ACCELERATOR MASS SPECTROMETRY AT VERA

Walter Kutschera
Institut für Isotopenforschung und Kernphysik der Universität Wien,
Währinger Strasse 17, A-1090 Wien, Austria

Abstract

The Vienna Environmental Research Accelerator (VERA) is a dedicated facility for accelerator mass spectrometry (AMS), operated at the University of Vienna since 1996. The principle of AMS with VERA and the advantage of measuring long-lived radioisotopes by atom counting (AMS) rather than decay counting (ß decay) is described. A brief overview on AMS applications is also presented.

1 INTRODUCTION

Mass Spectrometry (MS) is a well-known method to determine the abundance of stable isotopes. Typically, an ion beam with a well-defined energy in the keV range is produced from some sample material, and is sent through a magnet where the ions are separated according to their mass. MS is widely used in research and industry, and isotope ratios of stable isotopes can be measured with high precision. This method, however, has not enough selectivity to separate the extremely low abundances of long-lived radioisotopes in nature (10^{-12} to 10^{-16}) from an overwhelming background. Such a selectivity can be reached by using an entire accelerator system common in nuclear physics as a mass spectromter, thus extending MS to AMS (accelerator mass spectrometry). AMS allows one to measure long-lived radioisotopes by counting atoms rather than decays, which increases the detection sensitivity by many orders of magnitude. This makes it possible now to utilize a variety of cosmogenic radioisotopes which were virtually not detectable by decay counting.

2 ATOM COUNTING VERSUS DECAY COUNTING

Long-lived radioisotopes can be detected either through their radioactivity (decay counting) or by mass spectrometry (atom countig). In the latter case the radioisotopes are detected before they decay, and for measuring times much shorter than the half-life this method is much more sensitive. In Table 1 the conditions for measuring ^{14}C by decay counting and by AMS at VERA are compared. For this comparison it is assumed that the efficiency of detecting ß-rays from the decay of ^{14}C is 100% (upper limit), whereas the ^{14}C atom counting numbers are taken from actual measurements at VERA. As a practical consequence of the enormous gain in sensitivity, one can perform ^{14}C measurements with AMS requiring 1000 times less sample material (1 mg instead of 1 g), and 100 times less counting time (0.5 h instead of 2 d).

Table 1: ^{14}C detection: beta counting *versus* atom counting

Isotopic abundance of ^{12}C	89.9	%
Isotopic abundance of ^{13}C	1.1	%
Isotopic abundance of ^{14}C in modern (pre-bomb) carbon	1.2x10^{-12}	^{14}C/^{12}C
^{14}C content of 1 mg of modern carbon	6x10^{7}	^{14}C atoms
Half-life of ^{14}C	5730 ± 40	y
Activity of 1 mg of modern carbon	0.8	^{14}C decays/h
Activity of 1 mg of 50,000-year old carbon	17	^{14}C decays/y
Negative ion ^{12}C^{-} output from 1 mg of modern carbon [a)]	30	µA
	1.9x10^{14}	^{12}C^{-} ions/s
Negative ion ^{14}C^{-} output from 1 mg of modern carbon	230	^{14}C^{-} ions/s
Counting rate of ^{14}C in the final AMS detector	115	^{14}C^{3+} ions/s
	4.1x10^{5}	^{14}C^{3+} ions/h
Counting rate of ^{14}C from 1 mg of 50,000-year old carbon	970	^{14}C^{3+} ions/h
Gain in ^{14}C detection sensitivity with AMS	5.1x10^{5}	AMS/ß-counting

[a)] 1 mg of carbon lasts typically about one hour in a cesium beam negative ion sputter source.

2 THE VIENNA ENVIRONMENTAL RESEARCH ACCELERATOR

Figure 1 shows the schematic layout of VERA, a dedicated AMS facility based on a 3-MV Pelletron tandem accelerator [1]. As an example we describe the isotopic analysis of carbon, which starts with the production of negative carbon ions in a cesium beam sputter ion source.

