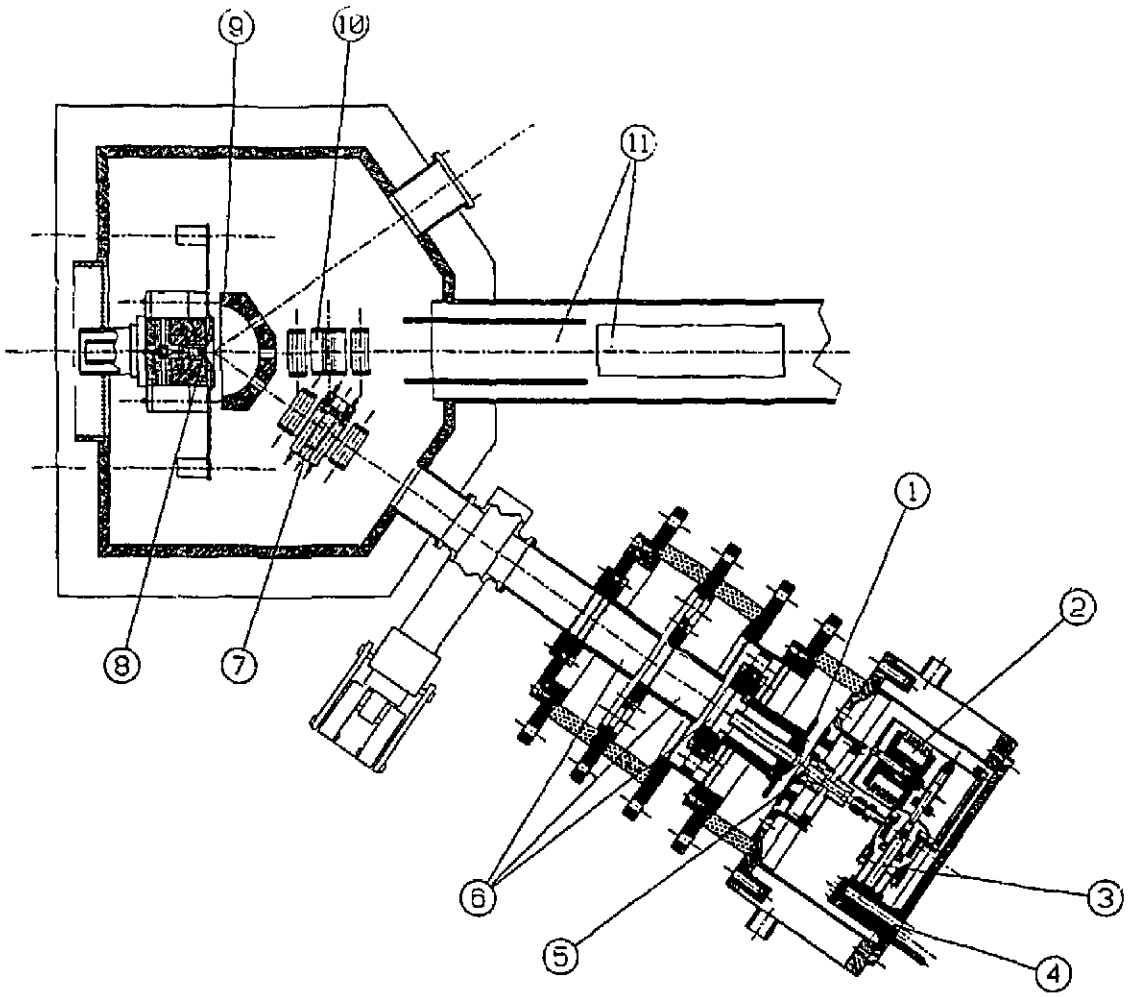

Fig. 6. A drawing of the new Cs-sputter ion source in Lund. The legends correspond to: Cs extraction electrode (1), bellows (2), Cs ampoule (3), Cs heater, cooling facility and thermocouple (4), tungsten crystal and heater (5), Einzel lenses for the Cs beam (6 and 7), sputter target (8), hemisphere (9), Einzel lens for the negative ion beam (10) and electrostatic steerer (11).

control the rotation of the sample head and the rotation and wobbling of the samples, respectively. The rods are manoeuvred by 2 air motors. A screen, which defines the potential surface of the sample head, also serves as a protection of the samples not in use, preventing cross-contamination.

