



ELSEVIER

Nuclear Instruments and Methods in Physics Research B 172 (2000) 1–7

NIM B
Beam Interactions
with Materials & Atoms

www.elsevier.nl/locate/nimb

The PSI/ETH small radiocarbon dating system

H.-A. Synal^{a,*}, S. Jacob^b, M. Suter^b

^a Paul Scherrer Institute, clo Institute für Teilchenphysik, F-13, ETH Hönggerberg, Building HPK, Zürich CH-8093, Switzerland

^b ETH Hönggerberg, Building HPK, Zürich CH-8093, Switzerland

Abstract

A small and compact radiocarbon dating system has been built at PSI/ETH. The system is based on a National Electrostatics Corporation (NEC) pelletron accelerator with a maximum terminal voltage of 550 kV. It is the first accelerator mass spectrometry (AMS) system that uses 1^+ ions at the high-energy end of the accelerator. Interfering isobaric molecules are destroyed by collisions in the gas stripper inside the accelerator. The system has been designed to fulfill two primary goals. First, it can be used as an experimental platform to study the relevant charge exchange and molecular break up processes at low energies. Second, it is able to perform high quality radiocarbon dating measurements. A detailed system description is given and results of performance tests are discussed. © 2000 Elsevier Science B.V. All rights reserved.

PACS: 0.7.75.+h; 41.85.p; 82.80.Ms

Keywords: Accelerator mass spectrometry; Radiocarbon dating; Molecular destruction

1. Introduction

State-of-the-art radiocarbon accelerator mass spectrometry (AMS) systems, which are able to provide high precision measurements, are now commercially available [1,2] and several so-called dedicated AMS facilities have been set up during the past few years. These systems are based on relatively large tandem accelerators (2.5 and 5 MV, respectively) for which the investment and operating costs are high and a sophisticated environment is required for their operation. For many

projects, where limited funds may impede a more extensive use of the AMS technique, it is highly desirable to have compact and cost-effective analytic instrumentation available to be operated directly by the users and under operating conditions of a standard laboratory.

In AMS, the high sensitivity is reached by destroying the interfering molecules during the stripping process and usually, ions in charge state 3^+ or higher are used for which small molecules are unstable. Interfering molecules in charge states 1^+ or 2^+ have bound states, but they can also be destroyed in collisions with the stripper gas [3,4]. Recent experiments performed at the AMS facility of the Paul Scherrer Institute and the ETH (PSI/ETH) [5,6] have shown that the molecular

* Corresponding author. Tel.: +41-1-633-2027; fax: +41-1-633-1067.

E-mail address: synal@particle.phys.ethz.ch (H.-A. Synal).

component at mass 14 can be sufficiently suppressed to background levels suitable for radiocarbon dating. Based on these results, a new compact radiocarbon dating system has been designed and set up at PSI/ETH, which can be regarded as an initial step towards a new generation of compact AMS systems.

2. Description of the system

The new instrument was developed at PSI/ETH in collaboration with National Electrostatics Corporation (NEC, [1]). In Fig. 1 the main components of the system, having overall dimensions of 4.5 m × 6.5 m, are shown. The small and compact instrument is based on a 1-SDH pelletron accelerator and operates with ions in charge state

1⁺. The required acceleration voltage can be utilized with short high-gradient acceleration tubes of only 30 cm length, enabling a compact design of the accelerator. On the other hand, the gas density of about 2 μg/cm², which is necessary to achieve sufficient suppression of interfering molecules [6], requires a relatively long terminal stripper. In our system, the overall length of the stripper housing is 76 cm, the stripper tube is 45 cm long and has an inner diameter of 8 mm. The diameter of the last 5 cm of the stripper tube is enlarged to 9 mm to account for the increasing beam size caused by angular straggling. At present, argon gas is used as stripper gas and experiments are planned to find the optimal stripper medium. About 95% of the stripper gas is recirculated with two 150 l/s turbomolecular pumps and two tubes of low conductance (≈0.6 l/s and ≈1.5 l/s, respectively) are

