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THE ETH/SIN DATING FACILITY: A STATUS REPORT

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ABSTRACT. The ETH/SIN AMS dating facility is routinely used for ^{10}Be, ^{14}C, ^{26}Al, and ^{36}Cl measurements. The present status and performance of this facility are reviewed and some of its major applications summarized.

INTRODUCTION

Accelerator mass spectrometry (AMS) has become a powerful tool for the detection of the cosmogenic long-lived radioisotopes such as ^{10}Be, ^{14}C, ^{26}Al, and ^{36}Cl, which occur in concentrations of $10^{-12}-10^{-16}$ relative to the stable isotopes. This technique opens up many new applications in different fields of science, where these radioisotopes are used for dating or as tracers. Progress in this field and its applications has been discussed most recently during the Third International Symposium on Accelerator Mass Spectrometry held at the ETH Zurich, April 10–13, 1984.

At the ETH Zurich the existing 6 MV Tandem Van-de-Graaff accelerator has been transformed into a high precision and fully automated dating facility. Presently it is routinely used for ^{10}Be and ^{14}C, ^{26}Al, and ^{36}Cl measurements. A total of 750 ^{10}Be, 568 ^{14}C, 120 ^{26}Al, and 150 ^{36}Cl samples (unknown, standards, and blanks) were analyzed in 1984. Measurements of >4000 samples have been requested for 1985. The present capacity is of 50 ^{10}Be, 25 ^{14}C, 25 ^{26}Al, and 25 ^{36}Cl samples per day. The measurements of such large series of samples with high precision requires, as will be discussed here, a carefully designed layout, an efficient data acquisition system, as well as some additional improvements which guarantee ease of operation, long-term stability, and precise determination of the isotopic ratios independent of current and transmission variations.

LAYOUT

The experimental arrangement used for ^{14}C measurements is shown in Figure 1. The ion source was installed on a separate 90° entrance port of the inflection magnet. The analyzing system on the high energy side is on a separate beam line, so that most of the components used for mass spectrometry are independent of other experiments performed with the same accelerator. Except for the injection and the final analyzing magnet at the high energy end, only electrostatic steering and focusing elements are used to transport the beam.

The negative ions are produced by a Cs-sputter source, specially designed for low contamination between specimens (cross talk) and convenient loading of samples. A cassette containing up to 25 samples can be loaded during full operation of the source. All samples within a cassette can be exchanged by computer control. The ions extracted at energies of 30–