5.3 Transmission

5.3.1 Stripping yield and terminal pumping

The terminal of a tandem accelerator is equipped with a stripper arrangement. The stripper can be either solid (thin carbon foil) or gaseous. When a beam of fast ions passes through the stripper material, ionisation and excitation processes will change the incoming negative ion beam into a multiply charged, positive ion beam. The charge state distribution emerging from the stripper is a function of the atomic number and velocity of the incoming ion beam and the atomic number, thickness and density of the stripper target (Suter, 1990). If the stripper target is thick enough, the ion beam will establish an equilibrium charge state distribution (Betz, 1972). The stripper efficiency can be optimised by the appropriate choice of ion velocity, charge state and target material. For this optimisation a knowledge of the charge state distribution is necessary. In the publications presented by Wittkower and Betz (1973) and Shima et al. (1985), previously reported, experimentally derived, equilibrium charge state distributions are summarised. According to these reports, charge state distributions have been obtained for the following stable isotopes to long-lived ("AMS") radionuclides, using carbon stripper foils and a terminal voltage of between 1.5 and 3 MV, ^{9}Be and ^{12}C by Stoller et al. (1983), ^{12}C by Girardeau et al. (1971), ^{27}Al by Druetta et al. (1983) and Lennard et al. (1981), ^{35}Cl by Wittkower and Ryding (1971) and by Hofmann et al. (1984) and ^{127}I by Wittkower and Ryding (1971). All these results were obtained by measuring the charge state distribution close behind an external stripper medium. If the transmission through the high-energy side of the accelerator is charge-state dependent, the charge state distribution emerging from the accelerator will be different compared to the distribution measured close to the stripper foil. Therefore, the charge state distribution obtained, when using the permanent stripper arrangement inside the tandem accelerator, has been derived for a number of ions (^{9}Be, ^{12}C, ^{13}C, ^{16}O, ^{27}Al, ^{35}Cl, ^{48}Ti and ^{58}Ni) in the Lund Pelletron accelerator. The results for ^{12}C and ^{13}C are shown in Fig. 7. Any difference between the results obtained when

using the terminal stripper compared to the results obtained when using an external stripper must, if no fundamental difference between the experiments exists, be due to differences in the transmission between the stripper foil and the detector set-up.

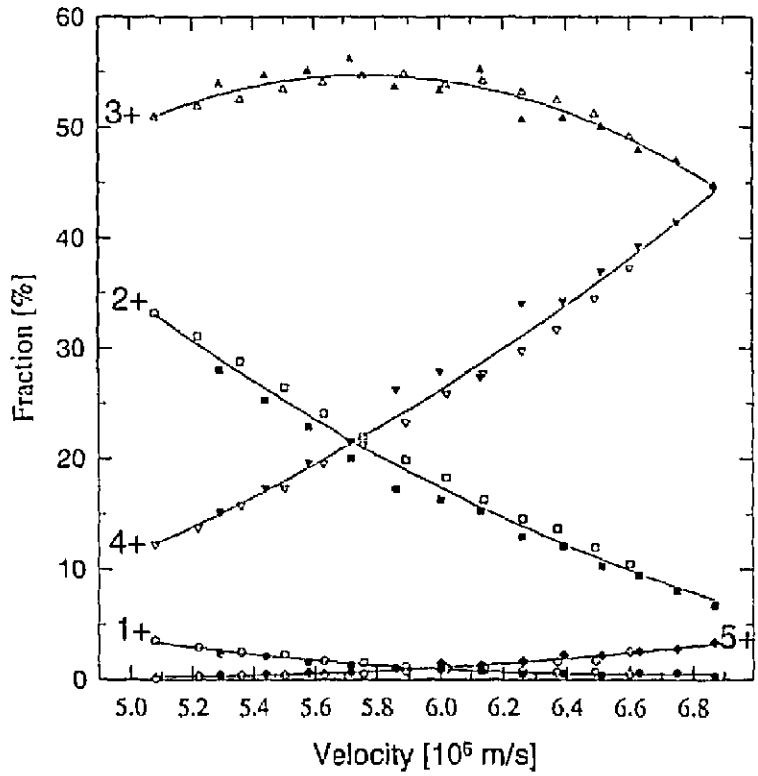

Fig. 7. Equilibrium charge state distribution for a ^{12}C (solid symbols) and ^{13}C (open symbols) obtained with a 2 μg/cm² carbon stripper foil. Lines are to guide the eye.

For ions, such as Be and Al, which have a low affinity for electrons, and are therefore commonly injected as negatively-charged molecules viz., as BeO^- and Al_2^-, the emittance of the beam will increase due to Coulomb repulsion between the positively charged fragments as the molecules break up in the stripper foil. The repulsion increases with the charge state of the ions (Hofmann et al., 1987). The effects due to Coulomb repulsion can be minimised by using stripper gas exclusively, instead of a solid stripper medium, or gas, at low pressure, in combination with a stripper foil (Hofmann et al., 1987). The molecules will then break up, due to the low-density gas, prior to their being stripped in a gas of higher density or by the foil.

The carbon stripper foils normally used when performing AMS measurements in Lund have a thickness of 2 μg/cm². During irradiation, the thickness of the carbon foils change in the irradiated area, and eventually they are destroyed (Tait et al., 1979). This change in thickness leads to a variation of the energy loss and in the scattering of the ions passing through the foils, and therefore to a time dependent transmission through the accelerator system (Tait et al., 1979). Gas cells have a much better homogeneity over time and are indestructible, leading to an improved beam stability compared to stripper foils. This is a most important property when performing high-precision measurements using heavy ions, such as AMS and beam foil spectroscopy (BFS). However, in many tandem accelerators, as in the case of the Lund Pelletron accelerator, the stripper gas is pumped through the accelerator tubes, using pumps outside the accelerator tank. This leads to a, using gas, significantly higher pressure in the accelerator tubes and gives rise to a lower transmission due to the spreading of the beam by collisions and by charge-exchange processes. In fact, the maximum transmission through an accelerator without terminal pumping is reached at a stripper-gas pressure below that required to obtain equilibrium charge state distribution (Bonani et al., 1990). The most significant transmission losses take place on the low-energy side accelerator tube due to the high cross-section for charge exchange, in the case of carbon, from C^- to C^0 (Bonani et al., 1990). Therefore, a suggestion for a new stripper design has been presented, based on our rebuilt NEC stripper house (Hellborg and Håkansson, 1982), see Fig. 8.

