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Chapter 2

Basics of AMS

2.1 Isotopic-ratio measurements

In almost all cases, AMS does not measure the trace nuclide alone, but relies on a stable isotope of the same element which is measured as a reference. AMS can not directly detect the trace atoms in the sample quantitatively. In this sense AMS is similar to conventional (low-energy) mass spectrometry, where in general ratios of isotopic abundances are measured. Only the ratio of trace to reference ions, i.e. an *isotope ratio*, is considered as the relevant result. The raw count rate of the trace nuclide is the product of chemical yield, sputter rate, negative ionization yield, and the detection efficiency of the complete AMS machine. The effects taking place after the ion source may be corrected by normalizing to a standard material, and the sputter rate will not matter if the complete sample is used up; however, the negative ionization yield strongly depends on the chemical and physical properties of the sputter target, therefore only in exceptional cases (e.g [Rucklidge et al., 1990]) will the number of trace-element counts alone allow meaningful conclusions.

The reference isotope may be present in the original sample material, as is the case for radiocarbon dating, or it may be added during the chemical treatment, as e.g. for ^{10}Be exposure dating [Lal 1991]. It is assumed that the reference isotope then follows the trace isotope through the further chemical treatment and through the ion source into the primary beam.

The final AMS result is the ratio of trace to reference isotope. If one is interested in the number of the trace atoms in the sample rather than in the isotopic ratio (i.e. for ^{10}Be exposure dating), it has to be calculated from the known or independently measured content of the reference-isotope in the sample.

2.2 Normalization to a standard material

In AMS one radioisotope of interest and (at least) one stable isotope are measured. The transmission of both ion beams influences the measured isotopic ratio. To compensate for transmission differences a standard material is needed as a reference. The accuracy of the AMS measurement depends on the reproducibility between the unknown and the reference sample. Unavoidable differences in the geometry of the samples serving as a target in the Cs-sputter source induce changes of the beam geometry and consequently of the transmission through every aperture where parts of the beam are lost. Generally a reduction of the losses will increase the accuracy of the results (see Section 5.2.4). The losses can be reduced by careful accelerator tuning, which is crucial both if high precision is needed (^{14}C) or if high mass resolution requires operation with narrow slits (heavy ions). Chemical fractionation does not introduce uncertainties into the final results if it is identical for the unknown sample and for the standard material. Therefore both materials should be subjected to sample preparation procedures as similar as possible.

2.3 Isotopic fractionation

Although isotopes of one element behave very similar in all chemical and physical processes, there exist small mass-dependent shifts of isotopic abundances. At the precision obtained for ^{14}C dating this *fractionation* is significant (sometimes a few percent), which would introduce a significant uncertainty into the measurement. Fortunately, the presence of two stable isotopes allows to determine the fractionation of $^{14}\text{C}/^{12}\text{C}$ by measuring the fractionation of $^{13}\text{C}/^{12}\text{C}$. This allows to separate the change in $^{14}\text{C}/^{12}\text{C}$ due to fractionation from the change due to decay of ^{14}C . Only the latter is the value of interest for ^{14}C dating. Without this correction, the usual ^{14}C dating precision of a few per mil could not be achieved.

Also for ^{36}Cl , the measurement of two stable isotopes (^{35}Cl and ^{37}Cl) allows fractionation correction [Fifield et al., 1987, Snyal et al., 1994].

2.4 The AMS facility

A typical AMS facility consist of an ion source for negative ions, an *injector*, the central tandem accelerator, the *analyzer*, and finally a detector (see Figure 1.1). Both the injector and the analyzer combine electrical and magnetic components to form mass spectrometers. Molecular ions break up during the stripping process in the accelerator (no light molecular ions with charge state 3+ or more are observed). The injector separates out the negative ion beam which contains the radionuclide of interest, but also molecular ions, often at the nA level. The accelerator breaks up all molecules, and the analyzer is tuned to select the atomic mass of the radionuclide, which is measured in the detector at a typical rate between 10^3 cps and 10^{-3} cps. Neither the injector nor the analyzer provide separation of atomic

isobars.

An additional advantage of acceleration is the reduction of the beam emittance, which improves the separation of the following mass filters. Additionally the higher energy may allow one to discriminate atomic isobars by means of their different interaction with matter (e.g. different energy loss or different average charge). However, with tandem accelerators these methods work only up to a mass of about 70. Therefore [Kilius et al., 1990] proposed that high energies are of no special advantage for heavy ions. [Kilius et al., 1987] have presented measurements of ^{129}I , where the isobaric ^{129}Xe does not form negative ions (see Section 2.4.1) at a terminal voltage as low as 0.75 MV. However, in the meantime it turned out that higher energies have the additional advantage of smaller cross sections for scattering and charge change at the residual gas in the analyzer. Most involved cross sections fall off with E^{-n} , where n is in the range of 1 to 5/2 for the various processes [Suter et al., 2000].