Whereas it is relatively easy to measure the isotope ratio of the two stable carbon isotopes ($^{13}C/^{12}C = 1.1x10^{-2}$) with classical mass spectrometry at energies in the keV range, it is virtually impossible to measure the extremely low abundance of the long-lived radioisotope ^{14}C ($^{14}C/^{12}C = 1.2x10^{-12}$) in this way. After selecting ions with mass 14 by the first analysing magnet, the $^{14}C^-$ ions are burried under an enormous background of molecular ions such as $^{13}CH^-$, $^{12}CH_2^-$ (see fig.2a). Fortunately, $^{14}N^-$ ions are absent in the mass-14 peak, because nitrogen does not form stable neagative ions and thus gets completely suppressed already in the ion source. An important step in the consecutive acceleration in the tandem is therefore the stripping process in the terminal, which dissociates the $^{13}CH^-$ and $^{12}CH_2^-$ molecules very effectively when sufficient electrons are stripped off. With the high energy analysing magnet set to select $^{14}C^{3+}$ ions, one observes the energy spectrum shown in Figure 2b. Although the residual ^{12}C and ^{13}C peaks are greatly reduced in intensity, there is still a large number of background peaks which happen to have the same magnetic rigidity as $^{14}C^{3+}$. However, Figure 2c shows that the $^{14}C^{3+}$ ions can be cleanly selected by sending the mix of ions in figure 2b through a Wien filter (see fig. 1) set to the velocity of $^{14}C^{3+}$. In order to determine $^{14}C/^{12}C$ ratios with high precision (~0.5%), $^{12}C^-$, $^{13}C^-$, and $^{14}C^-$ are injected in fast sequence (10 times per second) into the tandem. The stable isotopes, $^{12}C^{3+}$ and $^{13}C^{3+}$, are measured via their respective ion currents in Faraday cups located at the exit of the high-energy analysing magnet, and $^{14}C^{3+}$ ions are counted in a Si surface barrier detector at the end of the beam line (see fig. 1). For details of $^{14}C/^{12}C$ ratio measurements and of other radionuclides at VERA the reader is referred to references [3-8]. We now operate VERA fully automated and unattended during nights and over weekends, with the person in charge monitoring (and controlling) the performance from home via internet [9].