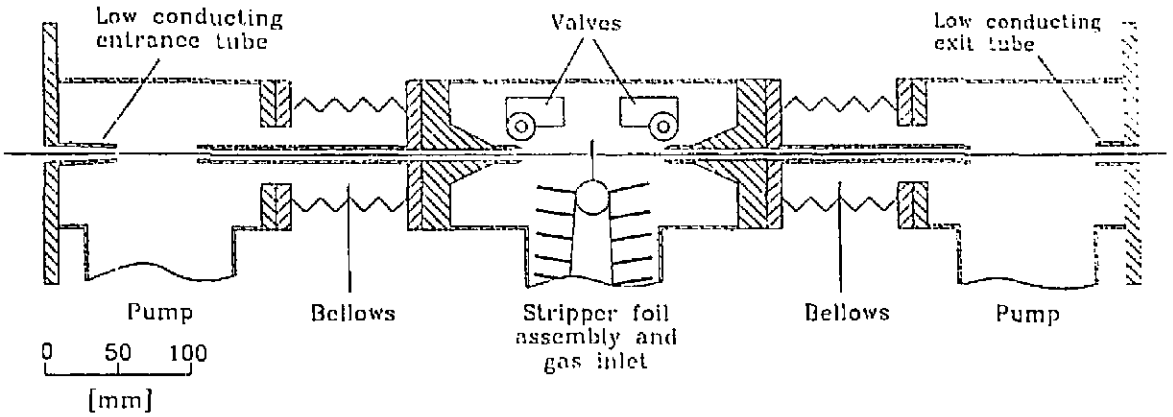

Fig. 8. Schematic design of the new stripper.

The incorporation of the new stripper into the terminal of the tandem accelerator will yield an improved transmission through the accelerator system, compared to our present gas-stripper arrangement, and to improved beam stability.

5.3.2 Isotopic fractionation

Isotopic fractionation by the measuring apparatus is a problem that has to be taken seriously in AMS. Since the charge state distribution obtained for an ion beam is velocity-dependent, the distributions will be different for the different isotopes in the beam. The effect can change the isotopic ratios measured by many percent, but since the measurements are performed relative to a standard, the effect is not as serious as it first appears. However, there are velocities or ion energies at which the isotopic fractionations are minimised. The isotopic fractionation is the smallest when the charge state fractions vary only slightly with energy, i.e. near the maximum yield of the charge state chosen (Hofmann et al., 1984). It is clear that the energy of the negative ions must not vary before the electrons are stripped if accurate relative measurements are to be made (Litherland, 1980). The isotopic fractionation, F(m), is defined here as one minus the ratio of the charge state fractions between two isotopes. The isotopic fractionation for $^{14}C^{3+}$ relative to $^{13}C^{3+}$, stripped using a 2 μg/cm^2 thick carbon foil, is shown in Fig. 9. F(14) was calculated from the measured result of $^{13}C^{3+}$, shown in Fig. 7.

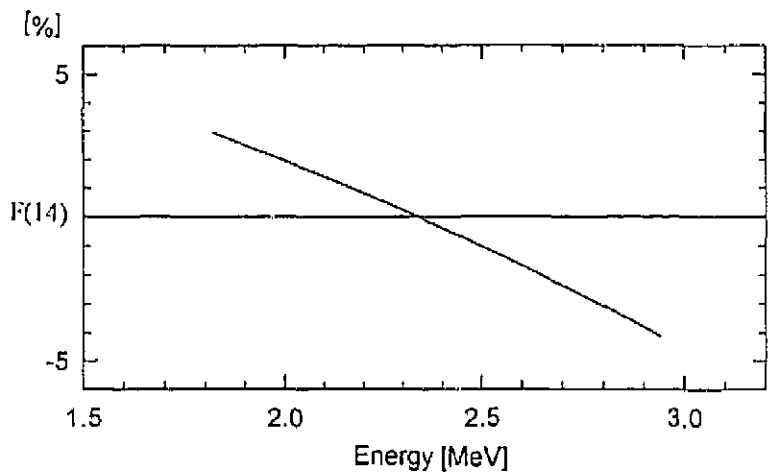

Fig. 9. Isotopic fractionation $F(14) = 1 - \phi_3^{14} / \phi_3^{13}$. *The line was calculated from the experimental values of* ^{13}C.

The results show that in order to obtain the smallest isotopic fractionation between $^{13}C^{3+}$ and $^{14}C^{3+}$ a terminal voltage of approximately 2.29 MV should be applied, which, in addition to the injector voltage of 40 kV, yields a stripping energy of 2.33 MeV.

Effects of isotopic fractionation in the rest of the system are potentially more serious, especially if they are time-dependent. According to Litherland (1980), time-dependent isotopic fractionation can be encountered in the sputtering process in the ion source. Another problem that might occur is if the degradation of the stripper foil during a measuring cycle affects the transmission of the isotopes to be measured differently.