2.4.1 Ion source

AMS with tandem accelerators has to use negative ions, which has led to the success of the Cs-sputter source [Middleton 1983]. Cs is evaporated, ionized at a hot surface, and a Cs^+ beam of a few keV energy is used to sputter the sample atoms. In addition to providing the sputter beam, a Cs layer on the sample surface serves a donor for electrons, leading to an enhanced yield of negative sample ions [Nadeau et al., 1987]. The Cs-sputter source does an important first step in isobar separation in *not* producing many species of negative ions. A rough estimate of the negative ion yield for a certain element is the electron affinity, but even for negative electron affinities in the atomic ground state, metastable ions in an excited state can exist which live long enough to reach the accelerator terminal. However, the Cs-sputter source creates no interfering negative ions for $^{14}\text{C}^-$, $^{26}\text{Al}^-$, $^{41}\text{CaH}_3^-$, and $^{129}\text{I}^-$, because the corresponding stable isobars ^{14}N , ^{26}Mg , $^{41}\text{KH}_3$, and ^{129}Xe do not form negative ions. Additionally, the Cs-sputter source creates readily many species of negative molecular ions, allowing to measure elements which do not create negative atomic ions easily, such as Ca (see above). As a general rule, larger molecules will have higher electron affinities. Beams of noble gases cannot be created by the Cs-sputter source, since they generally form neither negative atomic ions nor molecules.

Due to the widespread use of the Cs-sputter source, the sample is often called *target* in AMS. This may be confusing for accelerator physicists, since this target is mounted at the “wrong” side of the accelerator.

2.4.2 Electrostatic and magnetostatic filters

Different ion species follow different trajectories in electrical and magnetic fields. This is applied by the following beam filters to separate the ions of interest from the background. Filtering also requires beam focusing through slits to suppress the separated background. The separators may be focusing on their own, or additional focusing lenses may be used.

2.4.3 Electrostatic Analyzers

An electrostatic analyzer (ESA) consists of cylindrical or spherical electrodes at a distance d and a potential difference ΔU between which the nominal beam follows a circular trajectory of radius r_0 . The electrodes extend over the angle ϕ . In the case of spherical electrodes, the device provides focusing both in horizontal and vertical direction. 1:1 imaging can be achieved for an object $r_0 \tan(\phi/2)$ before the entrance to the image at the same distance after the exit. The ESA acts as a filter for E/q , i.e. for ion energy over charge:

$$\frac{E}{q} = \frac{\Delta U r_0}{2d} \quad (2.1)$$

Ions with a small relative energy deviation $\Delta E/E$ obtain a spatial separation of

$$\Delta x = r_0 \frac{\Delta E}{E} \times 2 \quad (2.2)$$

at the image position.

2.4.4 Bending Magnets

A bending magnet consists of a gap between two pole shoes of opposite magnetic polarity. In the constant field B inside the gap the ions follow circular trajectories. The bending angle α is the deflection angle of the nominal beam which follows a nominal radius r_0 . The focusing properties depend on the so called *shimming angles* at which the beam passes the entrance and exit pole faces. For magnets with deflecting angle $\phi = 90^\circ$, symmetrical shimming angles of $\tan^{-1}(1/2)$ supply symmetric 1:1 focusing from an image position $2r_0$ before the entrance to a symmetrical position after the exit. The bending magnet acts as a filter for momentum over charge:

$$\frac{\sqrt{2mE}}{q} = Br_0 \quad (2.3)$$

where m is the mass of the ion, and B is the magnetic field strength. Ions with a small relative energy or mass deviation obtain a spatial separation of

$$\Delta x = 2r_0 \left(\frac{\Delta E}{E} + \frac{\Delta m}{m} \right). \quad (2.4)$$

2.4.5 Wien filters

A magnetic field is combined with an electric field, perpendicular to the incident beam and to each other, so that the forces on the ions of interest cancel out and the nominal

trajectory is straight. This is achieved by electrostatic deflector plates with a distance d and a potential difference ΔU placed within the gap of a bending magnet with field strength B . To pass a Wien filter, particles must either be neutral or have the velocity

$$v = \sqrt{\frac{2E}{m}} = \frac{\Delta U}{Bd}. \quad (2.5)$$

A Wien filter provides no (useful) focusing, therefore usually a lens is placed before the Wien filter. If the image is at a distance s after the center of a Wien filter with length l , we obtain the separation

$$\Delta x = \frac{lsq\Delta U}{4Ed} \left(\frac{\Delta m}{m} - \frac{\Delta E}{E} \right). \quad (2.6)$$

We see that the separation can be increased by simultaneously increasing the electric field $\Delta U/d$ and the magnetic field B . The latter is necessary to fulfill the condition of 2.5.