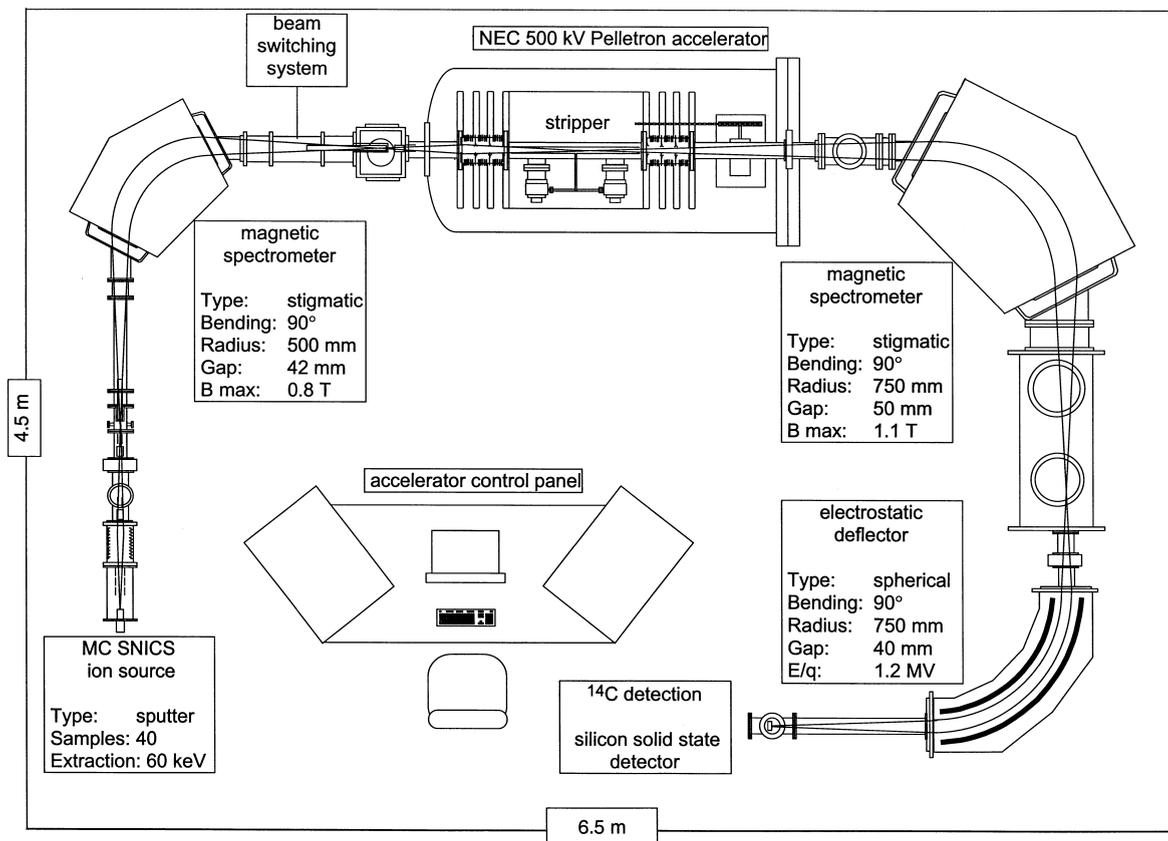


Fig. 1. The PSI/ETH compact AMS system for radiocarbon dating. The system is based on a 1-SDH pelletron accelerator and has been developed in collaboration with National Electrostatics Corporation (NEC).

placed at entrance and exit of the stripper housing to prevent an extensive gas flow into the acceleration sections. At the gas density required for radiocarbon dating measurements, the pressure in the backing line of the terminal pumps is $\approx 6 \times 10^{-2}$ mbar. The pressures measured are $\approx 8 \times 10^{-5}$ mbar in the stripper housing and $\approx 3 \times 10^{-7}$ and $\approx 9 \times 10^{-7}$ mbar at low- and high-energy end of the accelerator, respectively. The higher gas flow into the high-energy tubes is related to a larger opening at the exit of the stripper housing (diameter 11–12 mm compared to 8 mm at the entrance), which is required to account for the angular straggling introduced in the stripping process.

