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Authors

Bertsche, K.J.

Karadi, C.A.

Muller, R.A.

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RADIOCARBON DETECTION WITH A SMALL LOW ENERGY CYCLOTRON

Kirk J. Bertsche, Chandu A. Karadi, and Richard A. Muller

Lawrence Berkeley Laboratory

University of California

Berkeley, CA 94720

ABSTRACT

A small cyclotron (the "cyclotrino") was proposed for direct detection of radiocarbon in 1980. This combined the suppression of background through the use of negative ions, which had been used effectively in tandem accelerators, with the high intrinsic mass resolution of a cyclotron. Development of a small electrostatically-focused cyclotron for use as a mass spectrometer was previously reported but the sensitivity needed for detection of ^{14}C at natural abundance was not achieved.

A high current external ion source has now been integrated with a small flat-field, electrostatically-focused cyclotron to comprise a system capable of measuring ^{14}C at natural levels. The carbon ion beam from the ion source is pre-separated with a Wien filter and is transported to the cyclotron via a series of electrostatic lenses. Beam is injected radially into the cyclotron using electrostatic deflectors and an electrostatic mirror. Axial focusing is entirely electrostatic. A microchannel plate detector is used with a phase-gated output.

The design and performance of this system are presented here. In its present form the system is capable of improving the sensitivity of detecting ^{14}C in some biomedical experiments by a factor of 10^4 . Modifications are discussed which could bring about an additional factor of 100 in sensitivity, which is important for archaeological and geological applications. Possibilities for measurements of other isotopes, such as ^3H , ^{10}Be , and ^{26}Al , are discussed.

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1. INTRODUCTION

Since the introduction of radiocarbon dating by Libby in the 1950's, ^{14}C measurements have primarily been performed by decay counting. This method works well for samples which are large enough and recent enough to give a high decay rate (one gram of modern carbon produces about 14 disintegrations per minute [dpm]), but for old or small samples the necessary counting time becomes impractical.

This limitation of decay counting has been answered by accelerator mass spectrometry (AMS). Rather than counting decays of ^{14}C , the ^{14}C atoms themselves are detected and counted using a particle accelerator. This provides a huge advantage in sensitivity over decay counting techniques.

An accelerator was first used as a mass spectrometer by Alvarez when a large cyclotron was used for detection of ^3He , reported in 1939. [1-3]. The technique was re-introduced by him in the mid-1970's as part of a search for anomalous particles (quarks) with integral charge [4, 5]. It was first suggested as a means of measuring radioisotopes in 1976, and a successful age determination was made using tritium [6]. Although this early work was done with cyclotrons, measurements quickly shifted to tandem Van de Graaff accelerators, especially for ^{14}C measurements due to the elimination of interference from ^{14}N [7-10]. During the past ten years or so, techniques have been refined so that AMS is now a quite accurate method of dating ^{14}C and other radioisotopes.

This method has been relatively costly due to the need for a large, high-energy particle accelerator. In response to the increasing demand and high cost of accelerator mass spectrometry (AMS) techniques, the small cyclotron ("cyclotrino") project was begun at Berkeley in 1981 [11]. The basic idea was to combine the excellent properties of a cyclotron used as a mass spectrometer with the capabilities of negative ion sources to reject unwanted backgrounds such as ^{14}N [7,8].

A small cyclotron (the "cyclotrino") was built at Berkeley to test these concepts [12-15]. This cyclotron incorporated a miniature Cs sputter negative ion source at the center of the cyclotron, injecting negatively-charged carbon ions at about 3 keV. Extraction energy was about 40keV at about a 10.5 cm radius. The cyclotron was operated at the 11th to 15th harmonic to obtain the necessary resolution and relied solely on electrostatic focusing for ion confinement. It was found that such a device does indeed produce the necessary resolution for detection of ^{14}C .

Unfortunately, ^{14}C could not actually be detected in the early version of the cyclotron because of the low output of the internal ion source which was used. It was necessary to rebuild the cyclotron to provide for a high-current external ion source. The cyclotron reported here is very similar in design to the earlier machine, with the addition of an external ion source and injection beamline, and is shown in Fig. 1.

Research on similar small cyclotrons as mass spectrometers has also begun in China [16-18] and Yugoslavia [19]. These machines are both somewhat more complex than the cyclotron discussed here and differ in a number of important details. They show great promise, though they have not yet progressed as far as this work.

Our initial work has emphasized ^{14}C , partly because of the many users interested in this isotope and partly because the required mass resolution and beam currents are relatively easy to obtain. Measurements of other radioisotopes, such as ^{26}Al , ^{10}Be and ^3He are also possibilities with the cyclotron but have not been pursued (see section 6).