2.4.6 Tandem accelerators and stripping

This type of accelerator first accelerates negative ions towards a positive terminal, where the ions lose several electrons by interacting with a thin gas or a foil. After this so-called *stripping* the ions are positively charged and undergo another acceleration when continuing their path from the terminal to ground potential. Since the ions usually lose several electrons, the second acceleration is usually larger than the first (for large accelerators with a terminal at about 15 MV, heavy ions may lose a dozen electrons, and the resulting energy is above 150 MeV).

2.4.7 Detectors

The rare-isotope ions in the filtered beam are finally measured as events in detector systems common in nuclear physics. Solid state detectors (e.g. surface barrier Si) are the most easy to handle, but are quite vulnerable to radiation damage and show a low energy resolution for heavy ions. Ionization chambers show a better energy resolution for heavy ions. If discrimination of isobars is required, a chamber with a split anode allows to obtain several ΔE (energy loss) signals.

The detectors with the highest mass resolution are time-of-flight systems. They usually derive timing signals from secondary electrons emitted when the ion passes through a thin foil.

The main difference between ion separation by filters and identification by detectors is the limited number of background events $\lesssim 10^4$ cps the latter can accept. Therefore the

first method is to be preferred in AMS.

2.4.8 Beam sequencing

Two principally different methods exist for measuring both the reference and the trace isotope: simultaneous and sequential injection.

In the case of *simultaneous injection*, both beams are injected at the same time into the accelerator. Since it is not possible to use the un-analyzed beam directly from the ion source, simultaneous injection requires recombination of the two analyzed beams. This is usually done with two magnets (*analyzer* and *recombinator*, see e.g. [Southon et al., 1990]). The symmetry plane between the magnets is the focal plane for all masses, and apertures and shields can be used to select and suppress different masses. All selected beams are combined in the second magnet and injected into the accelerator. A very advanced implementation of simultaneous injection exists at the Naval Research Laboratory [Grabowski et al., 1997], where a so called *270°-Brezel* magnet separates and recombines masses with a dynamic range of 1 to 200 amu. Simultaneous injection has a significant disadvantage: since large currents of the reference ion are present in the accelerator while the trace isotope is measured, the background caused by charge changing collisions in the stripper tube (which usually is the limiting factor for heavy ion measurements) will be unavoidably enlarged by several magnitudes. On the other hand, the simultaneous injection has the advantage that all elements of the system until the separation of the isotopes after the accelerator are passed simultaneously and all beams experience the same, unavoidable fluctuations.

The alternative is *sequential injection*. The machine setup is periodically changed to inject the trace and the reference ion alternately. The disadvantages of this method are obvious: First, the time used to measure the reference ions, and even more the time needed to change the power supplies, reduces the net counting time for the trace isotope. Second, source current or machine transmission variations which are faster than the rate of sequential injection will not be reflected in the data which introduces an uncertainty. The contradictory requirements for frequent sequencing and long net counting time can be addressed by reducing the time needed to switch between the different beams. The magnetic elements impose the major problem in this task. With the usual bending magnet design switching times of tens of seconds are required, with sophisticated controllers switching times of a few seconds can be reached [Perry et al., 1997]. Laminated iron for the pole shoes and the housing will further lead to significant improvements. An ingenious solution for the injector magnet is to adjust the energy of the ions rather than the magnetic field. This is performed by applying a switched voltage to the insulated magnet box [Purser et al., 1980]. The achieved switching time is of the order of microseconds [Suter et al., 1984]. This method will be called *fast sequencing* in the following. In order to correct for unavoidable differences in the pathways of the trace and the reference beams, additional fast-switched steerers are required. The large beam load variations caused by fast switching may impose a problem, not only for the accelerator, but also for power supplies of beam guiding elements [Suter et al., 1984].

