

MACS: AN ACCELERATOR-BASED RADIOISOTOPE MEASURING SYSTEM

KENNETH H PURSER, REUEL B LIEBERT, and CARL J RUSSO

General Ionex Corporation, 19 Graf Road,
Newburyport, Massachusetts 01950

ABSTRACT. A description is given of an accelerator-based radioisotope measuring system, being supplied to the Universities of Arizona and Toronto and in part to the University of Oxford. This system will be capable of measuring $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ ratios in milligram samples of cracked acetylene. At present 200 μg of carbon obtained from cracked acetylene appears to be adequate for an isotopic ratio determination. Previous performance and new calculations indicate that a precision better than 1 percent will be achieved in a half-hour period only limited by counting statistics. A precision of 0.2 percent will be obtained in a ten-hour period. Using a carbon sample with an age greater than 60,000 years, the measured background $^{14}\text{C}/^{12}\text{C}$ ratio will be less than 0.07 percent of modern.

INTRODUCTION

Recently, several groups have described accelerator techniques for the detection of single radioisotope atoms independently of their radioactivity (Bennett and others, 1977; 1978; Elmore and others, 1979; Kilius and others, 1978; Muller, 1977; Nelson, Korteling, and Scott, 1977; Purser, 1977; Purser and others, 1977; Purser, Litherland, and Gove, 1979; Raisbeck and others, 1978a; 1978b). The common feature of these accelerator procedures is that the material to be analyzed is formed into a beam of charged particles which is accelerated to energies of several MeV before electromagnetic analysis and final detection. The advantage of detection at these comparatively high energies is the elimination of molecular interferences which in the past have plagued sensitive mass spectrometry measurements. Nuclear detectors can also be used to greatly overdetermine the kinematic and intrinsic properties of individual events.

This paper describes an accelerator-based detection system, MACS (Mass And Charge Spectroscopy), being built by the General Ionex Corporation for both the Universities of Arizona and Toronto. In these systems, the isotopic ratios $^{14}\text{C}/^{12}\text{C}$ for the carbon present in the sample is compared to the ratio for a standard such as 1850 wood. The subsidiary $^{13}\text{C}/^{12}\text{C}$ ratio is also measured simultaneously for each sample so that corrections can be made for the effects caused by chemical isotopic fractionation.

Apparatus description

The principles of MACS are shown in figure 1. For carbon measurements, a primary ion beam of cesium atoms having an energy of 30keV is used to sputter negative carbon ions from the surface of a graphite-like material prepared by cracking acetylene produced from the original sample.

After production the $^{12}\text{C}^-$, $^{13}\text{C}^-$, $^{14}\text{C}^-$ ions are accelerated to an energy of 20keV together with various atomic and molecular backgrounds. These ions are then mass-analyzed using a magnet system having a resolution, $M/\Delta M \sim 100$. This high resolution virtually eliminates contributions from the neighboring masses 13 and 15; the mass-15, NH^-

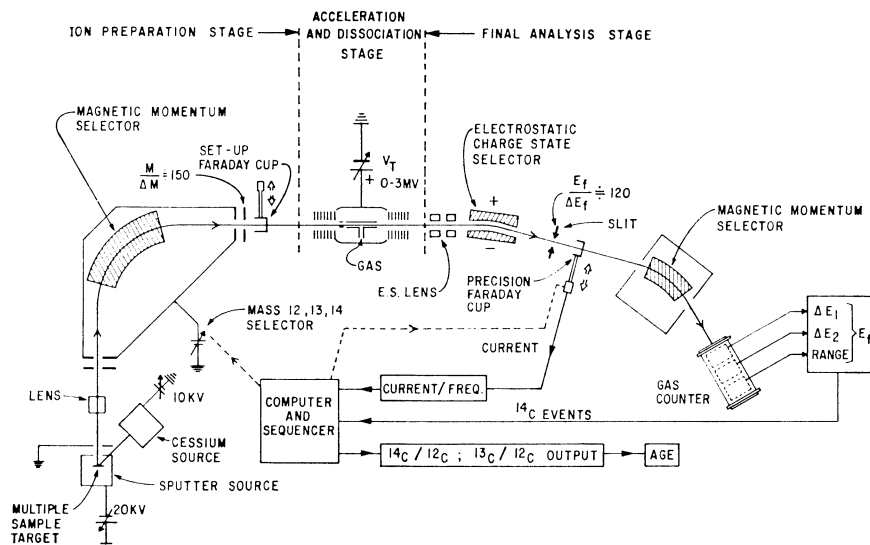


Fig 1. Ultrasensitive spectrometer system.

contaminant is particularly troublesome since it dissociates at the stripper to give ^{14}N atoms which only differ in mass from ^{14}C by 1:500,000.