While the initial impetus for the small cyclotron was archaeological research, biomedical researchers have become interested in this tool as well. In studies of the human body only a limited amount of ^{14}C can be safely given to a patient. For some metabolic studies it is necessary to separate blood into its various constituent fractions and to determine the amount of ^{14}C in each fraction, which may be so small as to contain less than 1 μg of carbon. Decay counting does not yield the required sensitivity for many such studies. Since an activity of 1 dpm or so is needed to make a measurement in a reasonable amount of time, a 1 μg sample must be enriched 7×10^4 times above modern levels to be measurable. A small cyclotron having a carbon current of 10 μA and a detection efficiency of 0.1% would give about 4 counts per minute with modern carbon, improving the count rate by 5 1/2 orders of magnitude! Clearly, the cyclotron could be very beneficial in such metabolic studies.

2. CYCLOTRON DESIGN

2.1 General Design Considerations

In a cyclotron, charged particles orbit freely in a magnetic field. The orbital frequency of these ions (the cyclotron frequency, $\omega = qB/m$) is a constant for non-

relativistic particles. The kinetic energy of a charged particle in a cyclotron may be increased by the imposition of electric fields which are driven synchronously at the cyclotron frequency or at a multiple of it. These electric fields establish a resonance condition for ions of the selected mass. Off-mass ions eventually fall out of synchronization with the alternating electric fields and are lost.

In a traditional two-dee cyclotron, each dee subtends 180° . Thus the electric fields are imposed at two positions in the particle's orbit, displaced 180° from each other. In such a design the electric fields may be driven at the cyclotron frequency or at any odd harmonic of the cyclotron frequency. Such harmonic operation will improve the resolution of a cyclotron used as a mass spectrometer, since mass resolution is roughly proportional to the product of the number of turns and the harmonic [12,14]. Typically, sinusoidal excitation is used, though with the relatively low RF voltages occurring in a cyclotron used as a low-energy mass spectrometer, non-sinusoidal excitation could be considered and has been suggested by other researchers [18].

This two-dee design allows one dee to be fixed at ground potential while the other is driven with the alternating RF waveform. The grounded dee may be replaced by a narrow strip ("dummy dee"), freeing a large area for insertion of electrostatic deflection channels, dee probes, etc.

Design of this cyclotron is essentially the same as that described previously by our group [12-14, 20-22]. A beam of carbon ions is introduced into the cyclotron. The cyclotron's magnetic field and RF frequency are tuned such that only the desired species (^{14}C) is accelerated; other species eventually fall out of phase with the RF and cease being accelerated, never reaching extraction radius. The cyclotron operates with negative ions to avoid interference from ^{14}N . With ^{14}N eliminated, the nearest interfering mass is the molecular ion ^{13}CH , which is heavier than ^{14}C by a part in 1800. This sets the minimum resolution requirements. The necessary resolution is obtained by operating at a high harmonic (11th to 15th) of the fundamental cyclotron frequency and by using a very flat magnetic field, which causes orbits to be isochronous and hence allows ions to make many turns (50 to 100) in the cyclotron. Focusing is purely electrostatic; the flat magnetic field provides no weak focusing. While beam bunching has been suggested and should improve transmission, this machine uses an unbunched, continuous beam.

A 12-inch NMR-type laboratory magnet is used, operated at about 10kG. Beam is injected at an energy of 5 keV with a 2.8 cm radius and extracted at about 40keV with a 10

cm radius. Ions must gain about 260 eV in the first two gap crossings for about half of them to clear an electrostatic deflection channel in the center of the cyclotron [24].

This cyclotron is typically operated with a harmonic of 11 to 15 and a peak dee voltage of 300 to 400 volts. The cyclotron frequency is about 1 MHz with a magnetic field of about 1 Tesla, so the dees must be driven at 11 to 15 MHz at 600 to 800 volts peak-to-peak. This is done with a modified amateur radio transmitter with an oven-controlled crystal driving a linear amplifier [22].

2.2 Stability

In traditional cyclotrons, orbit stability is dependant primarily on the magnetic field index of the machine [25 ch. 2]. The magnetic field is given a radial gradient; the field index is defined as the ratio of the fractional change in B to the fractional change in radius. In the cyclotrino it has been necessary to keep the field index as close to zero as possible to retain orbit isochronicity. Isochronous orbits are necessary if particles are to be retained for a large number of turns, which in turn is necessary for high resolution. Thus, orbit stability mechanisms in the cyclotrino are somewhat more subtle than in traditional designs.

In this machine, axial focusing is purely electrostatic. This provides sufficient vertical focusing to contain most of the beam from the ion source. The magnetic field provides radial focusing with radial betatron frequency $\nu_r = 1$. This is potentially unstable, but does not cause problems with careful design and a sufficiently flat magnetic field. There are no phase oscillations, but there is significant phase bunching. These stability mechanisms have been analyzed in detail [22,24].