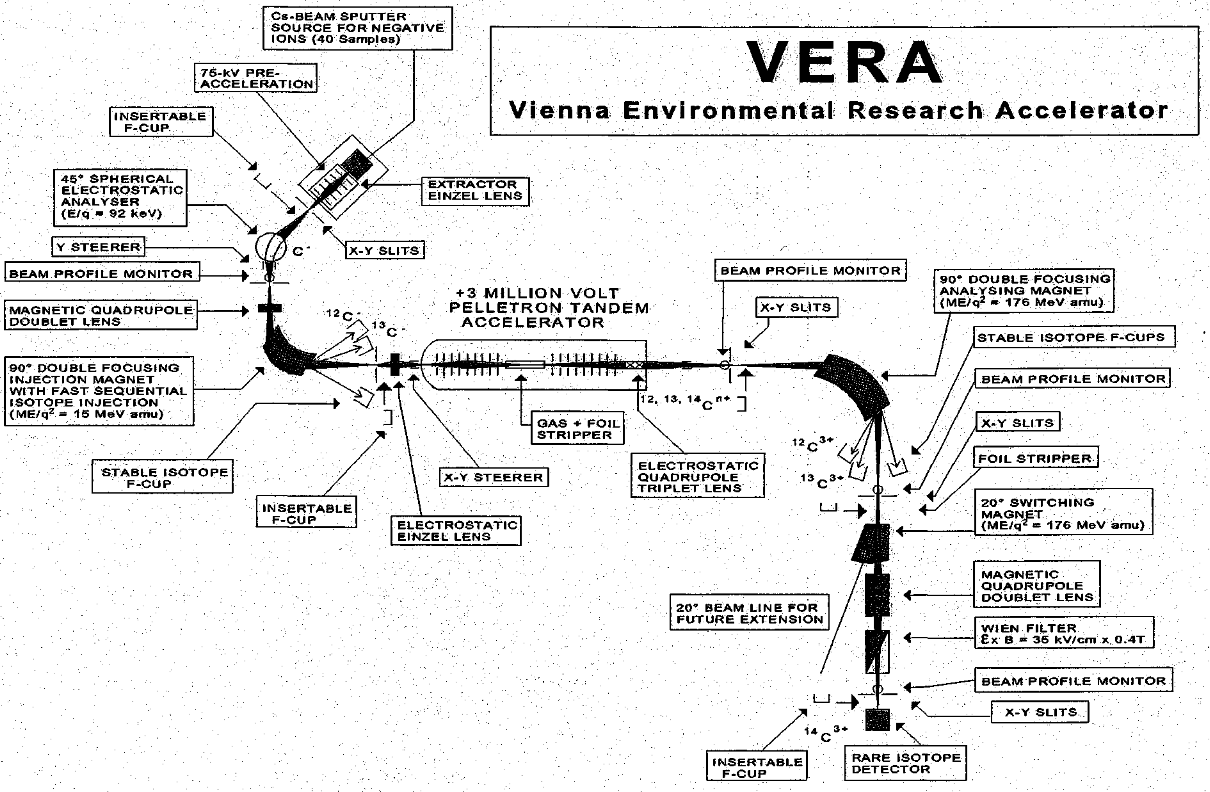

Figure1: Schematic layout of VERA showing the essential features of the AMS system. ^{14}C measurements are typically performed at a terminal voltage of 2.7 MV.

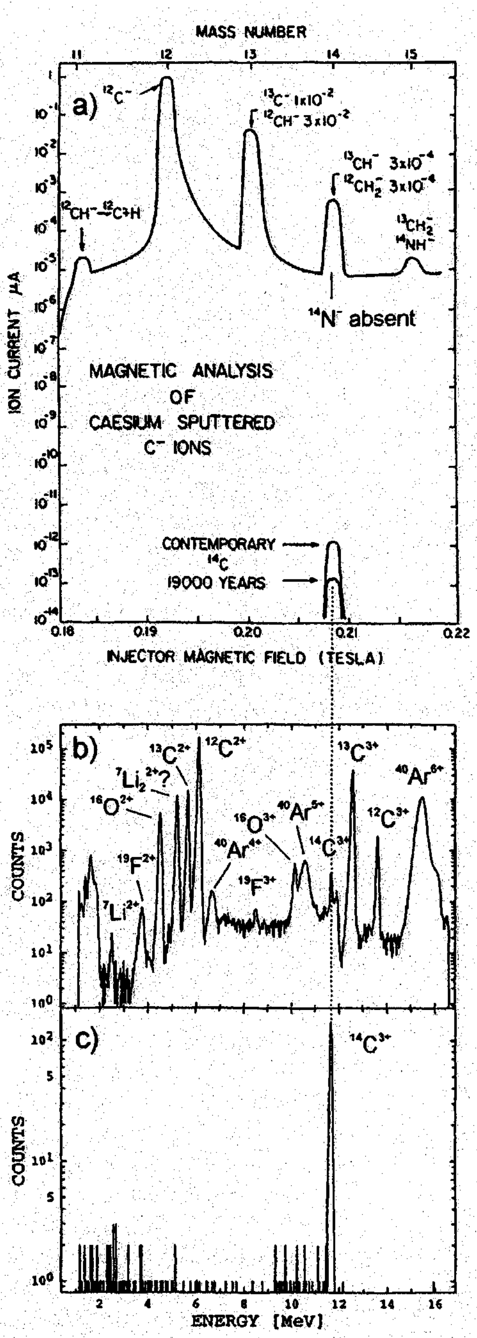

Figure 2: The three steps in the detection of ^{14}C with AMS at VERA. a) In the negative ion mass spectrum after the ion source the ^{14}C signal is burried under an enourmous background of mass-14 molecules [2]. Most important, however, is the absence of ^{14}N because nitrogen does not form negative ions. b) After acceleration through the tandem and analysis in the high-energy magnet the energy spectrum measured in the rare isotope detector (Si surface barrier detector) still shows many background peaks.This spectrum is measured with the velocity filter (Wien filter) switched off. c) After final analysis through theWien filter, a clean ^{14}C signal emerges.