The system is equipped with an MC SNICS caesium sputter ion source [7]. It is mounted on a high voltage deck, which makes ion beam energies up to 60 keV feasible. The ions extracted from the source are mass analyzed in a double focusing spectrometer magnet. The focusing properties of the low-energy acceleration tube of the pelletron are used to form a waist in the center of the stripper channel. No additional lens is used to match the emittance of the ion source to the acceptance of the accelerator. The focusing strength of the low-energy acceleration tube is adjusted by the source potential. Good ion optical matching is achieved at an injection energy that corresponds to about 10% of the terminal voltage. A maximal terminal voltage of 550 kV can be generated with the pelletron accelerator, which allows the coverage of the region with maximal stripping yield for 1^+ ions. At the high-energy end of the accelerator a combination of a magnetic and an electrostatic sector field is used to mass analyze the ion beam. Both devices have stigmatic focusing properties and the same bending radii (0.75 m). The combination of these two elements is made in a way that an achromatic beam transport becomes possible. The analyzing magnet directly follows the accelerator and again the beam is transported without an additional lens. The acceptance of the system is large enough to transport almost all ions of interest to their final detectors and beam losses associated with angular straggling occur primarily inside the stripper channel. In order to keep the dimensions of the system compact, the center of the stripper is imaged with a spatial demagnifica-

tion factor of about two, resulting in a short image distance at the exit of the magnet. The following electrostatic sector field magnifies the intermediate image to almost its original size onto a silicon solid state detector. Here, a measurement of the total energy of the mass filtered ions is made. At the beam energies that can be reached for 1^+ ions (about 1 MeV), ions can be unambiguously detected by means of a total energy measurement, whereas a proper particle identification using the $dE/dx - E$ technique is not possible.

During radiocarbon measurements all beam elements are tuned to mass 14 and the two stable isotopes of carbon (^{12}C ; ^{13}C) are analyzed using the fast beam switching technique [8]. Both isotopes are injected into the accelerator in a fast sequence at a repetition rate of 12.5 Hz. For mass 12 (^{12}C) a beam pulse of 20 μs duration is integrated whereas, for mass 13 (^{13}C & ^{12}CH) the integration time is 200 μs . One off-axis Faraday cup is used to measure the currents of ^{13}C & ^{12}CH and of ^{12}C at the low-energy end of the accelerator. This cup is placed in the focal plane of the low-energy magnet at the position of mass 13 and the total current of the $^{13}\text{C}^-$ and $^{12}\text{CH}^-$ ions is measured during the time of the injection of the mass 14 beam. The ^{12}C current is measured during the mass 13 beam pulse. During this pulse (200 μs), the ^{12}C ions go into the off-axis cup and the beam current is integrated over a time period of 20 μs . From the measurements of both currents, the ^{12}CH molecular component can be calculated by taking into account the natural $^{13}\text{C}/^{12}\text{C}$ ratio. At the high-energy end of the accelerator the two stable isotope beams are measured in individual Faraday cups, which are placed in the focal plane of the high-energy magnet. The positions of these cups can be individually adjusted in order to compensate for energy loss differences of various carbon isotopes at different stripper densities and energies. In addition to the ^{12}C and the ^{13}C cups, another Faraday cup is placed to measure ^{13}C ions which are fragments of the injected ^{13}CH molecules. This current is used to measure the molecular component $^{13}\text{CH}^-$ of the beam during the injection of mass 14 ions.

Together with the information collected at the low-energy end, important information on the in-

tensity of the molecular component of the ion beam is obtained. At a given stripper density, the fraction of destroyed molecules is constant and an excessively high molecular content of an individual sample can cause background events, which impede a proper ^{14}C measurement. Our measurements show that the molecular component of different sample materials varies within narrow limits. Typical values for both, the $^{12}\text{CH}^-$ and $^{13}\text{CH}^-$ molecular currents, are in good agreement and vary between 0.5% and 1.0% of the respective stable isotope currents. By adjusting the stripper gas pressure, a molecular destruction rate is selected, which is well above the range given by the variation of the molecular content of the individual samples. Thus, undesired molecular background is avoided. During radiocarbon dating measurements, stripper gas pressure and both molecular currents are logged to control the measurement conditions.