At the high energy side, switching of components is not required for light ions. The different mass results in different deflection in the analyzer magnet, and so called *offset* Faraday cups can be placed at the positions of the ^{12}C and the ^{13}C beam. However, the combination of molecular ions and electrostatic separators requires switched power supplies, if the electrostatic elements are placed before the analyzer magnet [Suter et al., 1984].

If both the reference and the trace isotope are to be measured with the same detector, switching of the analyzer magnet is also required. A sufficient energy change is not possible by applying a voltage to the magnet chamber at the high energy side. As an alternative, electrostatic deflector plates can be installed before and after the magnet which compensate the different deflection in the magnet [Sie et al., 2000].

2.5 Background and Sensitivity

The principal limit for AMS sensitivity is the total number of events of the rare isotope species during a measurement. This number follows from the number of atoms in the sample multiplied by the chemical yield multiplied by the total detection efficiency of the machine. However, a second limitation exists due to the background which is measured for samples containing nominally no atoms of the rare isotope. This background can arise from contamination in the environment or in the chemical treatment, or from ions of other species which are not sufficiently separated by the AMS machine and detector. The *lower limit of detection* (LLD) is usually defined as three times the $1\text{-}\sigma$ uncertainty of the background. Therefore the uncertainty (including the reproducibility) of the background determines the limitation rather than its actual value (however, in the case of event counting, there is a fixed relation between the background and its minimum uncertainty due to Poisson statistics). We will not cope with chemical contamination in the following, and also omit atomic isobars which were explained before.

2.5.1 Sputter tails

The spectrum of the ions generated by sputtering shows a high energy portion which results from excess energy imparted to the neutral secondary ion by the sputtering beam. The resulting distribution has an approximate E^{-n} dependency with n in the range 2 to 3 [Kilius et al., 1990]. Therefore both electrostatic and magnetic separation is required in the injector, since the higher energy in the tail may result in equal magnetic rigidity for abundant ions of lower mass.

2.5.2 Ions with same m/q

When choosing a charge state for the high energy side, one must avoid charge states which have common dividers with the rare isotope mass. As we will show in Section 7.1, ions with

the same m/q will not only pass through the analyzer, but also follow the ions of interest through the injector if they form molecules with the same mass. Therefore no separation for these ions exists before the detector, which will in many cases render measurements impossible.

2.5.3 Charge change in the accelerator tubes

Molecular negative ions with the same mass as the ion of interest impose a problem, even in those cases where they do not have the same m/q . As a part of the molecular fragments will always undergo charge exchange in the accelerator tubes due to collisions with residual gas, the final energy of the ion depends on the position of the charge change. This leads to an energy continuum of these fragments. For every mass and charge state a matching energy will exist which leads to the same magnetic rigidity and therefore allows the ions to pass through the analyzer magnet. These ions must be suppressed by electrostatic filter elements, i.e. a (pure) electrostatic analyzer or a Wien filter.

2.5.4 Scattering at residual gas

Based on geometrical arguments and the relevant scattering cross sections, the fraction F of a primary isotope that scatters from a magnetic spectrometer into the defining slits can be described [Kilius et al., 1990] by

$$F \propto N\sigma \left(\frac{M}{\Delta M} \right)^{n-1} \Delta x \quad (2.7)$$

where σ is the scattering cross section, N the gas density measured in atoms per volume, Δx the slit width, and n depends on the scattering process. Considering the different cross sections, it turns out that charge change is much more relevant than elastic scattering. For $^{129}\text{I}^{6+}$, [Kilius et al., 1990] estimate that $F \leq 4.3 \cdot 10^{-5}$ of all $^{127}\text{I}^{6+}$ ions injected into their magnet will pass their analyzer magnet due to this effect. Since a “wrong” ion has to change its charge anew within every additional spectrometer, this background can be reduced by adding more filters.

Chapter 3

A short description of VERA

3.1 Ion Source

VERA has a MCSNICS (Multi Cathode Source for Negative Ions by Cesium Sputtering) ion source (see [Norton 1992], built by NEC) with a sample wheel for 40 targets. The diameter of the sputter target is 1.1 mm. This source has a spherical ionizer for the production of the Cs^+ sputter beam.

3.2 Injector ESA

A 45° spherical electrostatic analyzer with a radius of 300 mm and a gap of 50 mm removes sputter tails and provides symmetrical 1:1 focusing from the slits after the source to the entrance lens of the beam sequencer.

