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SUPERCONDUCTING MINI-CYCLOTRONS AS AMS INSTRUMENTS

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ABSTRACT. We have studied the limitations of conventional mass spectrometry and have examined accelerator based methods which could help circumvent these limitations. In particular, cyclotron-based accelerator mass spectrometric (AMS) techniques are discussed with an emphasis on evaluating performances of superconducting mini-cyclotrons designed for use as AMS instruments. We discussed the design of superconducting mini-cyclotrons dedicated to radioisotope dating research.

INTRODUCTION

Conventional carbon dating is based on the measurement of the beta-decay rate of the ^{14}C content of a biological sample, the age of which has to be determined. The ^{12}C to ^{14}C ratio is ca 10^{12} so that 1g of organic matter will contain ca 6 10^{10} ^{14}C atoms. The half- life for the decay of ^{14}C is 5730 years, so that statistical accuracy of 1%, *ie*, 10,000 counts, could be achieved after 12 hours of measurement. Evidently, the efficiency of the use of ^{14}C content is only $10^4/(6\ 10^6) = 1.67\ 10^{-7}$.

An alternate approach in the measurement of ^{14}C content in samples of biological origin could be based on the use of mass spectrometers. If negative ions are employed, the only mass-14 ions interfering with ^{14}C will be $^{12}CH^-_2$ and $^{13}CH^-$ since ^{14}N cannot form the negative stable ion. The former differs by 1 part in 1000 and the latter by 1 part in 2000 in mass from $^{14}C^-$. If both molecules form negative ions with the same efficiency, the ^{14}C current produced by the typical organic sample will be lower than $^{13}CH^-$ current by a factor of 10^{10}. If the current peak shapes are Gaussian, the required mass resolution should be >15,000 in order that the local yield ratio of $^{14}C^-$ to $^{13}CH^-$ at peak position of the former will be $>10^{10}$. However, the unavoidable elastic scattering of ^{12}C ions from surfaces and residual gas further complicate the experimental situation making conventional mass spectrometry practically impossible for ^{14}C dating.

ACCELERATOR MASS SPECTROMETRY

Accelerator mass spectrometric (AMS) techniques provide ultra-sensitive methods of ^{14}C ion counting. The efficiency of AMS is determined by the product of three factors: ion source efficiency $\approx 1/20$, accelerator transmission efficiency $\approx 1/2$ and beam extraction efficiency $\approx 1/10$ to 1. The typical efficiency value e = 1/400–1/40 evidently exceeds the efficiency of the technique based on beta decay measurements by a factor of 10^4 to 10^5, allowing us to reduce the weight of the sample to the order of a few tens of micrograms. The sputter negative ion source which is most frequently used, produces negative ions with a cesium beam striking the surface of the sample. For AMS using a tandem accelerator, the produced beam is roughly analyzed, and after additional acceleration for increased transmission efficiency, the beam is injected and accelerated through the first half of the tandem. In the terminal the negative ions pass through a stripper canal using noble gases (Ar), where, after stripping, they become positive ions. The removal of the electrons greatly increases the molecule dissociation probability, eliminating the molecular ion inference. After acceleration in the second stage, the relevant ion species are analyzed, passing through a series of magnetic and electric deflection elements, and are finally identified and counted in appropriate detection systems.

Cyclotron-based AMS precludes the need for a bulky system of magnetic and electric deflecting elements. Rather, cyclotrons inherently represent high mass resolution devices. When tuned for a given atomic mass, they are detuned for molecules of the same atomic mass number but different true masses, due to the acceleration phase slip. With a sufficiently high magnetic field, the cyclotron dimensions become considerably smaller (radius could be retained in the range of 10–20cm) than those of the bulky tandem machines (linear dimension of the order of tens of meters). Especially if built in the superconducting version in persistent mode, the cyclotron magnet does not need a power supply while operating. The only power consumption would be that used to operate the RF accelerating system and the ion source.