3 SMALL IS BEAUTIFUL: THE MINATURIZATION OF AMS FACILITIES

For twenty years it was believed that $^{14}C^{3+}$ ions (or higher charge states) must be produced in the stripping process to break up the molecules for sure. It is interesting to note that this "dogma" of stripping has now been proven to be obsolete. In a collaborative effort of National Electrostatics Corporation (NEC) and the AMS laboratory of the ETH/PSI Zurich it was demonstrated that high quality ^{14}C AMS measurements can be performed using a 0.5 MV Pelletron tandem and stripping only to ^{14}C ions in charge state 1+ [10, 11]. The most surprising result was the measurement of $^{14}C/^{12}C$ ratios down to a level that corresponds to a radiocarbon age of 48,000 years. It therefore looks feasible to perform ^{14}C dating measurements with much smaller tandem accelerators than presently in use, approaching essentially the size of table top machines. Such small machines may prove particularly useful for biomedical application with labelled compounds, where generally less precision is required, but ease of operation, sample throughput, and space requirements are important factors.

4 APPLICATIONS OF AMS IN THE ENVIRONMENT AT LARGE

Applications of AMS are numerous, and penetrate almost every field of our environment on earth and beyond. The reason is that every piece of matter on Earth and beyond (meteorites, Moon, etc.) contains a certain number of long-lived radionuclides produced mainly by cosmic ray interaction, so-called cosmogenic radioisotopes. Other sources are man-made radioisotopes (anthropogenic) and those which originate from the decay of very long-lived radionuclides such as ^{238}U (radiogenic). AMS makes it possible to measure an increasing number of radioisotopes in the various domains of our environment (Table 2). Among the radioisotopes listed in Table 2, ^{14}C is still by far the most-used one. Approximately 90% of all AMS measurements performed in 50 AMS laboratories worldwide [12] are applications of ^{14}C. This is not surprizing since prior to the invention of AMS in 1977 there were already 30 years of ^{14}C applications through the classical ß-counting technique. Most other radioisotopes shown in Table 2 only became available for applications through AMS. It is not possible in this brief report to discuss any of the applications in detail. For this, the reader is referred to a recent, more extended review paper by the author [12], and to the Proceedings of recent AMS Conferences [13, 14]. Some of the work performed at VERA is presented in references [3-9]. For a more general source of information on both the technique and application of AMS, the reader is referred to a recent monograph by Tuniz et al. [15].