The system is not only designed for the detection of radiocarbon. The electrostatic deflector is capable of bending particles up to an electric rigidity of 2.5 MV, which covers the total energy range provided by the accelerator. Ions with a maximum magnetic rigidity of 0.4 Tm can be analyzed with the low-energy magnet, allowing a mass range up to 130 amu at the highest possible injection energy. The maximal magnetic rigidity covered by the high-energy magnet is 0.9 Tm, which enables the detection of ions up to mass 130 amu in charge state 3^+ . Inside the analyzing chamber of the high-energy magnet, Faraday cups can be placed at the image plane to measure ion currents of stable isotopes from different radionuclides. In the focal point of the high-energy magnet, variable apertures (4–12 mm) can be set. The size of these apertures defines the accepted energy spread of the mass spectrometer. A second stripper ($3 \mu\text{g}/\text{cm}^2$ carbon foil) can also be introduced in the beam path at this position. This stripper foil can be used to break up remaining molecular ions. This is useful to study the molecular destruction process at lower gas pressures of the terminal stripper. In particular, the different molecular species, i.e. $^{12}\text{CH}_2^+$ and $^{13}\text{CH}^+$, can be distinguished by selecting the appropriate electrostatic rigidity of the molecular fragments by the

following electrostatic deflector. It is planned to add a time-of-flight detector to the system. It will be used to improve detection capabilities in the case of heavy element measurements. These options make the system well suited as experimental platform, to evaluate the potential of the new principle of molecular destruction and to investigate the relevant processes of charge changing and molecular dissociation. In particular, possibilities for the measurements of heavier radionuclides such as ^{26}Al , ^{41}Ca and ^{129}I can be investigated, because their detection is not interfered by isobaric ions. Of course, in the case of calcium, CaH_3^- ions have to be extracted from the ion source in order to reduce the potassium interference.

3. Performance

The system is operational since July 1998 and the measurements were made to study charge changing and molecular destruction processes [5] as well as to study background and stability. From 250 to 550 keV the yield of the 1^+ charge state is more than 50%, and cross-sections for molecular dissociation are high enough to destroy the molecular component at stripper densities of about $2 \mu\text{g}/\text{cm}^2$ [6]. With decreasing energy, the main limitation comes from beam losses due to increasing angular straggling. At a terminal voltage of 460 kV, favorable operating conditions were found and an ion beam transmission of typically 42% is reached at stripper densities needed for dating measurements.

To study the radiocarbon dating capability, various tests have been performed with samples of standard reference material and processed blanks. From the oxalic acid standard a clear peak of ^{14}C events is observed [5]. Processed blank samples, with a typical ^{13}CH molecular interference of less than 10^{-4} compared to the extracted negative ^{12}C current, show events clustering in the region of the ^{14}C peak. By integrating over these peaks, isotopic ratios of $2\text{--}8 \times 10^{15}$ are calculated, corresponding to radiocarbon ages between 39 000 and 48 000 yr. Similar radiocarbon ages were measured for the same blank material at the large PSI/ETH radiocarbon system [9], where ions of 26 MeV in charge state 4^+ are used.

Radiocarbon dating requires high precision $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ isotope ratio measurements. This requires the same optimal beam transport through the whole system for the three carbon isotopes. In our system, the most critical part is the injection into the tandem accelerator. To avoid isotope-specific beam losses, all three beams have to be matched as close as possible. For optimal tuning conditions, Fig. 2(a) shows normalized beam intensities of the three carbon isotopes measured at the high energy end of the system as a function of the field of the injection magnet. In Fig. 2(b) normalized isotope ratios are shown. Over quite a wide range of magnetic field intensity, a plateau of measured isotopic ratios is observed and small variations of the initial beam parameter