2.3 Acceptance

Cyclotron acceptance must be matched to ion source emittance to avoid losing beam. Extrapolating from estimates of the source emittance at 20 keV, it was estimated that about 80% of the source output should fall within 40π mm-mrad at 5 keV. The acceptance of the cyclotron has been analyzed [22,24] and is found to be approximately 40π mm-mrad also. Thus, with appropriate focusing, nearly the entire beam profile should be accepted.

Beam is accepted over only about a 45° RF phase window due to the focusing and acceleration properties of the cyclotron. Thus, without a beam buncher, about 12% of the beam should be accepted into the cyclotron.

3. ION PRODUCTION AND INJECTION

3.1 Overview

A beamline transports the beam from the ion source to the cyclotron, shown schematically in Fig. 2 and pictorially in Fig. 3. An einzel lens captures and slightly focuses the highly divergent output of the ion source. An electrostatic quadrupole lens further focuses and steers the beam into the object slit of a Wien filter. The Wien filter (a velocity selector) performs the dual purpose of reducing the ion load entering the cyclotron (including elimination of the high energy tails from ^{12}C and ^{13}C) and allowing ion source output to be monitored. It focuses the mass 14 beam from the object slit to an image slit, deflecting the mass 12 and 13 ions to a Faraday cup to provide normalization for ion source output fluctuations. The mass 14 beam (containing ^{14}C and molecular ions such as ^{13}CH) is focused and steered into the cyclotron with a combination of four electrostatic quadrupole lenses. The beam is injected into the cyclotron radially using electrostatic deflection channels and an electrostatic mirror.

A computer program was written to simulate the optical performance of the beamline and cyclotron injection, based on 3×3 optical matrices [22]. This program allows easy examination of the first-order optical properties of the system. Either an individual ray or the beam envelope may be traced through the system. This allowed adjustment of beamline composition and geometry, leading to the beamline shown in Fig. 2 with the beam envelope shown in Fig. 4.

In addition to these first-order optical analyses, aberrations caused by higher order effects must be considered. For elements which are symmetric in the off-axis dimensions (e.g. einzel and quadrupole lenses) even-order aberrations cancel; the lowest-order aberration remaining is third order. For asymmetric elements (e.g. Wien filters) there are second-order aberrations as well.

In the beam injection system, aberrations are primarily due to the einzel lens, with the Wien filter contributing to a smaller extent. The quadrupole lenses contribute negligibly to the overall aberration.

3.2 Ion Source

The earlier version of the small cyclotron did not have enough current to enable ^{14}C detection at modern abundance levels [12,14]. For this reason the current machine was built with an external ion source.

A Cs-sputter negative ion source was purchased from General Ionex, capable of more than $20\text{ }\mu\text{A}$ of C^- output using graphite or CO_2 . The source is designed to operate with about 20 keV extraction energy. However, this machine is designed to have only a 5 keV input beam energy; thus, beam properties were not well known. Extrapolating from performance at 20kV [26], estimates were that at 5 keV the beam should have an emittance of about $40\pi\text{ mm-mrad}$ with an apparent origin over a region of about 1 mm diameter at about the actual sample location. These estimates are probably not extremely accurate, since there is significant loss of beam before the exit of the Wien filter. The ion source may perform better with initial acceleration to 20 keV followed by deceleration to 5 keV.

An RF plasma source was also investigated [27]. This has the advantages of a very simple design and a small beam emittance and seems like a promising approach for future AMS measurements. It was not used in this system because in early tests the source could not reliably generate C^- , though it now may be able to do so [28].

3.3 Einzel Lens

An einzel lens is a rotationally symmetric electrostatic lens. Typically, an einzel lens is composed of a grounded conductor at each end, either a tube or aperture, with an intermediate tube or aperture which is operated at a potential at or near the potential of the ion source [29 ch.6, 30, 31].

Einzel lenses generally have severe spherical aberrations. Thus it is important to minimize the spherical aberration coefficient, C_s , for an einzel lens design. Calculations of the focal lengths and spherical aberration coefficients for a number of einzel lens designs

may be found in the literature [28, 29 ch. 10, 32-36]. It has been found that C_s may be reduced in einzel lenses by making lenses with hyperbolic surfaces [37], by adding foils or grids [38-41], or by making them axially asymmetric [37, 42-45].

An axially asymmetric lens similar to that of Riddle [42] and Drummond [45] was constructed for the cyclotron (Fig. 5). This seems to work fairly well, but a significant amount of aberration seems to be introduced by the combination of the ion source and einzel lens.