After this initial mass analysis, the C^- ions are accelerated to an energy of 3MeV at the high voltage terminal of an Ionex Tandatron accelerator. Here, the particles have several outer electrons stripped from them by gas collisions (Yntema, 1974) to produce ions with positive charge. For carbon ions at these energies, the most probable charge state is 3+ and approximately 50 percent of the original C^- ions are found in this charge state. These particles, together with their companions having unwanted charge states and the molecular fragments that originate from the dissociation of the mass-14 particles $^{12}\text{CH}_2^-$, $^{13}\text{CH}^-$, $^7\text{Li}_2^-$ at the stripper, are all further accelerated back to ground by the second half of the electrostatic machine using the same 3MV potential. At the exit the wanted particles have an energy of 12MeV. There is also a broad spectrum of particles derived from mass-14 ions with a variety of discrete energies and masses.

Each of these particle species has a unique signature (Purser, Litherland, and Gove, 1979) which is easily identified and, thus, accepted or rejected. As an example, consider the unwanted charge state particles $^{14}\text{C}^+$, $^{14}\text{C}^{2+}$, $^{14}\text{C}^{4+}$, etc. These positive ions produced at the terminal are all accelerated by the second stage of the accelerator to a final kinetic energy, E , given by

$$E = eV_T(1+q) + eV_I \quad (1)$$

or

Here, eV_I is the injection energy of the ions before entering the tandem, V_T is the terminal potential, and q is the positive charge state. In contrast to conventional mass spectrometers, which operate without charge exchange and where the value of E/q is the same for all charge states, equation (2) shows that in the Tandetron configuration there is a unique value of E/q associated with each charge state so that one can identify particles of a particular charge state coming from the ion source. An electrostatic deflector with $E/\Delta E=150$, because it transmits particles with constant E/q independently of their mass (Septier, 1967), is used as the primary high energy filter.

In the machines for Toronto and Arizona, further filtering of the $^{14}\text{C}^{3+}$ particles is effected by a magnetic deflector having $M/\Delta M > 200$.

The surviving particles are stopped in a gas ionization chamber which measures for each particle its kinetic energy, E , and its rate of energy loss, dE/dx . The importance of measuring dE/dx can be seen from the fact that, at these energies, which are of the order 1 MeV/nucleon, the energy loss of a charged particle in material or a gas is given by the simplified Bethe-Bloch equation (Purser, Litherland, and Gove, 1979)

$$dE/dx = k \cdot \frac{Z^2}{V^2} \quad (3)$$

where Z is the effective charge on the particle and varies monotonically with the atomic number (Northcliffe and Schilling, 1979). Because the velocity of the particles is implicitly specified by passage through the magnet and electrostatic deflection system, equation (3) can be written

$$dE/dx = K \cdot Z^2$$

Thus, dE/dx is directly related to the atomic number of the particle. In ^{14}C measurements, this dE/dx parameter permits a clear distinction between ^{14}C particles and rare background ^{14}N ions which may have entered the system by unexpected routes. Such a route is the dissociation of NH_2^- in the residual gas before the inflection magnet where the residual NH^- has the same mass-energy product as the wanted $^{14}\text{C}^-$ ions.