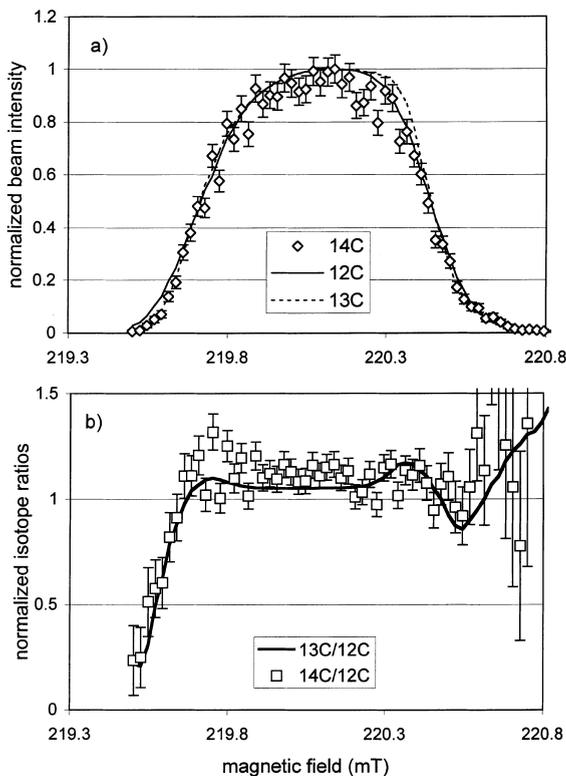


Fig. 2. Beam intensity profiles of the different carbon isotopes ^{12}C , ^{13}C and ^{14}C . (a) The field of the injection magnet is scanned. The three isotopes are injected quasi-simultaneously into the accelerator using the fast beam switching technique and are measured at the high-energy end of the accelerator; (b) The resulting normalized isotope ratios for the scan of (a).

will not cause significant changes in isotopic ratios. Our test measurements carried out so far have shown that reproducibility of isotope ratio measurements depends also on a good alignment of the ion source. Under best conditions a performance test with a set of seven oxalic acid standards has been made. Each graphite sample (2 mg) was subdivided into two fractions (A/B). The fractions were pressed into individual cathodes suitable for AMS analysis. Each cathode was measured in three individual measurement runs, which were subdivided into 10 cycles of 20 s of measurement time. The results of the measurements are shown in Table 1. For each cathode the average $^{13}\text{C}/^{12}\text{C}$ and the $^{14}\text{C}/^{12}\text{C}$ isotopic ratios for the three individual runs are shown together with the internal error due to counting statistics, the statistical (external) error of the mean values over the three runs and the associated χ^2 values. The $^{13}\text{C}/^{12}\text{C}$ ratios are not affected by counting statistics. Therefore, the deviations from the overall mean value are given. Averaging all results, the mean $^{14}\text{C}/^{12}\text{C}$ ratio has a standard deviation of 0.5% and an error of 0.13% for the mean value. This compares nicely with the internal error computed from counting statistics alone (0.5% and 0.13% mean statistical error of the three individual runs of one cathode and overall statistical error, respectively). The standard deviation of the $^{13}\text{C}/^{12}\text{C}$ measurements is about 1‰. The average values of the two different fractions A and B agree in both, the $^{13}\text{C}/^{12}\text{C}$ and $^{14}\text{C}/^{12}\text{C}$ isotopic ratios. These results clearly show that carbon isotopic ratio measurements at the 2–3‰ level can be performed with this compact AMS system. Moreover, the system may have the potential for even better performance, but this has to be demonstrated in routine radiocarbon dating measurements. At present the system is being prepared for routine operation. Data acquisition and system control software are under development to meet the requirements of unattended operation.

4. Conclusion

During the last years, large efforts have been made to extend the AMS technique to an energy range that permits the construction of small

Table 1
Compilation of results from the measurement of seven oxalic acid I standard reference material samples. Each sample was subdivided into two fractions (A/B) and pressed in individual cathodes for the analysis^a

Sample/series	$^{13}\text{C}/^{12}\text{C}$ (‰)	Error of mean (‰)	Deviation from mean (‰)	Difference (#A-#B) (‰)	$^{14}\text{C}/^{12}\text{C}$ $\times 10^{-12}$	Statistical error (‰)	Error of mean (‰)	Deviation from mean (‰)	χ^2 ext./int.
1A	1.0448	0.03	0.11	0.05	1.1945	0.51	0.37	1.10	0.5
2A	1.0436	0.02	-0.01	0.08	1.1843	0.50	0.58	0.24	1.3
3A	1.0429	0.06	-0.07	-0.08	1.1704	0.52	0.51	-0.93	1.0
4A	1.0429	0.03	-0.08	0.08	1.1819	0.52	0.57	0.04	1.2
5A	1.0447	0.04	0.10	0.20	1.1792	0.52	0.24	-0.19	0.2
6A	1.0444	0.06	0.07	0.13	1.1795	0.50	0.57	-0.16	1.3
7A	1.0456	0.01	0.19	0.16	1.1802	0.37	0.49	-0.10	1.7
Mean A	1.0441	0.04			1.1814	0.20	0.27		1.9
σ	0.10‰				0.72‰				
1B	1.0443	0.01	0.06		1.1795	0.47	0.93	-0.17	3.8
2B	1.0427	0.01	-0.10		1.1883	0.47	0.64	0.58	1.9
3B	1.0438	0.02	0.01		1.1825	0.42	0.60	0.09	2.0
4B	1.0420	0.03	-0.16		1.1845	0.51	0.63	0.26	1.5
5B	1.0427	0.04	-0.10		1.1745	0.51	0.08	-0.59	0.0
6B	1.0431	0.01	-0.06		1.1764	0.50	0.22	-0.43	0.2
7B	1.0439	0.02	0.02		1.1846	0.50	0.25	0.26	0.2
Mean B	1.0432	0.03			1.1815	0.18	0.19		1.1
σ	0.08‰				0.49‰				
Mean of all samples	1.0437	0.026			1.1815	0.13	0.13		1.1
σ	0.099‰				0.50‰				