3.4 Quadrupole Lenses

Good control is needed of beam characteristics (beam position, beam width, and focal plane locations) on injection into the cyclotron. A combination of four electrostatic quadrupole lenses provides the needed flexibility. By independent adjustment of the powers of the four lenses, control is gained over the four degrees of freedom associated with beam focal properties. (These four degrees of freedom may be described as the position and size of the beam focus for each of the two dimensions orthogonal to the beam axis.) Beam steering may be accomplished by driving the quadrupoles asymmetrically; this allows adequate control of the four additional degrees of freedom associated with beam position. Custom-built power supplies allow independent adjustment of the focal power and vertical and horizontal deflections of the quadrupole lenses [22 Appendix C].

The lenses which were built for the small cyclotron have a length of 4.0 inches, a pole diameter of 2.0 inches, and an aperture of 1.74 inches (Fig. 6). The aberration of the quadrupole lenses was found to be relatively unimportant [22]. Although the beam becomes quite large in the quadrupoles (Fig. 4), there is virtually no loss of beam in traveling from the Wien image slit to the cyclotron's injection channel.

3.5 Wien Filter

A Wien filter (also known as a crossed-field separator or a velocity selector) is used in this system to pre-separate the incident ion beam, reducing the amount of ^{12}C which is injected into the cyclotron itself. A Wien filter relies on an electric and a magnetic field which are orthogonal to one another. The ion beam travels along an axis orthogonal to both. The electric and magnetic fields are adjusted to pass the desired species undeflected; any species with incorrect velocities are deflected away from the main path.

The Wien filter in this system is composed of a slit, a drift length, the crossed field region, a second drift length and a second slit. The length of the crossed field region is such that:

$$L \equiv \frac{\pi}{2} r_c \quad (1)$$

where r_c is the nominal cyclotron radius for particles in the crossed field region. The two drift lengths are each about r_c in length. This causes an image of the first slit to be focused at the second slit [22 Appendix D]. It is possible to adjust the focus of the filter by changing the beam energy (velocity), either at the ion source, or inside the Wien filter by driving the Wien filter electric field plates asymmetrically [22].

A symmetric optical element such as a quadrupole lens or einzel lens will have no second-order aberrations due to symmetry about the beam axis. However, a Wien filter is not symmetric and has second-order aberrations which must be considered. These aberrations may be calculated by integrating the equations of motion for an ion traversing the filter [22]. Their effects may be controlled by limiting the angles of incident ions. The beam should not be focused to an extremely narrow ribbon at the slits, because the beam's angular spread will grow as a consequence of conservation of beam emittance. It is also advisable to physically limit the maximum angles which can traverse the filter; this may be done by inserting a field aperture at a point optically conjugate to the entrance slit. For our design this consists of merely inserting an aperture at the entrance to the magnetic field region.

The Wien filter in this system has been built with samarium cobalt permanent magnet material and a soft steel yoke, placed entirely in vacuum (Fig. 7). The magnetic field is 4.2 kG over an effective length of 12.0 cm, with a width of about 4.7 cm and a gap of about 2 cm. According to calculations [46], this should produce a field uniform to better than 0.2% over a 1 cm by 1 cm central region. Magnetic field shunts cause the field to drop quite rapidly at the ends, falling from 90% to 10% of its maximum value in less than 2 cm.

The electric field is produced between two parallel plates about 2.5 cm apart. Guard electrodes were placed near the magnet poles and two fringe field shunts were placed at each end to reduce electric field distortion [22,47]. The electric field assembly is shown in Fig. 8.

The electric potentials are applied asymmetrically in this design, with one electric field plate grounded and the other operated at a high negative voltage (about 2500 V). This slows the incident 5 keV beam significantly, shortening the necessary length of the Wien filter since ions spend more time in the crossed field region.

The separation of masses obtained with this Wien filter is shown in Fig. 9, measured at low currents with a Faraday cup at the output slit of the Wien filter. This plot greatly understates the off-mass rejection of the Wien filter. Many off-mass ions are able to exit the Wien filter's image slit but do not fall in the injection channel of the cyclotron because they exit the slit with a small angular deviation. Measurements of ^{12}C taken at the detector show that off-tuning the Wien filter by one mass unit (to mass 11) suppresses current by a factor of 50, while off-tuning by two mass units (to mass 10) suppresses by 10^5 . Thus, with the Wien filter tuned to mass 14, very little mass 12 should enter the cyclotron.

3.6 Injection

The ion beam is injected into the cyclotron using a novel radial injection method [48]. This was used because of concern that the more traditional axial injection would cause distortion of the magnetic field near the center and destruction of orbit isochronicity, due to the need to bore a hole through one of the magnet pole pieces. The radial injection system is shown schematically in Fig. 10, with a photograph in Fig. 11.