Changes in source efficiency and instrument transmission

While the MACS system is designed to have high and stable transmission efficiency, some variations of instrument and source efficiency with time are inevitable. This problem is eliminated for precision measurements by arranging that bursts of ion beams from the three individual isotopes, ^{12}C , ^{13}C , or ^{14}C , are injected in a rapid controlled time sequence and accelerated through the whole system. The period of the cycle is approximately 10 seconds with 10 percent of the cycle dedicated

Performance

During ^{14}C measurements the transmission efficiency of MACS is high with the only inevitable loss of ^{14}C particles at the terminal stripping canal. Even here a substantial fraction of the original particles (typically 40 to 50 percent) enter the acceptable charge state (Yntema, 1974). Measurement of the efficiency of various parts of the instrument indicate that atom detection efficiencies of 3 percent should be possible for individual ^{14}C atoms in a cracked acetylene ion source sample. Overall, for a sample of recent origin it will be possible to measure between 15 and 30 ^{14}C events per second for a consumption of about $100\mu\text{g}$ /hours of cracked carbon. One percent counting statistics should be available within 15 minutes; 0.2 percent precision should be available from a measurement taking between 10 and 24 hours.

Apparatus description

The system shown in figure 1 divides itself naturally into four sections: 1) an ion source section, 2) an acceleration region, 3) a post-acceleration analysis stage, 4) a gas counter.

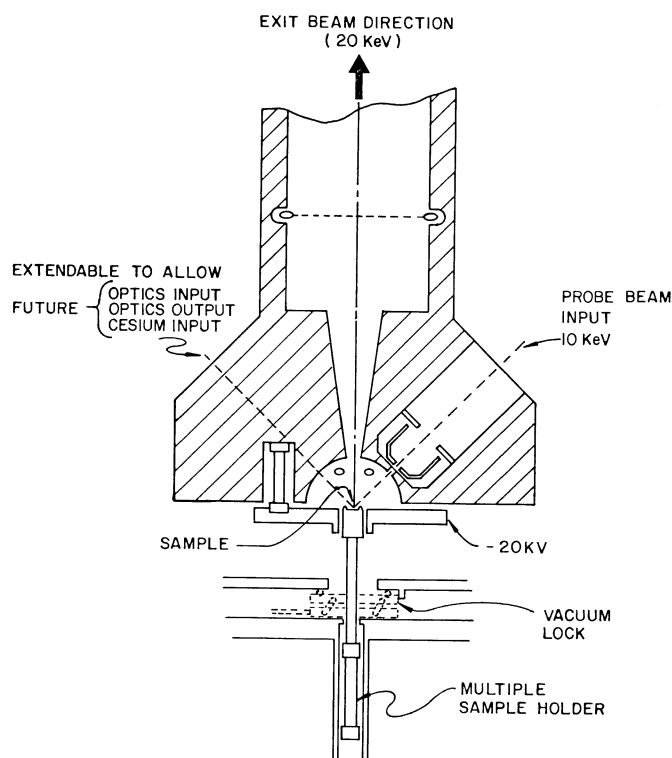


Fig 2. Ion probe source.

Ion source

The ion source used in MACS is an improved version of a sputter ion source (Brand, 1977a) that has been used for all previous radioisotope measurements at Rochester. The principles of this improved device are shown in figure 2. An incident beam of positive cesium ions with an intensity of a few hundred micro-amperes and an energy of 30 keV is focused to a small spot ($<1\text{mm}$) on the surface of the sample. Here, the incident cesium ions sputter the surface and also form a donor layer to produce the C^- ions, which are produced with about 8 percent efficiency (Brand, 1977b), are then accelerated by an axial extraction field into a beam having an emittance expected to be less than $5.5\pi\text{mm.mrad.}\sqrt{\text{MeV}}$. This sputter source demonstrates the following features: 1) high efficiency, 2) demonstrated lack of production of meta-stable $^{14}\text{N}^-$ ions (Purser and others, 1977), 3) rapid change from sample to sample, 4) readily cleaned to eliminate contamination from prior measurements, 5) isolation of individual samples to avoid cross-contamination.

The ability to eliminate carbon contamination is particularly important during low level ratio measurements and it is possible to bake the source occasionally to eliminate contamination from prior samples. High-speed turbomolecular pumps are employed near the ion source to maintain a pressure of 10^{-6} Torr or less during cesium bombardment of samples. Cryogenic vacuum pumps, operating at a temperature below 19K, provide the remainder of the vacuum capacity. At 19K, all gases other than hydrogen, helium, and neon have vapor pressures below 10^{-10} Torr. Four cryogenic vacuum heads are located on the instrument near the ion source, in the terminal, at the electrostatic deflector, and near the final detector.