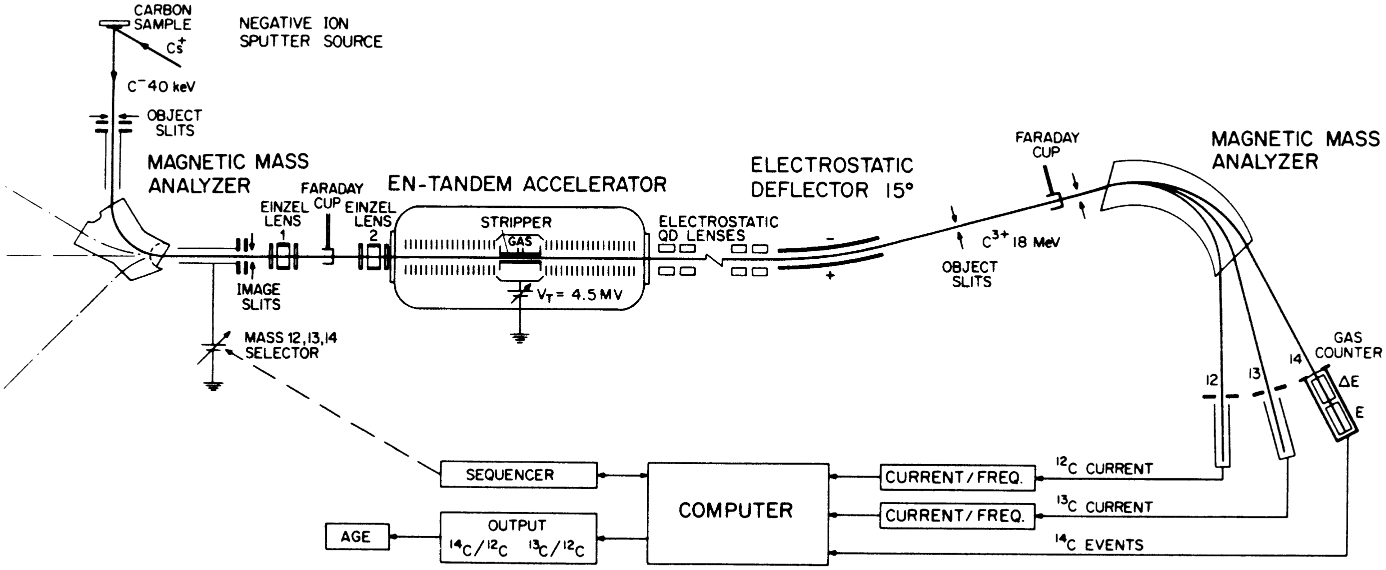

Fig 1. Schematic layout of the ETH/SIN dating facility. Only those parts relevant for AMS are shown.

50 kV are focused with a first einzel lens onto the object slits of the 90° inflection magnet. The chamber of this magnet is insulated so that a high voltage can be applied to it. Acceleration and deceleration gaps are located symmetrically to the magnet and are positioned close to the object and image slits. Pulses of up to 10kV can be applied to this magnet chamber to change the energy of the ions entering the magnet. This allows rapid switching from the measurement of one isotope to another leaving the magnetic field unchanged. A Faraday cup is placed behind the inflection magnet in such a way that the ^{12}C beam can be monitored while ^{14}C is injected into the accelerator. Two einzel lenses and electrostatic steerers are used to direct the beam in the accelerator. Two electric quadrupole lenses are used to focus the beam at the high energy side. A cylindrical electrostatic deflector selects the ions with the desired energy and charge state. The ions are then analyzed by a 90° magnetic spectrometer with a resolution of $p/\Delta p \simeq 300$. Several apertures had to be mounted in front of as well as behind the magnetic spectrometer to eliminate scattered particles.

The Faraday cups and the ΔE-E counter are mounted in such a way that they can be rearranged easily for measurements of the isotopic composition of other elements of interest. For the detection of ^{36}Cl an additional time of flight measuring system placed in front of the ΔE/E gas counter telescope is used.

FAST ISOTOPE SWITCHING SYSTEM

For a ^{14}C measurement, the isotopic ratios ^{14}C/^{12}C and ^{13}C/^{12}C of the unknown samples have to be measured with a precision of at least 1% relative to that of a standard (Suter *et al,* 1984a, p 117). This can only be achieved if the rare as well as the abundant stable isotopes are accelerated and measured at the very end of the facility. Most of the existing accelerators, however, in particular, electrostatic tandem accelerators, are neither designed to handle large beam intensity variations occurring when switching from the rare to the abundant isotope, nor capable of sustaining intense heavy ion beams continuously. Therefore, the intense beams from the ion source have to be attenuated or injected in very short pulses only, so that loading effects of the accelerators are insignificant.

As indicated in Figure 1, we are using a fast switching system, which can inject the stable isotopes in short pulses of 20–200μs duration with a repetition rate of 10–30Hz. This pulsing method (Suter *et al,* 1984b, p 242) requires a medium power pulse source delivering rectangular pulses with an amplitude of up to 10kV to a load of 11nF the capacity of the insulated deflection chamber of the analyzing magnet. Details of this pulse source have been published (Balzer, 1984, p 247).

In order to measure the beams of the stable isotopes during the stable part of the applied high voltage pulses, current integraters with start/stop, hold, and reset features have to be used. A quartz-controlled clock determines the time sequence and produces all signals required for the pulsed power supply, the current integrators and the data acquisition for the rare isotopes (Suter *et al,* 1984b, p 242).