Beam crosses the magnet edge at an angle, providing the proper focusing characteristics for a nearly parallel incident beam to be axially focused to a beam waist at the first dee gap crossing [22]. After entering the magnet region it is deflected in an electrostatic channel. This deflects the beam through about a 90° angle with a 3.8 cm radius. The beam then curves through a 180° arc, whereupon it strikes an electrostatic mirror at normal incidence. After reflection the beam travels along a circle which is nearly, but not exactly, centered on the cyclotron axis. A final electrostatic deflection channel (1.9 cm radius over about 30° , with a 2 mm gap) near the center of the cyclotron shifts the beam orbit slightly to center it and to provide clearance between the electrostatic mirror and the beam's first orbit.

The electrostatic mirror is composed of a grounded metal mesh with a flat, high voltage electrode behind it. A 5 keV beam passing through the mesh drifts in the electric field until it reaches a 5 kV equipotential surface, turns around, and re-exits the mesh. The

mirror electrode was operated at a potential slightly above the incident beam energy to keep beam from reaching the electrode and depositing on it.

The optical properties of the electrostatic mirror and electrostatic deflection channels have been calculated [22]. When the matrices for these elements are inserted into the beamline program mentioned earlier, the beam envelope of Fig. 12 is obtained.

4. ION EXTRACTION AND DETECTION

4.1 Extractor

An electrostatic channel is used to deflect ions out of the cyclotron. The entrance to the channel is located 90° from the dee gap (Fig. 10,11). This prohibits any low energy ions from entering the channel by drifting along the dee gap.

The channel has a 45° arc with a 7.9 cm radius and a 2 mm gap between the two concentric electrodes. The extracted ions exit the magnetic field and strike the detector.

The first order optical effects of the channel and the crossing of the B-field edge combine to produce a focus in the axial dimension with size of about 2 mm at a distance of about 15 cm from the actual edge of the magnet. At this point the radial dimension has a size of about 13 mm.

4.2 Detector

A custom microchannel plate detector was designed and built to detect the extracted ions [49,50]. This detector uses an aluminum oxide conversion dynode which is impacted by the extracted ions at glancing incidence. A number of secondary electrons are liberated from the aluminum and pulled into the microchannel plate. This allows discrimination between the 30-40 keV output ions and any low energy ions or photons, which will release fewer secondary electrons.

The microchannel plate is operated in saturation, so that each pore which is struck will produce a saturated pulse at the anode of the microchannel plate. Thus the output

charge is proportional to the number of secondary electrons collected by the microchannel plate.

The extracted ions should fall into a region about 2 mm by 13 mm at a distance of 15 cm, where the detector is placed inside a magnetic shield made of magnet-grade soft steel. Ions strike the detector's aluminum dynode with an incident angle of about 12° . This angle reduces its area of 19mm by 13mm to a projected area of about 4 mm by 13 mm which should provide a good match to the extracted beam size. At this angle, an average of about 17 secondary electrons should be liberated by an incident ion; the peak of the distribution should be at 10 to 15 electrons [49].

A large number of neutral particles, presumably UV photons, seems to be produced in the injection channel. This is probably due to secondary electrons which are produced in the channel by incident ions, gain energy from the electric field, then strike surfaces at high velocity. Copper plated optical shields were installed to block and absorb these photons. Unfortunately, a significant amount of UV still strikes the detector. The tail of the UV-induced single electron distribution extends well into the distribution for high energy ions. Because of this it was necessary to reduce the angle at which ions strike the dynode by both physically rotating the detector and by increasing the dynode voltage to deflect beam electrically. The angle of ion beam incidence was reduced from about 12° to about 6° , shifting the peak of the high energy ion distribution from about 10 secondary electrons to about 17 secondary electrons. This also reduced the projected area of the dynode by a factor of two, making alignment much more critical.

4.3 Detection Electronics

In a further attempt to reduce the interference of UV photons, pulses from the detector were gated in time (Fig. 13). Pulses were only accepted if they fell into a phase window of about 90° width. This reduced the number of UV photons by about a factor of four, while reducing the number of high energy ions by about 25%. The width of this window can not be reduced arbitrarily; ions are spread in phase due to a small spread in extraction energy (which is due to the radial spread in position of orbit centers, as discussed in section 2.3.2), and due to their striking the dynode in different places, which correspond to different distances from the extractor. For our design, the time spread at the detector due to the ions' velocity spread is about ± 6 ns and the time spread due to the

dynode size is about ± 9 nS, resulting in a ± 11 nS spread (adding in quadrature). For our typical operation at the 15th harmonic, this corresponds to about $\pm 60^\circ$.