5.3.3 Mathematical description of the beam transport

The effects caused by the charge exchanging processes in the accelerator tubes and in the stripper can be calculated in order to model the beam transport if the pressure profile in the accelerator system and the different cross sections for charge exchange are known. The charge of a fast ion moving through material fluctuates as a result of electron loss and capture in collisions with the atoms of the target material.

The change in the proportions of the charge states in an ion beam within a gas target is described by the following system of linear, coupled, differential equations (Tawara and Russek, 1973):

$$\frac{dF_m}{dx} = \sum_{j \neq m} (F_j \sigma_{j \to m} - F_m \sigma_{m \to j}) \qquad m = -1, 0, 1, \cdots, Z$$

with the normalisation:

$$\sum_{m=-1}^{Z} F_m = 1$$

where F_m represents the fraction of ions with charge m, $\sigma_{j \to m}$ the charge exchange cross section for changing from charge state j to m, and $\frac{dF_m}{dx}$ the change in the fraction of ions in charge state m over a target thickness dx, where x refers to the number of target particles per cm^2 traversed by the beam. At large values of x, $\frac{dF_m}{dx} \to 0$ and equilibrium conditions are reached.

In solid targets, the density effect becomes important. This is due to excitation processes in the ion beam. At higher densities the ions will not

have enough time to de-excite between two collisions (Betz, 1972), leading to an average equilibrium charge state approximately 5% higher than for gas targets (Booth and Grant, 1965).

5.3.4 Beam transport losses and detection efficiency

In order to achieve a high transmission through the accelerator for heavy ions the beam optics on the low-energy side must be optimised to focus the beam through the stripper channel in the terminal.

The beam loss in a tandem accelerator due to charge exchange processes in the accelerator tubes can, under foil stripping conditions, be shown to be very small. Instead the optical transmission is often the limitation. The electrostatic field due to the accelerator tubes focuses the beam. This effect varies with the energy and charge state of the ion. Angular and energy straggling in the stripping process will also lower the overall transmission. For C, the maximum transmission through the Lund Pelletron accelerator system using carbon stripper foils has been measured by comparing the positive ion current beyond the accelerator to the negative ion current just in front of the accelerator and was found, after correction for the charge state distribution, to be about 60%.

For a standard type of ΔE-E gas counter telescope the detection efficiency is close to 100%. For a gas filled magnet in combination with time-of-flight detection the detection efficiency can be significantly lower (Suter, 1990). The efficiency of detecting the characteristic X-rays of the projectiles, as described in section 3.4.4, is typically less than 1% (Artigalas et al., 1994).

5.5 Present accuracy and efficiency of the Lund AMS system

Over the past few years several modifications of the Lund Pelletron accelerator facility have led to improvements both of the accuracy and efficiency, thus making the accelerator system suitable for AMS measurements. The optical system on the low-energy side has been re-built to improve the transmission through the accelerator when experiments using heavier ions are performed, and vacuum pumps with higher pump capacity and lower final pressure have been installed, also improving the transmission through the system (Hellborg et al., 1990). A new magnetic quadrupole triplet lens and a Wien filter have been installed on the experimental beam line. Computer control of the dipole magnets and the Wien filter has also

been implemented, making accurate and efficient alternation between the different settings of the system during each measuring cycle possible. The new ion source will significantly improve the accuracy and reproducibility of the system due to its multiple target wheel, ensuring identical conditions for the different samples, stable operation conditions and lower background compared to the sputter ion source previously used. The higher beam current of the ion source and much reduced interruptions for sample changes will significantly improve the efficiency of the system. The present accuracy of the AMS system is shown in Fig. 10, presenting some results obtained in April 1995. Future improvements of the AMS system will include, further development of the ion source including an electrostatic deflector on the injector, installation of a terminal pump in the accelerator facilitating the use of a gas stripper with only a minimal transmission loss, improved accelerator voltage stability, the installation of a new current integrator and the use of a ΔE-E gas counter detector in combination with a two-parameter data collection system yielding three-dimensional presentations as shown in Fig. 5.

With the new ion source the beam current of $^{13}C^{3+}$ obtained, measured at the beam stop just in front of the detector set-up, has increased by approximately a factor of 3 compared to the old ion source, and at present a current of 50-60 nA can be produced. With further development of the ion source the current is expected to increase. With this current enough ^{14}C counts for a 1% statistical uncertainty can be achieved within 15 minutes for a contemporary carbon sample. The precision of the AMS system is at present around ±1.5%.