AUTOMATIC MEASURING PROCEDURE

The measurement of a whole series of samples is automated in the following way: When the samples have been loaded into the ion source box, a list containing the identification of the samples is entered into the data acquisition system based on a LSI 11/23 computer. A run schedule is then defined. For each run, the sample position in the cassette, the maximum run time, and the desired statistical accuracy are given. The samples can be measured in any order, and each more than once. The run schedule can be modified and extended even during a measuring sequence. A run is divided into intervals of 10 to 50 seconds' duration. For each interval, the ^{12}C current, measured at the low energy side during the ^{14}C injection time, the ^{12}C and ^{13}C currents in the final Faraday cups and the number of ^{14}C events registered in a predetermined ΔE and E window, are stored. From these numbers, the isotopic ratios are derived and the transmission for the ^{12}C beam is determined. In addition the statistical error of the ^{14}C counts is calculated and mean values and standard deviations are determined, when more than one interval is measured. These results are continuously displayed. At the end of each run, a complete list of the results is printed and, together with the ΔE-E spectrum, stored on a disk.

The schematic layout of our data acquisition system is shown in Figure 2. The same computer is used to control the operation of the accelerator, the power supplies for the magnetic analyzer and the electrostatic beam focus and transport system. All ADC's and power supplies are interfaced to a CAMAC crate containing the microprocessor (Nessi *et al,* 1984, p 238).

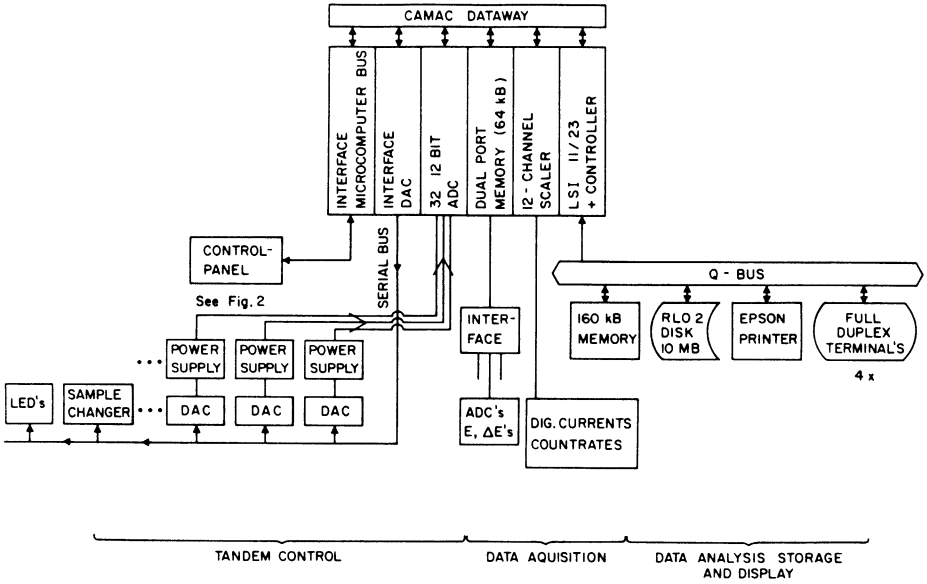

Fig 2. Layout of the computer control and data acquisition system. All power supplies of the facility and the ADC's are interfaced to a CAMAC crate containing the microprocessor.