Sample preparation

One important consequence of the important development work at Rochester (Bennett and others, 1977; 1978; Elmore and others, 1979; Kilius and others, ms; Purser and others, 1977) has been the recognition of the critical nature of sample preparation for precision measurements. For one percent or better reproducibility, a standardized sample preparation method is essential.

In the instrument described here, precision ^{14}C measurements will be made using solid carbon rather than gas. Gas is unsatisfactory because of poor conversion efficiency and memory effects in the ion source. The carbon for each measurement will be graphitized onto small metal buttons using the electrical discharge dissociation of acetylene.

Beukens (pers commun, 1979) investigated the cracking of acetylene for the production of suitable carbon samples for the sputter source. He has found that while electrical dissociation in an apparatus similar to that shown in figure 3 is a satisfactory technique, the deposition mechanism is strongly dependent upon the gas pressure. Optimum results are obtained at gas pressures between 5 and 10 Torr. In a conventional sputter source stable ^{12}C currents up to $7.6\mu\text{A}$ from cracked acetylene samples have been obtained after approximately one hour of operation, com-

parable to the $10\mu\text{A}$ obtained from a graphite target in the same source. With the new MACS source, currents of several times this value are expected when operating experience has accumulated. To allow for cratering during cesium sputtering, sample thickness must be at least 0.1mm. The sample diameter, however, need only be large enough to contain the cesium beam spot. Since cesium spot size can be less than 1mm, cracked acetylene samples that are datable can be considerably smaller than $200\mu\text{g}$. The cracking conversion efficiency has been measured in the most recent radio-frequency discharge tests to be 16 percent. The rest of the acetylene is converted to carbon but is not deposited in the active area of the target. It is our opinion that the radio-frequency discharge method is sufficiently developed for use in accelerator dating applications.

^{14}C , ^{13}C , ^{12}C mass selection

An important feature of the optics of the inflection magnet is that the magnet vacuum chamber is insulated from ground to permit the energy of the beam within the magnetic field region to be varied as a

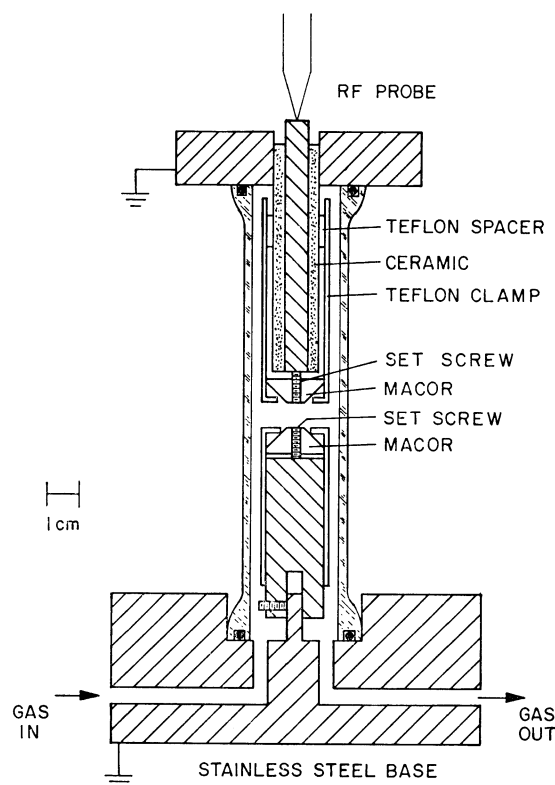


Fig 3. Radiofrequency glow discharge apparatus.

function of time. The mass-energy product for a magnet remains constant at constant field (Septier, 1967), *ie*,

$$ME = B^2 \rho^2 e^2 \frac{q^2}{2}$$

Thus, increases in energy within the magnetic field cause lower masses to be deflected through identical trajectories and injected into the machine. For the magnets used at both Toronto and Arizona, $ME = 8 \text{ AMU} \cdot \text{MeV}$. The energy fluctuations needed for selection are shown in figure 4.