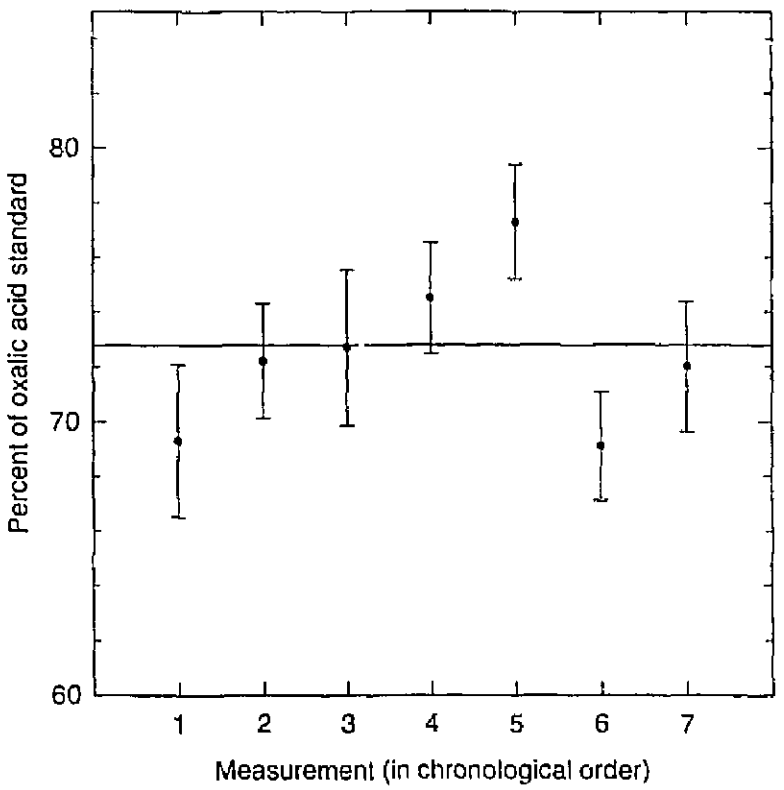

Fig. 10. These results were obtained for 7 different measurements of an old charcoal sample, over a period of 2 days in April 1995. Each measurement was performed over 10 cycles, and from these results the standard deviation of the measurement was calculated. The error bars correspond to ±1σ, where σ is the larger one of either the statistical uncertainty of each measurement or the calculated standard deviation for each measurement. The ^{14}C age was calculated, using the results obtained for an NBS oxalic acid standard, to 2259±120 years, well in line the result 2000±70 years, obtained at the Radiocarbon Dating Laboratory in Lund. The uncertainties in these results correspond to ±1σ. The statistical uncertainty of the measurement of the oxalic acid standard was less than 1%..

6. Summary of the papers

6.1 Paper I

"The charge state distribution of carbon beams measured at the Lund Pelletron accelerator"

A. Wiebert, B. Erlandsson, R. Hellborg, K. Stenström and G. Skog

Paper I was presented at The Third European Conference on Accelerators in Applied Research and Technology (ECAART) held in Orleans (France) 31/8-4/9-93 and published in the proceedings, Nuclear Instruments and Methods in Physics Research B 89 (1994) 259-261.

In this paper the first results of the obtained equilibrium charge state distribution for 1.7-2.8 MeV ^{12}C and ^{13}C ions stripped in the high-voltage terminal were presented. The measurements were performed by using 2 $\mu g/cm^2$ thick carbon stripper foils. If the charge state distribution of the ion beam is known, isotopic fractionation effects, due to drifts in the accelerating voltage, can be minimised by the choice of the accelerating voltage.

The energy of the different charge states was measured by using a particle detector. In order to reduce the intensity of the ion beam, ions backscattered on a 50 $\mu g/cm^2$ gold foil (ϕ=16 mm) were analysed. The backscattering angle was 165°. The experimental set-up is shown in Fig. 11.

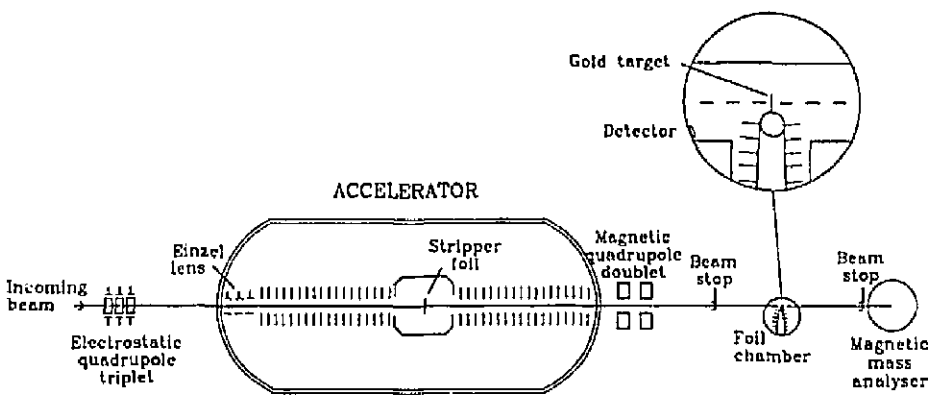

Fig. 11. The schematic sketch of the experimental set-up.

For each terminal voltage setting, at least three measurements were performed using different target foils. This was done in order to verify that the target foils were not broken, since a crack, for instance in the centre of the foil would reduce the intensity of the ions at higher charge states, as the focusing of the high-energy accelerator tube and the magnetic lens act more strongly on the higher charge states. Also for each new terminal setting, the beam, prior to being charge state analysed, was focused through a diaphragm (ϕ=8 mm) which replaced the target foil, in order to verify that the beam was well focused so that the majority of the ions in the beam would hit the gold target. The stripper foils were also checked repeatedly.