Table 2: Overview of AMS applications in the environment at large

Sphere	**Areas of interest. Radionuclides used in the respective fields are given in parenthesis.*)**
Atmosphere	Production and distribution of cosmogenic and anthropogenic radionuclides (^{3}H, ^{7}Be, ^{10}Be, ^{14}C, ^{26}Al, ^{32}Si, ^{36}Cl, ^{39}Ar, ^{81}Kr, ^{85}Kr, ^{129}I) Study of trace gases: CO_2, CO, OH, O_3, CH_4 (^{7}Be, ^{10}Be, ^{14}C) Transport and origin of aerosols (^{14}C)
Biosphere	Dating in archaeology and other fields (^{14}C, ^{41}Ca) ^{14}C calibration studies in tree rings, corals and sediments (^{14}C) Studies in forensic medicine through bomb-peak dating (^{14}C) In-vivo tracer studies in animals and humans (^{3}H, ^{14}C, ^{26}Al, ^{41}Ca, ^{79}Se, ^{129}I)
Hydrosphere	Dating of groundwater (^{14}C, ^{36}Cl, ^{39}Ar, ^{81}Kr, ^{85}Kr, ^{129}I) Global ocean circulation pattern (^{14}C, ^{39}Ar, ^{129}I) Paleoclimatic studies in ocean sediments
Cryosphere	Ice core analysis from Greenland, Antarcita, and other glacial areas on earth Paleoclimatic studies (^{10}Be, ^{14}C, ^{32}Si, ^{36}Cl, ^{39}Ar, ^{81}Kr) Variation of cosmic ray intensity with time (^{10}Be, ^{36}Cl) Bomb-peak identification (^{36}Cl, ^{41}Ca, ^{129}I)
Lithosphere	Exposure dating and erosion of surface rocks (^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl) Paleoclimatic studies in loess (^{10}Be) Tectonic plate subduction studies through volcanos (^{10}Be) Platinum group elements in minerals (stable trace isotopes)
Cosmosphere	Cosmic ray record in meteorites and lunar materials (^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{44}Ti, ^{59}Ni, ^{60}Fe, ^{107}Pd, ^{129}I); life on Mars ? (^{14}C) Evidence for supernova occurrence through extinct and life radionuclides in meteorites and manganese crusts (^{10}Be, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{60}Fe, ^{107}Pd, ^{135}Cs, ^{146}Sm, ^{182}Hf, ^{210m}Bi, ^{244}Pu, ^{257}Cm) Geochemical solar neutrino detection (^{41}Ca, ^{81}Kr, ^{98}Tc, ^{126}I (^{126}Xe), ^{205}Pb) Search for exotic particles (superheavy elements, fractionally charged particles, strange matter)
Technosphere	Releases from nuclear industry (^{14}C, ^{36}Cl, ^{85}Kr, ^{90}Sr, ^{99}Tc, ^{126}Sn, ^{129}I) Temperature measurement of fusion plasma (^{26}Al) Neutron flux of the Hiroshima bomb (^{36}Cl, ^{41}Ca, ^{63}Ni) Fission material characterization (^{236}U, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{242}Pu, ^{244}Pu) Ultra-high purity tests of semiconductor materials (stable trace isotopes)

*) The half-lives of radionuclides are as follows (the most used radionuclides are marked in bold): ^{3}H ($t_{1/2}$ = 12.33 y), **^{10}Be (1.5x10^{6} y), ^{14}C (5.73x10^{3} y), ^{26}Al (7.1x10^{5} y),** ^{32}Si (135 y), ^{32}Si (135 y) **^{36}Cl (3.01x10^{5} y),** ^{39}Ar (268 y), **^{41}Ca (1.04x10^{5} y),** ^{44}Ti (60.0 y), ^{53}Mn (3.7x10^{6} y), ^{55}Fe (2.73 y), ^{60}Fe (1.5x10^{6} y), ^{59}Ni (9.2x10^{4} y), ^{63}Ni (100 y), ^{79}Se (1.1x10^{6} y), ^{81}Kr (2.3x10^{5} y), ^{85}Kr (10.8 y) ^{90}Sr (28.8 y), ^{99}Tc (2.11x10^{5} y), ^{107}Pd (6.5x10^{6} y), ^{126}Sn (2.35x10^{5} y), ^{126}I (13.1 d), ^{126}Xe (stable), **^{129}I (1.7x10^{7} y),** ^{135}Cs (2.3x10^{6} y), ^{146}Sm (1.03x10^{8} y), ^{182}Hf (9x10^{6} y), ^{205}Pb (1.5x10^{7} y), ^{210}Pb (22.3 y), ^{210m}Bi (3.04x10^{6} y) ^{236}U (2.34x10^{7} y), ^{237}Np (2.14x10^{6} y), ^{239}Pu (2.41x10^{4} y), ^{240}Pu (6.54x10^{3} y), ^{242}Pu (3.73x10^{5} y), ^{244}Pu (8.1x10^{7} y), ^{247}Cm (1.56x10^{7} y).

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