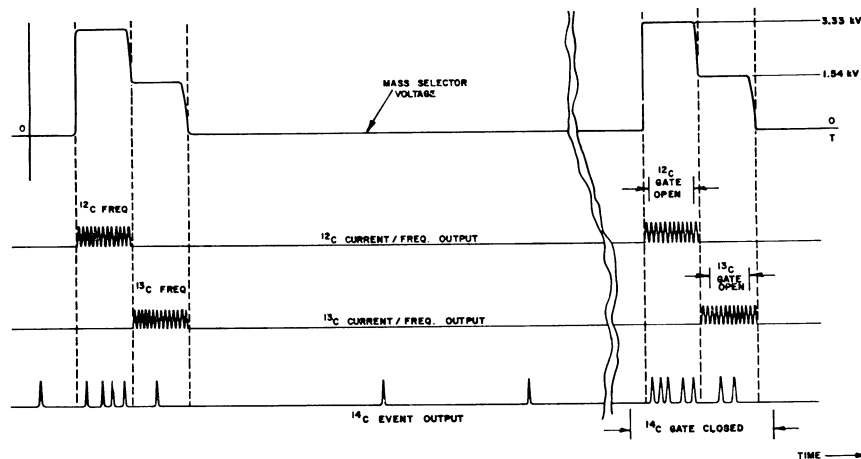


Fig 4.

The optics of the system are arranged so that the emittance of the selected particles leaving the magnet system is similar from mass to mass so that the transmission through the remainder of the components of the dating system is almost equal for all masses.

The 3MV power supply

The parallel-fed voltage multiplier consists of a series of high voltage rectifiers parallel-fed through the inherent coupling capacity between the capacitor coupling rings and an RF driver electrode driven at 35kHz (Cleland, 1959). The 3MV power supply is capable of very high currents ($\sim 3\text{mA}$), an essential feature for successive direct ^{14}C measurements because of the great disparity of the injected beam currents of $^{14}\text{C}^-$, $^{13}\text{C}^-$, $^{12}\text{C}^-$.

Terminal voltage stabilization

A precision generating voltmeter with extended frequency response is used to monitor and control the terminal voltage at all times. This voltmeter serves two functions: 1) to provide a feedback signal for 3MV terminal voltage control of better than $1:10^4$, 2) to provide a gating signal

to prohibit data collection during those periods when the terminal voltage has strayed more than $\pm 1.5\text{kV}$ from its assigned value.

Acceleration structure and optics

For radiocarbon dating, the acceleration tube structure must be designed to minimize isotope fractionation in order to eliminate excessive loss of C^- ions and to prevent fluctuations in transmission. To reach the above design goals, the tubes must be designed to keep a constant field along the length under varying beam loading; the stripper must have constant transmission and good vacuum must be maintained in the tubes.

Varying beam loading occurs during the transmission measurement test of each ^{14}C measuring cycle. Up to $10\mu\text{A}$ of ^{12}C will be injected into the machine for a fraction of a second. This current burst will not cause terminal voltage droop or optical element voltage changes in excess of 0.5kV , nor will it cause the relative transmission factor for each isotope to change more than 0.1 percent. To maintain the necessary gradient stability along the acceleration tubes from isotope to isotope, the tubes are electromagnetically suppressed and a high grading current is maintained along the acceleration tube structure. The grading resistors on the low energy tubes drain in excess of $300\mu\text{A}$ and the resistors on the less critical high energy tube drain in excess of $200\mu\text{A}$.

Variations in stripper transmission are minimized by using gas stripping. A small diameter stripper canal is desirable to minimize the gas load and the optical trajectories (fig 5) are chosen to form a waist in the stripping canal to minimize the emittance increase due to small angle scattering. Cryogenic terminal pumping is provided to handle the stripper gas load.

Good vacuum in the acceleration tubes prevents: 1) loss of C^- ions by charge exchange, 2) vacuum-dependent fractionation effects due to