The tradeoffs involved in adjusting the width of this phase window may be seen in Fig. 14. Data is shown at the 15th harmonic and 400 V peak for no phase windowing, a wide phase window, and a narrow phase window. Data for the "noise" curves was taken about 10kHz above the ^{13}CH peak, while data for the "signal" curves was taken on the ^{13}CH peak with some of the lenses in the beamline de-tuned to protect the microchannel plate detector from high count rates.

5. SYSTEM PERFORMANCE

5.1 Transmission

System losses are illustrated in Table 1. The transmission of the most complex part of the system (from the output of the Wien filter to the extraction channel of the cyclotron) is quite good, at about 2.5%. A factor of 8 loss in the cyclotron would be expected due to phase acceptance alone. (Only ions with a phase between about 15° and 60° have enough energy gain and focusing to reach extraction radius.) Transmission through the cyclotron itself is shown in Fig. 15, measured with a dee probe. The abrupt drop at about 10 cm is due to interception of ions by the extraction channel.

Table 1
Transmission losses in cyclotron system.

From:	To:	Loss Factor
ion source	Wien first slit	4*
Wien first slit	Wien second slit	5
Wien second slit	injection channel	1
injection channel	mirror	2
mirror	inner deflector	2
inner deflector	extraction radius	10
extraction radius	detector	5
phase discrimination		1.3
amplitude discrimination		4
Wien output	extraction radius	40
Wien output	detection	10^3
ion source	detection	2×10^4

* estimated

The overall efficiency of the system is somewhat lower than expected, with unexpected losses seen at each end of the system. Loss at the beginning of the beamline is probably due to aberrations (and possibly some mispositioning) of the einzel lens. It should be possible to improve this performance by re-designing the einzel lens on the basis of detailed emittance measurements or computer calculations, yielding a transmission increase of a factor of 5 to 10.

There is also unexpected loss in ion detection due to the discrimination of ions from UV photons. The loss from extraction radius to detector is larger than expected; a factor of two would be expected [12] while a factor of five is seen. This is probably due to part of the beam missing the dynode as a result of minor misalignments and uncertainties in beam focus position. A factor of four loss is due to setting a high discrimination threshold.

It may be possible to reduce these detection losses by a more thorough addition of optical absorbers, though this would probably not make a significant improvement. However, extending the size of the dynode (to present a larger projected area to the beam) and placing the dynode at more of a glancing incidence (to produce more secondary electrons from each incident ion) should reduce losses both from beam missing the dynode and from the detection threshold, improving the detection efficiency by at least a factor of

four. This would necessitate either a larger microchannel plate or focusing of the secondary electrons onto the plate.

It is also possible to add a beam buncher. This has been suggested for this system [20] and has been investigated in detail by Karadi [22 Appendix F]. A beam buncher should yield another factor of four increase in efficiency.

Making all of the above improvements (einzellens, beam buncher, and detector modifications) should boost system efficiency by two orders of magnitude.

5.2 ^{14}C Measurements

The resolution of the system has proven to be adequate for ^{14}C measurements, as shown previously [12,14]. The resolution peak is relatively flat over a frequency range of about $\pm$ one part in 6000, with very steep skirts outside this range which fall about six orders of magnitude for an offset of one part in 10,000 (Fig. 16).

At mass 14, the dominant species are $^{12}\text{CH}_2$ and ^{13}CH . The relative proportion of these two species is quite variable, depending on residual gases in the vacuum chamber and on ion source output current. (As source output is increased, the sample heats up and $^{12}\text{CH}_2$ production drops in relation to ^{13}CH .) A typical scan of these peaks is shown in Fig. 16.

A sample of modern carbon prepared by Erv Taylor from the OX-1 standard was measured in the system (Fig 17). The ^{14}C peak is easily identified. The shape of the peak was found by measuring an enriched sample of ^{14}C (Fig. 18). The peak should be at a frequency which is higher than the ^{13}CH peak by a part in 1800, or about 8.5 kHz. Since the ^{13}CH peak is asymmetric (see Fig. 16), its location was mis-identified systematically by about 1 kHz, and the ^{14}C peak lies 9.5 kHz above this "apparent" ^{13}CH peak location. At the ^{14}C peak, the count rate was about 0.2 counts per minute with an ion current of roughly 10 μA .

A sample of enriched carbon was also measured in the system (Fig. 18). This sample had been enriched roughly 15 times above modern levels in a reactor, courtesy of Frank Asaro and Helen Michel. The enrichment of this sample allowed a more efficient detection setting than that in Table 1; the amplitude discrimination setting was lowered,

giving a loss factor of about 1.5 rather than 4. The measured count rate for this sample was still higher than expected by about a factor of 6. This is not completely understood; it is possible that the enrichment was higher than expected, and it may be that other loss factors were reduced from the values in Table 1 due to the lower ion currents used for the measurements of the enriched sample. At the peak, the count rate was about 15 per minute with an ion source output of roughly 4 μA .