^a The total 14 cathodes were analyzed in three independent measurement runs. Mean values of the three runs are given for the isotopic ratios $^{13}\text{C}/^{12}\text{C}$ and $^{14}\text{C}/^{12}\text{C}$ together with associated errors of mean, and in the case of the $^{14}\text{C}/^{12}\text{C}$ ratios, the error due to counting statistics. In addition, mean values computed for the two subsets A and B are given.

systems [10–12]. At PSI/ETH a compact AMS system has been developed in collaboration with NEC. It has been designed as a versatile tool to investigate the relevant processes of AMS measurements at low-beam energies. The system is now operational and several performance tests have been conducted. It has been shown that molecular interference impeding radiocarbon detection can be destroyed efficiently so that ^{14}C detection is feasible at natural levels, even if 1^+ ions are used. In particular, the possibility of radiocarbon dating has been demonstrated at beam energies of less than 1 MeV. Both, the required abundance sensitivity and the necessary overall accuracy, have been reached with the new system.

Acknowledgements

Special thanks go to J.B. Schroeder and to National Electrostatics Corporation for their valuable contribution to the PSI/ETH small AMS system. We thank Ch. Schnabel for his comments on this manuscript, and Georges Bonani and Irka Hajdas for providing the ^{14}C test samples.

References

- [1] NEC, National Electrostatics Corporation, Middleton WI, USA.
- [2] HVEE, High Voltage Engineering Europa B.V., Amersfoort, The Netherlands.
- [3] H.W. Lee, A. Galino-Uribarri, K.H. Chang, L.R. Kilius, A.E. Litherland, Nucl. Instr. and Meth. B 5 (1984) 208.
- [4] M. Suter, S.W.A. Jacob, H.-A. Synal, Nucl. Instr. and Meth. B 172 (2000) 144.
- [5] M. Suter, R. Huber, S. Jacob, J.B. Schroeder, H.-A. Synal, AIP Conference Proceedings 475, Application of Accelerators in Research & Industry 2 (1999) 665.
- [6] S.A.W. Jacob, M. Suter, H.-A. Synal, Nucl. Instr. and Meth. B 172 (2000) 235.
- [7] G.A. Norton, R.L. Loger, J.E. Raatz, R.D. Rathmell, in: Proceeding of the Symposium of Northeastern Accelerator Personnel (SNEAP), Santa Fe, World Scientific, Singapore, 1992, p. 295.
- [8] M. Suter, R. Balzer, G. Bonani, W. Wölfli, Nucl. Instr. and Meth. B 5 (1984) 242.
- [9] H.-A. Synal, G. Bonani, M. Döbeli, R.M. Ender, P. Gartenmann, P.W. Kubik, Ch. Schnabel, M. Suter, Nucl. Instr. and Meth. B 123 (1997) 62.
- [10] B.J. Hughey, R.E. Klinkowstein, R.E. Shefer, P.L. Skipper, S.R. Tannenbaum, J.S. Wishnok, Nucl. Instr. and Meth. B 123 (1997) 153.
- [11] D.J. Mous, K.H. Puser, W. Fokker, R. van den Broek, R.B. Koopmans, Nucl. Instr. and Meth. B 123 (1997) 159.
- [12] M. Suter, St. Jacob, H.-A. Synal, Nucl. Instr. and Meth. B 123 (1997) 148.