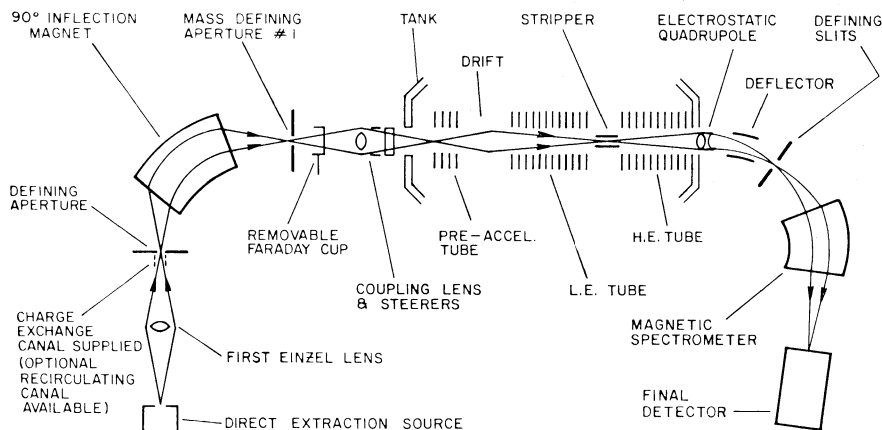


Fig 5. Optical ray diagram 3 MV tandetron accelerator.

different charge exchange cross-sections for different isotopes, 3) multiple charge exchange in the high energy tube. The cryogenic terminal pumping and cryogenic pumping at each end of the acceleration tube serves to provide excellent acceleration tube vacuum. The end pumps serve as cold traps to isolate the accelerator from the source region preventing the backstreaming of any carbon-containing species to the ion source region.

Post-acceleration analysis

No magnetic elements are used in the radioisotope system between the 90° magnetic mass-analyzer after the source and the exit from the high energy acceleration tubes. Because of this electrostatic character, the system avoids mass fractionation and the transmission efficiency for ^{12}C , ^{13}C , ^{14}C ions is identical. This mass independence is extended to the first part of the post-acceleration transport system through a combination of an electrostatic quadrupole and an electrostatic deflector. Although they are mass-independent, these two elements can discriminate against molecular fragments (Purser, Litherland, and Gove, 1979).

Electrostatic quadrupole double-deflector

An electrostatic quadrupole doublet lens focuses the particles leaving the high-energy acceleration tubes to a beam waist at the defining slits following the electrostatic deflector. Because it is electrostatic, the focal length is mass-independent for a particular charge state. This lens/deflector combination has a resolution $E/\Delta E \sim 150$ which is necessary to remove the fragmented ^{12}C and ^{13}C arising from the mass-14 beams, $^{12}\text{CH}_2^-$ and $^{13}\text{CH}^-$ and nitrogen from the mass-15 component $^{14}\text{NH}^-$.

Precision Faraday cup

A removable, precision Faraday cup is used after the defining slits beyond the electrostatic deflector to make current measurements on the ^{12}C and ^{13}C component beams. This Faraday cup is suppressed to avoid secondary electron loss and shielded to prevent high energy photon interception by the negatively biased electrode (M Nicolet, pers commun, 1979).

Magnetic spectrometer

During the mass-14 phase of the ^{12}C , ^{13}C , ^{14}C cycle, the precision Faraday cup will be withdrawn and the particles which have passed through the defining slits are momentum-analyzed by a strong-focusing 45° sector magnetic spectrometer (H Enge, pers commun, 1979) with $ME/Z^2 = 27\text{MeV-AMU}$. This spectrometer has a radius of curvature 0.6m.

Heavy ion Z-detector

The Heavy Ion Identifier is a gas-filled detector with many features similar to the Rochester heavy ion detector (Shapira and others, 1975). The Ionex detector has two dE/dx stages with transverse collection, and a final range stage with longitudinal collection. The signals produced by the detector include E_{total} , two dE/dx signals, and range.

A schematic diagram of the detector and its electronics is shown in figure 6. The active length of the counter is 500mm. The window size is