6. APPLICATIONS AND IMPROVEMENTS

The cyclotrino offers great potential for biomedical research. Samples with enriched levels of ^{14}C can be easily measured with a 10^4 increase in sensitivity over that of conventional scintillation counting. (This conclusion is based on measurements with the OX-1 standard; a factor of 5 enrichment over natural abundance would give a count rate of about one count per minute. Since a factor of 7×10^4 enrichment is necessary to give 1 dpm for a 1 μg carbon sample by decay counting, this represents an improvement of four orders of magnitude.)

Though the device has very good sensitivity for biomedical studies, at present it is not competitive with tandem accelerators for archaeological or geological measurements. However, with improvements in the einzel lens geometry to better match beam from the ion source to the Wien filter, a factor of 5 to 10 increase in efficiency should be attainable. With improvements in the detector system a factor of about 4 should be attainable. The addition of a beam buncher should add another factor of 4. Thus, count rates of 5 per minute without a buncher, or 20 per minute with a buncher should be attainable from modern carbon. This would make the system a practical tool for archaeological and geological applications.

In order to make high precision measurements, a rapid sample changing method is necessary to cancel errors due to small changes in the transmission and detection efficiency of the system. This could be provided with a rotary sample changing apparatus as is used with many tandem accelerator mass spectrometers. Rapid sample changing could also be done by making measurements with CO_2 gas rather than graphite and by simply switching gas sources.

It would also be possible to measure a number of other isotopes with the cyclotrino [22]. Tritium should be measurable. The nearest interfering mass is different by a part in

500, so the resolution requirements are much more relaxed than for carbon and are easily attained. It will be much lower in abundance than is ^{14}C , which may lead to some difficulties with backgrounds or count rates. Normalization of tritium to hydrogen may pose some minor difficulties since there tends to be a low level background of hydrocarbons in the vacuum.

If the resolution of the cyclotron can be increased by a factor of about 10, ^{10}Be would be measurable. This resolution increase would entail operating the system at a very high harmonic (higher than 50th), which would greatly reduce the transmission of the system. (At such high harmonics the transit time effect leads to insufficient radius increase for many ions on the first orbit.) Since the natural abundance of ^{10}Be is relatively high, this reduced transmission may well be tolerable. Be does not form negative ions, so it would be necessary to accelerate a negative molecular ion (e.g. BeH^-) or to generate Be^+ with a positive ion source.

A more modest increase in resolution should allow measurements of ^{40}K and ^{44}Ti . This would require about a factor of 3 increase in resolution, which can be achieved by doubling the harmonic to 31st [22,24]. There is probably no benefit to be found to using the cyclotron for ^{40}K , since conventional mass spectrometers are quite adequate for ^{40}K measurements. There may be some applications for ^{44}Ti as a tracer, though its relatively short half life implies that the advantages of the cyclotron over decay counting are not extremely large.

The most promising isotope other than ^{14}C seems to be ^{26}Al . The abundance of ^{26}Al is about the same as that of ^{14}C , so should pose no additional problems. The nearest interfering mass is different by a part in 3700, so the cyclotron resolution would have to be doubled; this can be done by operating at about the 25th harmonic [22, 24]. Furthermore, the abundance of the nearest interfering species (^{25}MgH) should be relatively low. There will probably also be some mass-26 hydrocarbons produced (e.g. $^{13}\text{C}_2$, $^{12}\text{C}_2\text{H}_2$), but these will be at least three times as far away in mass. Production of a beam of Al^- ions is problematic; it may be necessary to use AlO^- [51]. It seems that detection of ^{26}Al should be easily achieved if adequate ion beams can be produced.

7. CONCLUSIONS

A small low energy cyclotron similar to that of Welch [12] has been combined with a high current external ion source, a beamline, and radial beam injection to detect ^{14}C at natural levels. The system's overall efficiency is about 2×10^{-4} . In its present form, it would be quite useful for measurements of samples enriched in ^{14}C ; it could improve the sensitivity of detecting ^{14}C in $1\mu\text{g}$ biomedical samples by four orders of magnitude over conventional scintillation counting techniques.

Improvements in system efficiency by a factor of 100 should be possible by redesigning the optics of the einzel lens, adding a beam buncher, and optimizing the detector geometry. With these modifications the system would be well suited to archaeological and geological applications.

The system could also be used for measurements of radioisotopes other than ^{14}C . With minor modifications, ^{26}Al and ^3He should be detectable. With further modifications, ^{10}Be and other species may be measurable as well.