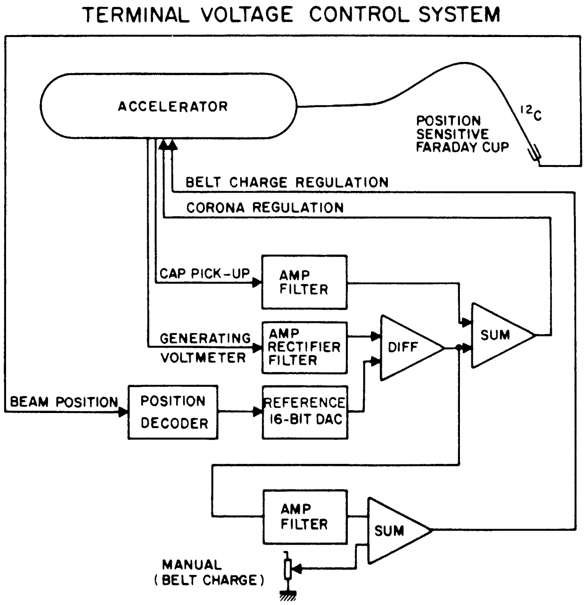

Fig 3. Terminal voltage control system of the ETH EN Tandem accelerator.

TANDEM VOLTAGE CONTROL SYSTEM

Precise isotope ratio measurements require well-stabilized ion beam energies. Usually the terminal voltage of a tandem accelerator is controlled by a signal derived from slits placed on both sides of the beam in the image plane of the analyzing magnet. This procedure cannot be applied in AMS, because no beam losses at slits can be tolerated. In addition, the beams are pulsed and a proper beam position signal can be derived only during the short injection time of ^{12}C. Therefore, the generating voltmeter regulation mode has to be used. In order to eliminate the short- and long-term drifts observed in the pulsed mode, a new position-sensitive control system shown in Figure 3 has been designed (Suter *et al,* 1983, p 1528). It consists of a conventional corona regulation loop, which derives the correction signals from the GVM (low frequencies) and from a capacitive pick-up electrode (high frequencies). The beam position is determined by measuring the x-ray intensity distribution produced by the heavy ion beam (eg, ^{12}C) stopped at the end of the Faraday cup. The X-rays are detected by a microchannel plate detector provided with a split anode. The difference of the two anode signals is proportional to the beam position and this information is used to adjust the reference voltage of the generating voltmeter feedback loop. The stability achieved with this simple design guarantees stable operation for many days (Suter *et al,* 1984c, p 251).

PERFORMANCE

The facility has been in routine use for the last two years for ^{10}Be and ^{14}C measurements and first measurements using ^{26}Al and ^{36}Cl samples have been made. Beam current, measuring time, detection limit, and the precision of an isotopic ratio measured relative to that of a standard strongly depends on the isotope, the origin and compound of the sample material, and the sample preparation procedure. The relevant numbers of our facility are given in Table 1.

BERYLLIUM 10

^{10}Be forms metastable negative ions only (Bunge, Galan & Jauregui, 1982, p 299). Therefore, negatively charged molecular BeO ions have to be extracted from the ion source and injected into the accelerator. Using milligram samples of BeO mixed with copper powder, reasonable BeO beam currents of 1–2μA are obtained. The molecular beam is stripped and dissociated in the terminal of the accelerator using a 10 to 15μg/cm^2 carbon foil. Multiple scattering in this foil and the Coulomb explosion of the molecules are responsible for a rather low beam quality at the high energy side (Stoller *et al,* 1983, p 1074). Consequently, the systematic errors in determining ^{10}Be/^{9}Be ratios are rather large, namely of the order of 2–3%. Usually BeO samples are contaminated with boron on the 0.1 to 1ppm level. Since boronoxyde readily forms negative ions, it is necessary to eliminate the isobar ^{10}B by means of a gas absorber cell placed in front of the gas counter ΔE/E telescope. Nuclear reactions of ^{10}B with traces of hydrogen in the absorber gas or on the surface of the entrance window produce ^{7}Be which may enter the telescope. These particles produce a background which partly overlap with the ^{10}Be region. However, our present detection limit of 10^{-14} is determined not by this effect, but by true ^{10}Be atoms occurring in all blanks produced from commercially available material. Lower blank values have been found so far only in BeO samples which have been extracted from a beryl crystal found in a deep mine (Middleton *et al,* 1984, p 511).