Measurements of the charge state distribution for carbon beams previously reported had been done using external strippers followed by a magnetic analyser (Stoller et al., 1983 and Girardeau et al., 1971). The main advantages of measuring the charge state distribution as presented in paper I are that all charge states can be measured simultaneously, so that normalisation of the beam current is unnecessary, and that any transmission through the high-energy side of the accelerator tube which is dependent on the charge state, is included in the measurements. The results obtained showed a small shift towards a higher average charge state as compared to the results reported by Stoller et al. (1983) and a somewhat larger shift towards a higher average charge state compared to the results obtained by Girardeau et al. (1971), suggesting a charge state-dependent transmission through the high-energy side accelerator tube. However, as the target foils tended to crack when heated by the beam, the measurement series was later repeated using more reliable gold targets, see paper III.

According to the results obtained, carbon ions should be stripped at an energy of 2.44 MeV in order to minimise the isotopic fractionation between $^{14}C^{3+}$ and $^{14}C^{3+}$.

6.2 Paper II

"The pressure profile in the Lund Pelletron accelerator"

A. Wiebert, B. Erlandsson, R. Hellborg, K. Stenström and G. Skog

Paper II has been accepted for publication in Nuclear Instruments and Methods in Physics Research A and is at the moment in press.

In this paper the pressure profile of the Lund Pelletron accelerator was derived in order to perform the transmission calculations presented in paper III. A series of measurement was performed both under foil stripping conditions, with no stripper gas input, and at several settings of the needle valve controlling the stripper gas input, in order to correlate the pressure in the beam tubes on either side of the tandem accelerator to the pressure in the terminal. With this information the pressure profiles could be calculated both under foil stripping conditions and under the gas stripping conditions under which the transmission of a ^{12}C beam is at a maximum, according to the measurements presented in paper III.

The calculations were based on the kinetic theory of gases originally presented by Knudsen (1909). As all parts of the accelerator system fulfil the inequality $D/\lambda < 0.4$, equations of molecular gas flow were used throughout (Roth, 1982). D/λ is known as the Knudsen number, where D is the diameter of the vacuum enclosure and λ is the mean free path of the gas molecules. The mean free path in centimetres can be calculated roughly using the expression $\lambda=6.6\times10^{-3}/P$, where P is the pressure in mbar (Grunderna i Tillämpad Vacuumteknik, 1988).

The results obtained in these calculations, in combination with the results presented in paper III, clearly showed the limitations of the present gas stripper arrangement in the terminal of the accelerator. Gas strippers are preferable when heavy ions are accelerated and when stable beam conditions are required, i.e. when AMS measurements are performed. However, in order to fully take advantage of the gas stripper, the terminal of the accelerator must be equipped with a terminal pump. In the case of the Lund Pelletron accelerator the stripper gas is pumped through the accelerator tubes using pumps outside the accelerator tank. This leads to a considerably higher pressure in the accelerator tubes and will lower the transmission by spreading the beam by collisions and by charge-exchange processes. Therefore, a

suggestion for a new gas stripper arrangement, including terminal pumping, was presented in the paper, and is shown in Fig. 8.

Some of the results obtained in the calculations could be compared with results of measurements previously made by the manufacturer of the accelerator. The results agreed beyond expectation and differed by less than 3%.

6.3 Paper III

"The transmission of a carbon beam through a tandem accelerator"

A. Wiebert, B. Erlandsson, R. Hellborg, K. Stenström and G. Skog

Paper III has been accepted for publication in Nuclear Instruments and Methods in Physics Research A and is at the moment in press.

In this paper both calculations and measurements of the non-equilibrium and equilibrium charge state distribution and transmission for a carbon beam were presented.

The measured equilibrium distributions for 1.7-2.9 MeV ^{12}C and ^{13}C beams were obtained in a similar manner as explained in paper I. However, in order to the reduce the uncertainty due to unreliable gold target foils, the measurements were performed using gold targets (approximately 40 $\mu g/cm^2$ thick), made by evaporating gold onto pieces of perspex (35×25 mm^2). The measured equilibrium distributions were very similar to the results presented in paper I, i.e. indicating a small shift towards a higher average charge, compared to the distributions obtained by using external stripper foils (Stoller et al, 1983). Again, the results obtained by Girardeau et al. (1971) were different from our result, with a noticeably lower mean charge state. Further investigations, to study the charge state distribution obtained for ^{16}O, see paper V, showed the same shift compared to the charge state distribution for ^{16}O obtained by Girardeau et al. (1971), indicated that a systematic shift existed between the measurements.

The measured non-equilibrium distributions were obtained for a 2.44 MeV ^{12}C beam. The measurements were performed using N_2 gas stripper with target thicknesses varying between approximately 0.01 and 0.5 $\mu g/cm^2$. The results of the measurements are shown in Fig. 12 together with the result of the calculations. The measurements showed that the maximum transmission of charge state 3+ through the accelerator was 10%, to be compared with a transmission of 33% using a foil stripper, and was obtained at a target thickness of 0.2 $\mu g/cm^2$.

The calculations of the non-equilibrium charge state distribution was performed by using the system of coupled differential equations given in section 5.3.3. On the basis of the relative size between single- and double electron capture/loss cross sections, all higher order cross sections for charge

exchange were assumed to be insignificant. The varying target thicknesses along the accelerator tubes and in the stripper were calculated using Eq. (5), from paper III, and the pressure profile for maximum transmission of a ^{12}C beam derived in paper II. The cross sections for charge exchange for the positively-charged ions were taken from Niklaus et al. (1994) on the assumption that the charge exchange cross section for C in N_2 gas and for C in Ar gas are, to the first approximation, very nearly equal (Betz, 1972 and Betz, 1983). The cross sections for charge exchange for singly-charged, negative, C ions stripped in N_2 gas were obtained from previous measurements (Tsuji et al., 1989 and Rahman and Hird, 1986). The charge exchange cross sections for zero-charged ions were assumed to be equal to the corresponding cross section for singly-charged, negative, C ions (Niklaus et al., 1994). The maximum transmission of charge state 3+ was 12% and was obtained at a target thickness of 0.2 μg/cm², well in line with the measured results.