A small cyclotron such as this has great potential as a mass spectrometer. It offers a huge advantage over decay counting methods for small samples. With efficiency improvements, it would be able to do archaeological and geological measurements which are now done by tandem accelerators, yet would require a small fraction of the space, construction costs, and operating costs.

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FIGURES

Figure 1 External view of the cyclotron. The ion source is enclosed in the cage on the left. The author is adjusting the dee probe. (CBB 874-3258)

Figure 2 Cyclotron system schematic, showing injection beamline. The einzel lens and first quadrupole focus beam onto the Wien filter object slit. The Wien filter pre-separates beam and provides monitoring of ion source output. The four remaining quadrupoles focus beam into the cyclotron. (XBL 874-1819)

Figure 3 Cyclotron system, retracted from magnet. The ion source is at the left, the lenses and Wien filter are inside the vacuum box in the center, and the cyclotron is at the right. (CBB 888-8200)

Figure 4 Approximate envelope of ion beam along beamline. The radial envelope (in the plane of the paper) is shown above, while the axial envelope (the plane out of the paper) is shown below. EQ stands for electrostatic quadrupole lens. (XBL 8911-4210)

Figure 5 Schematic diagram of the axially asymmetric einzel lens used in the beamline. The lens has rotational symmetry about the beam axis. (XBL 908-2807)

Figure 6 Quadrupole lens as used in the injection beamline. The beam travels on an axis connecting the two round holes. (CBB 874-3103)

Figure 7 Magnet assembly used in Wien filter, shown with end plates and end magnets removed. The field direction is vertical. Field is provided by permanent magnet material located between the pole pieces and return yoke. The entire assembly is in vacuum. (CBB 874-3087)

Figure 8 Electric field assembly used in Wien filter with front conductors removed to show construction. The electric field is produced between the top and bottom plates. Guard electrodes at the mid-potential shunt the electric field at each end and also extend along the front and rear faces to define the electric field near the magnet poles. (CBB 893-1972)

Figure 9 Mass resolution of Wien filter, measured at image slit. Some masses present in beam are labeled. (XBL 8911-4214)

Figure 10 Cyclotron schematic, showing beam injection with electrostatic deflectors and mirror. (XBL 874-1818)

Figure 11 The cyclotron shown as in Fig. 10, with the dees, deflectors and mirror visible. The central plug provides mechanical support of the vacuum envelope. (CBB 888-8210)

Figure 12 Approximate envelope of ion beam in cyclotron. The radial profile is shown above and the axial below. (XBL 8911-4174)

Figure 13 Block diagram of detection electronics. Adjustment of the single channel analyzer sets the phase window. (XBL 8911-4181)

Figure 14 Counts above threshold measured as a function of discrimination threshold. The upper curves of each set show the effects of amplitude discrimination with no phase gating, the center curves with a wide phase window (about 90°), and the lower curves with a narrow phase window (about 45°). (XBL 8911-4183)

Figure 15 Ion current as a function of radius in the cyclotron. The abrupt drop at about 10 cm is due to interception of ions by the extraction channel. (XBL 8911-4182)

Figure 16 Mass 14 hydrocarbon peaks. Operation is at the 15th harmonic with 400 V peak RF. RF frequency is along the horizontal axis, with count rate on the vertical axis. (XBL 8911-4222)

Figure 17 Carbon of modern age. Operation is at the 15th harmonic with 400 V peak RF. RF frequency offset from the ^{13}CH peak is along the horizontal axis. The vertical axis is the normalized count rate, expressed as the counts per coulomb of ^{12}C measured at the output of the Wien filter. (XBL 8911-4185)

Figure 18 Enriched sample. Operation is at the 15th harmonic with 400 V peak RF. RF frequency offset from the ^{13}CH peak is along the horizontal axis. The vertical axis is the normalized count rate, expressed as the counts per coulomb of ^{12}C measured at the output of the Wien filter. (XBL 8911-4186)

CBB 874-3258

Figure 1

XBL 874-1819

Figure 2

CBB 888-8200

Figure 3

Figure 4

XBL 908-2807

Figure 5

CBB 874-3103

Figure 6

CBB 874-3087

Figure 7

CBB 893-1972

Figure 8

Figure 9

ED = ELECTROSTATIC DEFLECTOR
EM = ELECTROSTATIC MIRROR

XBL 874-1818

Figure 10

CBB 888-8210

Figure 11

XBL 8911-4174

Figure 12

XBL 8911-4181

Figure 13

XBL 8911-4183

Figure 14

XBL 8911-4182

Figure 15

Figure 16

XBL 8911-4185

Figure 17

XBL 8911-4186

Figure 18

LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
INFORMATION RESOURCES DEPARTMENT
BERKELEY, CALIFORNIA 94720