CYCLOTRONS AS AMS INSTRUMENTS

The cyclotron resonance principle is the basis for the particle acceleration used in this type of machine. Ions produced in the ion source are extracted from the internal ion source using the puller configuration, or in the case of external ion source, they are axially or radially injected in the cyclotron median plane. The trajectories of the ions of mass m, charge q and impulse p in the homogeneous cyclotron magnetic field, the center of symmetry of which coincides with the machine center, are the stable circular equilibrium orbits of radius $r = p/qB$. The frequency of the particle motion along these orbits is $\omega = qB/m$, evidently constant, which does not depend on the particle energy $E = p^2/2m$ but does depend on the particle q/m ratio. If electrodes (dees) are introduced into the cyclotron median plane and electrode RF voltage frequency equals multiple (h) of particle eigenfrequency ions of given q/m ratio could cross the electrode gap in phase with the peak of RF voltage. If we have the two-component beam and RF frequency is tuned for q/m ratio of one of the components, the other beam component will experience phase slip, Φ, determined by:

$$\sin \Phi = 2\pi \, h \, n \frac{df}{f} \tag{1}$$

while energy gain per turn of both beam components is given by

$$dE = 2N \, qV \sin\left(\frac{hD}{2}\right) \cos \Phi \tag{2}$$

where h = harmonic number, n = number of achieved particle turns, f = eigenfrequency of the reference beam component, df = difference of the beam eigenfrequencies of beam components, V = maximum RF voltage, D = angular dee width, N = number of dees. The in-phase beam component will always experience a maximum energy gain at a gap crossings while the out-of-phase beam component, after executing a certain number of orbits at the given harmonic of RF field, will experience phase slip higher than 90°, *ie*, $\cos \Phi$ will have negative value and this beam component will be decelerated and fall toward the machine center. The in-phase beam component which does not interfere with the out-of-phase component may be then accelerated up to the energies suitable for extraction and particle identification.

THE SUPERCONDUCTING AMS MINI-CYCLOTRON

A superconducting mini cyclotron is proposed here for ^{14}C dating using AMS.

Design. The cyclotron dedicated to AMS should satisfy the high-resolution requirements, *ie*, those characterized by high-resolution value $R \approx \pi hn$ numbers (Clark 1984). This requirement does not necessarily imply high energy needs, *ie*, the machine could be small, accelerating the ions in the range of the energy which allows easy detection and identification. The high bending power of superconducting cyclotrons could accelerate the particles up to 25–50 times higher energies than cyclotrons at room temperature with the same polar radii. AMS cyclotrons could have azimuthally constant fields with or without radial gradients

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since energy requirements are not too high and relativistic effects that eventually change the particle eigenfrequencies could be considered negligible. This dipole field could be produced in an air-core-type (Subotic 1984, 1988) superconducting version by two symmetrically positioned, relative to the median plane, superconducting coil sections. The zero-radial-gradient field design substantially improves the resolution of the device.

The demands for high particle turn number mean the very low RF power requirements. Typical dee voltage requirements are in the range of 100–500 V, so that relaxed design criteria for dee structure could be employed and cheap commercial components in power supply circuits may be used. The requirement of high RF harmonic number to obtain high resolution performances also works along the line of improving the machine focusing capability. Actually due to the absence of the radial field gradient and/or AVF (azimuthally varying field), the beam can suffer from a lack of focusing power which can be provided only by electrical focusing power of the acceleration electrodes. The electrical focusing power contributes to the ν_z^2 with phase focusing term (Gordon & Marti 1980):

$$h \frac{\sin \Phi}{4\pi n} \tag{3}$$

The effect of the mandatory present high turn number, n, which reduces the focusing power, thus may be compensated by the high value of harmonic number, h. This provides the frequency value of vertical oscillations high enough to produce sufficiently powerful particle focusing ($F_z \approx \nu_z{}^2 z$). This feature is also relevant in determining the upper limit of intensity (in the first n turns necessarily) of the multi-component beam. The value of this limit which becomes extremely important when an intense contaminant beam component is accelerated to partial energy along with a low intensity measured beam component, reads (Blosser & Gordon 1961):

$$I = \epsilon_0 A \omega \nu_z^2 \frac{d\Phi dE}{2\pi q} \tag{4}$$

where ϵ_0 = permitivity, A = full beam aperture, $d\Phi$ = beam phase width, dE = energy gain per turn.