CARBON 14

Graphite, charcoal, or thin deposits of carbon reduced from CO_2 and $CaCO_3$ deliver typical beam currents between 10 and 30μA. Depending on the origin of the material, different preparation techniques are used. The properties of the various methods are described elsewhere (Bonani *et al,* 1984, p 284). In contrast to the ^{10}Be case, ^{14}C/^{12}C ratios can be measured

TABLE 1
Status of the ETH/SIN dating facility

Sample	Negative ion current [μA]	Terminal voltage [MV] (charge state)	Total efficiency [%]	Background	Systematic error [%]	Statistical* error [%]
BeO	1–2	5.2 (3+)	10^{-3}	10^{-14}	3	2
C	10–40	4.5 (3+)	$2.5 \cdot (10^{-3}–10^{-2})$	$5 \cdot 10^{-16}$	0.1–0.2	0.3–1.0
Al_2O_3	0.2–0.4	6.0 (7+)	$0.7 \cdot 10^{-4}$	10^{-14}	1–2	8
AgCl**	10–20	6.0 (7+)	$3.6 \cdot 10^{-3}$	$5 \cdot 10^{-14}$	10	1

* Statistical error for a concentration of 10^{-12} and 1h measuring time

** Sulphur contamination $\leq$ 1ppm

with much higher precision. Extensive tests showed (Suter *et al,* 1984a, p 117) that, in this particular case, the systematic error of a $^{14}C/^{12}C$ and $^{13}C/^{12}C$ ratio measurement relative to that of a standard can routinely be kept on the 1–2‰ level, if thin deposits of carbon (<0.1mm) are used. This means that the total error of a ^{14}C concentration measurement is usually determined by statistics only. Our present precision allows us also to determine the $\delta^{13}C$ for each sample during the same measuring procedure. The fact that the systematic errors are much smaller than the statistical ones is illustrated in Figure 4. It shows the distribution of a total of 515 independent $^{14}C/^{12}C$ ratio measurements using >200 different samples of ages between modern and 60,000 years. It agrees well with a chi-squared distribution. Larger systematic errors of the order of 1–2% are observed if thick samples are used. Phase space changes in the ion source due to crater formation during the sputter process are most probably responsible for this increase.

The dating range for ^{14}C is usually determined by the contamination of the carbon samples with modern CO_2 during the preparation procedure. The blank value depends on the preparation method and the available sample amount. For milligram dead carbon samples, the ^{14}C contamination varies between 2.5 and 0.1% of modern. The lowest blank values have been observed in spectroscopically pure graphite, untreated graphite from the bedrock of Finland and from the El Taco meteorite. These results are compiled in Table 2. By using an additional time-of-flight spectrometer placed in front of the counter telescope it has been verified that in all three cases the blank value is determined by true ^{14}C events. The origin of these events,