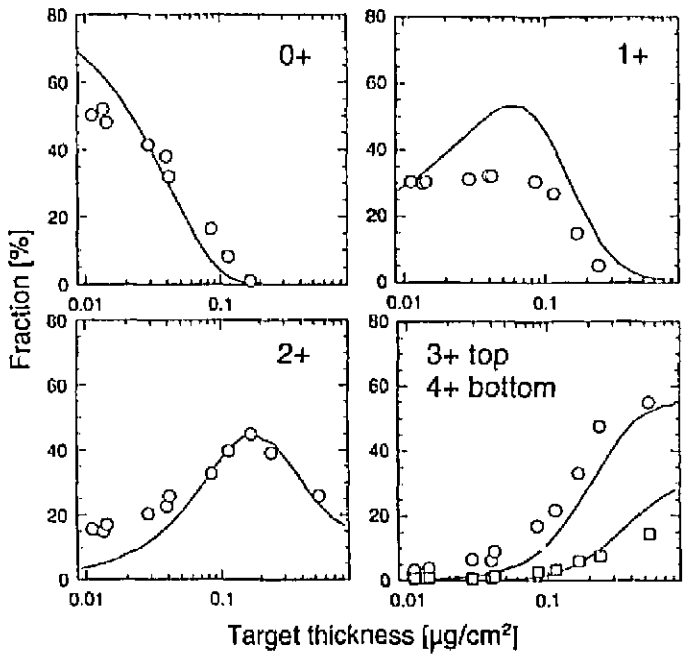

Fig. 12. Measured non-equilibrium charge state distribution for a 2.44 MeV ^{12}C beam stripped in N_2 gas, obtained at the high-energy side of the accelerator. The solid lines represent the calculated charge state distribution.

The calculations performed under foil stripping conditions, i.e. no gas input, performed in order to examine the size of the transmission loss due to charge exchange processes in the accelerator tubes, using stripper foils. The

results showed that these losses are very small, <1%. Since the total transmission using stripper foils for a carbon beam has been measured to be in the order of 60%, the losses are most probably due to the optical transmission of the system. The target thicknesses along the accelerator tubes and in the stripper were calculated using Eq. (5), from paper III, and the pressure profile under foil stripping conditions derived in paper II.

A hypothetical equilibrium distribution using N_2 stripper gas was shown to have an average charge state 5% lower than the measured equilibrium average charge state. This can be explained by the "density effects in solids" (Betz, 1972). At higher densities the ions will not have enough time to de-excite between collisions, leading to a higher average charge state. The higher average charge state in the measured result may also be due to a charge state-dependent transmission through the high-energy side accelerator tube due to ion optical effects, which is not included in the calculations.

6.4 Paper IV

"A negative caesium-sputtering source for AMS investigations"

K. Håkansson, R. Hellborg, B. Erlandsson, G. Skog, K. Stenström and A. Wiebert

Paper IV has been presented at the 7th International Conference on Heavy Ion Accelerator Technology held in Canberra (Australia) 18/9-22/9-95. The proceedings will be published in Nuclear Instruments and Methods in Physics Research A.

In Paper IV the new caesium sputter ion source was presented. The paper describes the mechanical construction and the special properties of our ion source design. The new source was constructed to meet the special demands required for accurate and efficient AMS investigations. Due to the fact that often very small samples are analysed, a high production of negative ions is essential. Wheel holders for multiple samples are necessary for repeated measurements of the standard sample. The availability of many samples in the ion source at the same time also ensures similar conditions for multiple measurements without changing any source parameters. This is very important since all measurements are performed relative to a standard of known activity. Short interruptions for sample changes and a high beam current imply more efficient measurements. The samples can be manoeuvred in order to find the optimum position of each sample or to increase the life-time of the samples. The construction also facilitates easy maintenance. A technical description of the ion source is given in section 5.2.2.

6.5 Paper V

"The charge state distribution of Be, O, F, Al, Cl, Ti and Ni ions"

A. Wiebert, B. Erlandsson, R. Hellborg, K. Stenström and G. Skog

Paper V has been submitted for publication in Nuclear Instruments and Methods in Physics Research B.

In paper V the charge state distributions for a number of ions, some with applications in the field of AMS, were obtained in a manner similar to that described in paper III. However, in these experiments the backscattering angle was changed to 110°, except in the case the Ni measurements, in order to increase the energy separation between the peaks which correspond to the different charge states. The results were compared with charge state distributions previously reported for Be (Stoller et a., 1983), O (Girardeau et al., 1971), Al (Druetta et al., 1983 and Lennard et al., 1981) and Cl (Wittkower and Ryding, 1971 and Hofmann et al., 1984) measured close to the stripper foil. For the ions injected as molecules, namely BeO and Al_2, it could be concluded that the transmission through the high-energy side accelerator tube was dependent on the charge state. This is mainly due to the Coulomb repulsion between the positively charged fragments as the molecules break up in the stripper. Of the ions injected in an elemental state, the only comparable data could be found for O and Cl in the reports by Wittkower and Betz (1973) and Shima et al. (1985). The charge state distribution for Cl was in good agreement with previously published results by Wittkower and Ryding (1971) and Hofmann et al. (1984) indicating a negligible, charge state-dependent transmission through the high-energy side accelerator tube. As previously mentioned in paper III, the charge state distribution for O obtained by Girardeau et al. (1971) clearly disagrees with our results.