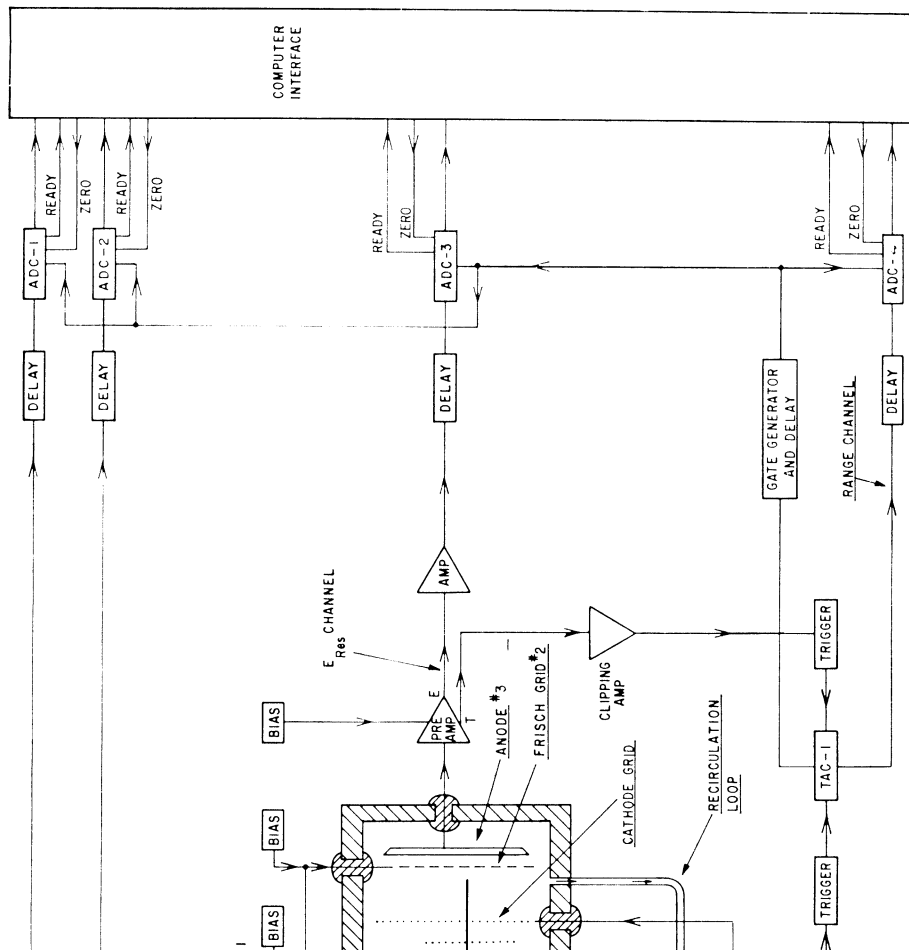


Fig. 6. Schematic diagram of converter and its electronics.

limited to a diameter of 5mm and is covered by a 1 μ m Paralene-C foil. The anode is subdivided into two parts to give independent dE/dx signals from each of two plates. The particles are stopped in a final section where the collecting electric field is rotated by 90° to have its direction along the axis of the counter. This geometry permits a determination of Z by measuring the range (Holt and Litherland, 1954) for each particle, independent of the two dE/dx measurements.

SUMMARY

A MACS system has been described which uses a 3MV tandem accelerator and can be extended for the direct detection of the radioisotopes ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , and others. Measurements at the University of Rochester (Bennett and others, 1977; 1978; Elmore and others, 1979; Kilius and others, ms; Purser and others, 1977) and other calculations indicate that, for ^{14}C measurements, a precision better than one percent will be achieved in a half-hour period only limited by counting statistics. A precision of 0.2 percent will be obtained in a ten-hour period. With a carbon sample older than 60,000 years, the measured background $^{14}\text{C}/^{12}\text{C}$ ratio will be less than 0.07 percent of modern as defined by Stuiver and Polach (1977).

There is less information on precision expected for the other radioisotopes. It is expected, however, that the system will also be capable of measuring ^{10}Be , ^{26}Al at sensitivities of magnitude 1:10¹⁵. This is about two orders of magnitude more sensitive than is needed for groundwater aging.

Finally, it should be noted that the same techniques in many fields of research are related to the characterization of surface and bulk solids and to high spatial resolution where stable isotopes are detected rather than radioactive atoms. Construction is technologically possible today of