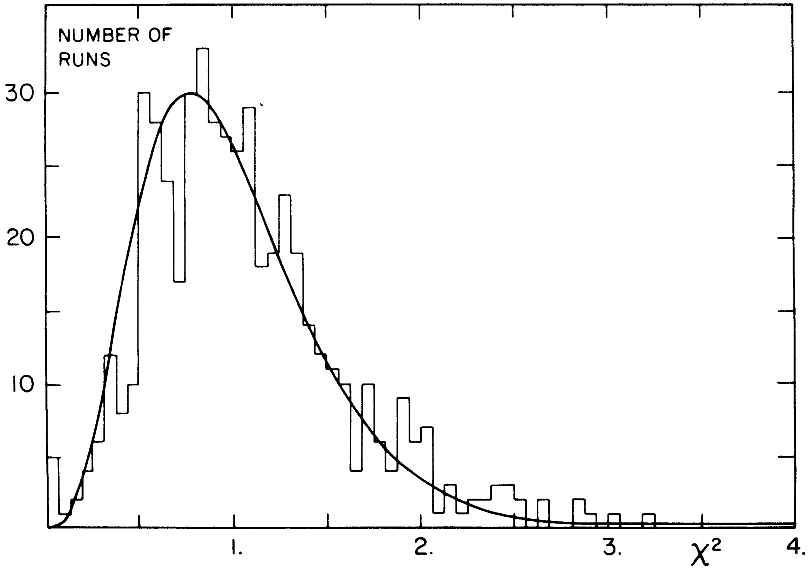

Fig 4. Experimental and theoretical chi-squared distribution of ^{14}C to ^{12}C ratio from 515 independent ^{14}C measurements of samples with ages between modern and 60,000 years.

TABLE 2

Apparent ^{14}C ages of chemically untreated graphite of different sources

Source of graphite	$^{14}C/^{12}C$	Apparent ^{14}C age
Spectroscopically pure	$7.4 \cdot 10^{-16}$	59,500 ± 1000
El Taco meteorite	$1.1 \cdot 10^{-15}$	56,500 ± 1500
Bedrock of Finland	$4.5 \cdot 10^{-16}$	63,500 ± 2000

which restrict the AMS ^{14}C dating range to ca 60,000 years is not yet understood. Trivial surface contamination with modern CO_2 can be excluded. No significant decrease in the count rate was observed during the many hours of measuring time.

ALUMINIUM 26

Al can easily be detected because ^{26}Mg, the isobar, does not form stable negative ions. However, the negative ion efficiency for Al is very low. Using Al_2O_3 samples mixed with silver powder yields negative ion beam currents of only ca $0.2\mu A$ for ^{27}Al. Higher beam currents can be achieved only with a different type of ion source (Middleton, 1984, p 193). Our detection limit for ^{26}Al is 10^{-14}.

CHLORINE 36

Very high ^{35}Cl beam currents of the order of $10–20\mu A$ are obtained from AgCl samples. As the mass resolution of the magnetic analyzers decreases with increasing mass, an additional time-of-flight spectrometer placed in front of the counter telescope is of importance here. It has a flight path of 2m and a time resolution of 200 ps for ^{36}S. This resolution is sufficient to discriminate between mass 35, 36, and 37 particles. However, the major problem is the isobaric separation. Sulfur readily forms negative ions and occurs in the AgCl samples as well as in the sample holders. Lowest sulfur concentrations at the ppm level has been found in high purity Ta (99.98%) and Au. Although the natural abundance of ^{36}S is $1.4 \cdot 10^{-4}$ only, it produces a very high background which can only be reduced electronically by measuring the energy losses of the two isobars in a multi-stage counter telescope. With a four-stage counter telescope a suppression factor of 10^3 has been achieved at our maximum particle energy of 48 MeV. This rather low suppression rate limits our detection sensitivity to ca $5 \cdot 10^{-14}$ provided the sulfur contamination of the sample and the sample holder can be kept below 1ppm. Higher beam energies, eg, larger accelerators (Conard *et al,* 1986) or sulphur-free samples and holders are required to improve this limit.

APPLICATIONS

Presently, about half of the available time is devoted to basic research in heavy ion physics relevant for the continuous improvement and development of AMS. Of particular interest here are the formation of negatively charged atomic and molecular beams, the dissociation or Coulomb explosion of high energetic molecular beams in gas and foils, and the charge state distribution of atomic heavy ions and molecular fragments after the stripping and dissociation process (Hofmann *et al,* 1984, p 254). The half-life of ^{10}Be (t1/2 ~ 1.5 10^6 y) is known with an accuracy of 15% only. An attempt is made to improve this accuracy.

In a number of national and international collaborations, ca 1000 hours of measuring time was spent in 1984 for applications in archaeology, geoscience, and cosmology. A few results of this research work was presented at this conference (Andrée *et al,* 1986a,b; Haas *et al,* 1985; Johnson *et al,* 1986).

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