6.6 Paper VI

"Isobar suppression in accelerator mass spectrometry by the detection of characteristic X-rays"

A. Wiebert, P. Persson, M. Elfman, B. Erlandsson, R. Hellborg, P. Kristiansson, K. Stenström and G. Skog

Paper VI was presented at the VII International Conference on Particle Induced X-ray Emission and its Analytical Applications and has been accepted for publication in Nuclear Instruments and Methods in Physics Research B.

In this paper the possibility of using a new detection technique for AMS investigations was explored. For heavier ions, the use of a traditional detector system is often limited due to the insufficient ion energy provided when using a small tandem accelerator. A new detection technique has recently been presented (Artigalas et al., 1993) in which the different incoming isobars are distinguished by their characteristic X-rays that are emitted as the beam is stopped in a target. A schematic sketch of the detector set-up used in the experiments is shown in Fig. 13.

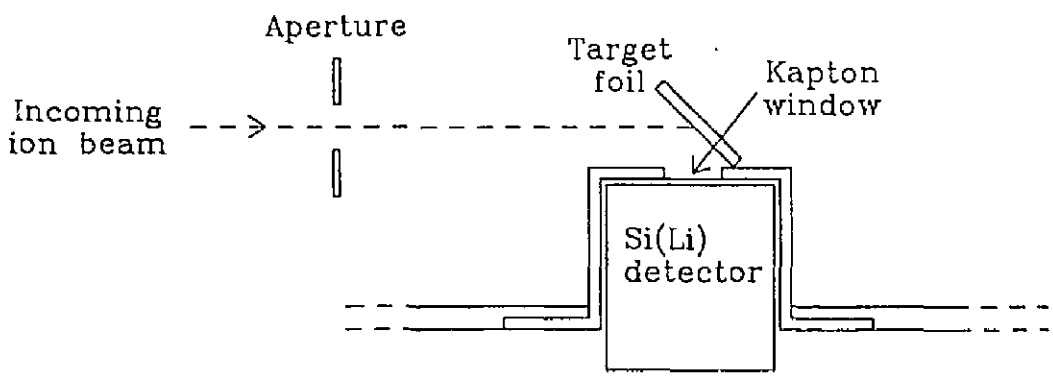

Fig. 13. The incoming ion beam hits the target foil that is thick enough to completely stop the ions. The X-rays emitted in the direction of the Si(Li)-detector have a high probability of being registered. In the actual experiments, performed with a ^{58}Ni beam, the solid angle of the detector was decreased to reduce the count rate.

The radioisotope ^{59}Ni was investigated because of its importance in nuclear waste management. ^{59}Ni is produced by neutron activation in the stainless steel shielding surrounding the fuel in nuclear power plants. Because ^{59}Ni decays only via electron capture (EC) and has a very long half-life ($t_{1/2}=7.6\times10^4$ y) it is very difficult to measure the radiation emitted in the radioactive decay.

When performing AMS measurements of ^{59}Ni by detecting the characteristic X-rays, the two major problems are the detection efficiency and the background in the X-ray spectrum.

The detection efficiency is a function of the X-ray production and of the solid angle of the detector. By bombarding different targets (Fe, Ni, Cu, Zn, GaP, Ge and Se) with a ^{58}Ni beam at different energies (10-27 MeV) it was found that the X-ray production increases with projectile energy and has a maximum for targets with a nuclear charge slightly higher than that of the projectile, i.e. Cu and Zn. Approximately one of every thousand incoming projectiles is registered when a solid angle of detection of 10% is used. The detection limit of the specific activity in a stainless steel sample (containing 10% Ni) can be calculated, and according to these results, is a few Bq ^{59}Ni per gram steel.

The background in the X-ray spectrum in a ^{59}Ni measurement is due either to stable Ni isotopes that have managed to slip through the AMS system or to isobars, emitting X-ray photons with an energy very close to the energy of X-ray photons produced by incoming Ni projectiles. In the case of ^{59}Ni, the most problematic isobar is ^{59}Co, which is relatively abundant in stainless steel (~0.02%). In the paper it was investigated whether the background due to the Co content of the beam could be suppressed by selection of the target foil. According to these measurements a Ge target seems favourable. It was found that the ^{59}Ni content in the sample in the ion source must not be less than 1/160 of the Co content of the sample, in order to achieve a signal-to-background of 1:1 in the energy spectrum, at a projectile energy of 22 MeV, using a Ge target. This corresponds to a specific activity in a stainless steel sample of a few kBq ^{59}Ni per gram steel. However, by performing a chemical purification of the sample the Co content can be significantly reduced, and the detection limit will be mainly due to the efficiency of the detection system.

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