REFERENCES

- Bennett, C L, Beukens, R P, Clover, M R, Elmore, D, Gove, H E, Kilius, L R, Litherland, A E, and Purser, K H, 1978, Results from radiocarbon dating of several established samples: *Science*, v 201, p 345-347.
- Bennett, C L, Beukens, R P, Clover, M R, Gove, H E, Liebert, R B, Litherland, A E, Purser, K H, and Sondheim, W, 1977, Radiocarbon dating using accelerators; negative ions provide the key: *Science*, v 198, p 508-510.
- Brand, K, 1977a, Performance of HICONEX sputter source: *Nuclear Instruments and Methods*, v 141, p 519-520.
- 1977b, A reflected geometry for a sputter source: *Rev Phys Appl*, v 12, p 1453.
- Cleland, M R, 1959: US Patent 2,875,394.
- Elmore, D, Fulton, B R, Clover, M R, Marsden, J R, Gove, H E, Naylor, H, Purser, K H, Kilius, L R, Beukens, R P, and Litherland, A E, 1979, Analysis of ^{36}Cl in environmental water samples using an electrostatic accelerator: *Nature*, v 277, p 22-26.
- Holt, J R and Litherland, A E, 1954, A range method for Z-identifying of charged particles: *Rev Sci Instrument*, v 25, p 450.
- Muller, R, 1977, Radioisotope dating with a cyclotron: *Science*, v 196, p 489-494.
- Nelson, E, Korteling, R G, and Scott, W R, 1977, Radiocarbon measurements using the McMaster Tandem accelerator: *Science*, v 198, p 506-508.
- Northcliffe, L C and Schilling, R G, 1979: *Nuclear data tables*, A7, p 233 (1970).
- Purser, K H, 1977: US Patent 4,037,100.
- Purser, K H, Liebert, R B, Litherland, A E, Beukens, R P, Gove, H E, Bennett, C L, Clover, M R and Sondheim, W E, 1977, An attempt to detect stable N^- ions from a sputter source and the implication for ^{14}C detection: *Rev Phys Appl*, v 12, p 1487-1492.
- Purser, K H, Litherland, A E and Gove, H E, 1979, Ultra-sensitive particle identification systems: *Nuclear Instruments and Methods*, v 162, p 637-656.
- Raisbeck, G M, Yioux, F, Fruneau, M, Lievin, M and Loiseaux, J M, 1978a, Detection of ^{26}Al using ALICE: *Nature*, v 275, p 731-733.
- Raisbeck, G M, Yioux, F, Fruneau, M and Loiseaux, J M, 1978b, Detection of ^{10}Be using the Grenoble cyclotron: *Science*, v 202, p 215.
- Septier, A, 1967, *Focusing of charged particles*: New York, Academic Press.
- Shapira, D, de Vries, R M, Fulbright, H W, Toke, J, Clover, M R, 1975, A broad band nuclear detector for use in focal planes of magnetic spectrographs: *Nuclear Instruments and Methods*, v 129, p 123-130.
- Stuiver, Minze and Polach, H A, 1977, Discussion: Reporting of ^{14}C data: *Radiocarbon*, v 19, p 355-363.
- Yntema, J L, 1974, Heavy ion stripping in Tandem accelerator terminals: *Nuclear Instruments and Methods*, v 122, p 45-52.

DISCUSSION

Willkomm: How many persons (physicists and/or technical staff) are needed for running the machine itself?

Purser: It is necessary to have an engineer/physicist who has as his primary responsibility the running of the machine. It would be desirable to have available also a technician for each operation shift who would insert samples, clean the ion source, and help with maintenance problems when they occur. These persons would be exclusive of the work needed for cleaning and preparing specimens. It should be recognized, however, that these numbers do not assume that any development of the machine will take place and that the instrument is used solely for ^{14}C dating. If detection of other more difficult isotopes are planned the staff would need to be considerably larger.

Farwell: We understand from Dr Gove's remarks in this morning's session that this process, or certain aspects of it, have been patented. Would you care to comment upon the possible implications of the existing (or

contemplated) patents in regard to the use of tandem accelerator radio-carbon dating techniques by other groups?

Purser: We would be delighted to provide licenses to use the patent to any bona-fide non-commercial group. If you have questions please feel free to write to me directly.

Roether: Would you be willing to give an estimate on cost per sample, for a ^{14}C precision of ± 1 percent, for a routine laboratory using the technique that you have described?

Purser: Excluding sample preparation costs, which may be substantial, the time for running a sample to give a precision of ~ 1 percent should be less than $\frac{1}{2}$ hour. Realistically, the machine probably needs 1 to 2 people dedicated to its maintenance. Allowing for overheads, I would plan \$50 to \$75 per sample as the basic cost.