3.3 Sequential Isotope Injection

VERA is designed for fast sequential injection of isotopes. This is achieved by applying different voltages to the insulated vacuum chamber of the injection magnet, which leads to the same magnetic rigidity for different isotopes. Although it takes only 20 μs to switch the voltage, stable beam current conditions are reached after about 200 μs . For a complete isotope cycle the voltage is set to 12 kV for 0.5 ms, to 6 kV for 3 ms, and to 1 kV for 100 ms. We use high-voltage TREK P0705 and TREK 50/750 power supplies for this purpose. Fast sequencing is hardware-implemented for the injector magnet chamber and for one pair of vertical and horizontal steerer plates before the accelerator entrance. It is used for measurements of ^{14}C and ^{10}Be . The hardware sequencer proceeds through several *states* in which different ions are injected. For carbon, the voltage applied to the chamber is about 12 kV to measure the $^{12}\text{C}^{3+}$, 6 kV for $^{13}\text{C}^{3+}$, and 1 kV for several 100 ms

of $^{14}\text{C}^{3+}$ counting. We call a complete pass through all three states one *cycle*. The data acquired for one cycle is one measurement of every beam current and a set of counts for the trace isotope.

3.4 Injector Magnet

The injector magnet (Walker) is a double focusing 90° sector magnet with a nominal radius of 18 inch and a (rather large) gap of 1.9 inch. The maximum (measured) field is 1.295 T at a current of 125 A (Danfysik Series 8000 power supply). After installation and first tests [Priller et al., 1997], we found that the shim angles of the injection magnet do not provide proper focusing of the beam at the image slits. NEC added a magnetic quadrupole doublet just in front of the injection magnet to correct for this deficiency. After the installation the measured horizontal width of the beam at the image slits was 5.5 mm, giving a mass resolution of about 80. Therefore, the corrections were sufficient for light ions only. Chapter 6 of this thesis describes our efforts to solve this problem.

3.5 Tandem Accelerator

The 3-MV tandem accelerator is of the Pelletron type (model 9SDH-2). Two charging chains can supply a total charging current of about $230\ \mu\text{A}$. The terminal voltage is measured using a generating volt meter, and regulated by means of a corona probe. Resistors divide the terminal voltage along the accelerator tubes. SF_6 at about 6 bar is used as insulating gas which is continuously recirculated and dried. The temperature of the recirculated SF_6 is regulated by a PID controller adjusting the cooling water flow through a heat exchanger. Before opening the accelerator tank (e.g. for maintenance), the SF_6 is transferred to a liquid storage system (DILO, Germany). The maintenance of the accelerator structure can be conveniently performed by rotating the tank clockwise around a pivot point near the low energy entrance, and pulling the whole accelerator structure out of the tank. A special feature is a supply line from ground to the stripper gas bottle in the terminal. It allows to refill the stripper gas bottle without opening the tank. Up to now we strip with argon from the bottle in the terminal, while the supply line is filled with 10 bar of N_2 . Two turbo pumps at the accelerator terminal accomplish differential pumping at the end of the stripper gas tubes, and the pumped-off stripper gas is recirculated. The pressure in the middle of the stripper tube is measured with a thermocouple gauge. This signal and various other parameters are transferred by a light link system from and to the terminal electronics. A foil changer allows to use stripper foils instead of gas. AC power for the terminal is supplied by a generator driven by a lucite rod connected to a motor at ground potential.

3.6 Analyzer magnet

The analyzer magnet (Danfysik) is a double focusing 90° sector magnet with a nominal radius of 1270 mm and a gap of 34 mm. The maximum (measured) field is 1.53 T at 320 A (Danfysik Series 8000 power supply). It provides bending power up to the heaviest radioisotopes, e.g. $^{244}\text{Pu}^{5+}$ ions at 3 MV terminal voltage.

3.7 Wien Filter

VERA has a relatively small Wien filter with pole length of 305 mm. The distance of the electric plates is 16.2 mm, the magnet gap is 55.4 mm. The maximum magnetic field is 0.44 T at 50 A, which is the limitation for heavy ions. For light ions, the resolution is limited by the ± 30 kV at the electrical plates.

3.8 Vacuum, power, and cooling

The base pressure in the beam line system is kept in the 10^{-9} Torr range by an all cryo pump system (seven CTI pumps). Only oil free roughing pumps are used to avoid contamination with pump oil. A maximum electric power of 100 kW is available for the operation of VERA. The water and air cooling system is designed to provide a maximum cooling power of 70 kW (but actually provides only 50 kW). The high power end is needed for AMS experiments with very heavy radioisotopes. During ^{14}C operation, 30 to 40 kW are consumed.