The chosen parameters of the proposed superconducting mini cyclotron provided for use as an AMS instrument are given in Table 1.

Ion Source. There are two possibilities for ion sources leading to the conclusion that 1) choice of internal source makes the machine simpler and less expensive and 2) the advantage of the external source is that there is more space for sample mounting and changing. This requires the construction of suitable injection lines, which makes machine hardware complex and more expensive.

However, it appears as desirable to use an external conventional sputter-type negative

TABLE 1
Cyclotron design parameters

Resolution	$df/f = \pi$ n h = 25,000
Extraction radius	10cm
Outer coil radius	14cm
RF frequency	35,566 Mhz
Outer machine radius	18cm
Cyclotron height	29cm
Dee voltage	100-500 V
SC current density	9-45kA/cm^2
Re consumption	0.1L/hr
^{14}C eigenfrequency	1.18433 Mhz

carbon ion source to facilitate rapid sample changing and to produce the required current intensities $\approx 5\mu A$ beam of ^{12}C (Bertsche *et al* 1987). The beam injection from the external ion source into the cyclotron median plane, in the case of the Berkeley cyclotron, was accomplished radially, due to problems with field non-uniformities when the axial injection scheme is applied to small iron core cyclotrons (Welch 1984). In our case, air-core design allows the use of the axial injection scheme. The study of an off-centered axial injection scheme based on the usage of electrostatic mirror to bend the beam, previously confined respectively by solenoid field and off-centered by an electrostatic deflector, gave satisfactory results.

Cyclotron Magnet and RF Structure. Figure 1 is a schematic representation of the cyclotron. The required cyclotron magnetic field is produced by two main coil sections at current density of 9kA/cm^2. A cylindrical iron shield for effective screening of the magnetic field also provides part of the cyclotron magnetic field and mechanical support to the machine components. An axial hole 22cm in diameter provides axial access to the coil interior 22cm in diameter, and openings 5cm high are reserved for radial access to the median plane for beam extraction and RF feed.

Checking the cyclotron magnet thermal and magnetic stresses and coil net body forces, using standard procedures (Subotic 1988), shows that the design values could be retained inside the standard limits. Various protection schemes for coil quenching that retain the temperature and voltage drop inside safe boundaries are considered.

The design elements of RF configuration demands have shown that low power and dee voltage requirements permit the use of standard commercially available components and do not require using sine wave voltage (C Bieth, pers commun 1988).

Orbit Characteristics and Particle Identification. The resulting temporal change of the orbit radii for $^{14}C^-$, $^{13}CH^-$ and $^{12}CH_2^-$ species are given in Figure 2. The field parameters and harmonic numbers employed are approximately the parameters of the Berkeley AMS cyclotron (Clark 1984). This small room-temperature low-energy cyclotron has been designed and built at Berkeley for direct detection dating of ^{14}C (Bertsche *et al* 1987). The time is measured by counting executed particle turns, where each turn requires one period of closed-orbit particle motion lasting $t = 1/f = 1/1.18433$ μseconds. The particles are accelerated in a homogeneous magnetic field of 1.08 T using 180° wide dees. Peak dee voltage used was 125 V, while the RF system was operating at $h = 29$ multiple of ^{14}C ion eigenfrequency $f = 1.18333$ MHz. We can conclude that at the field level of 1.08 T and RF harmonic number $h = 29$, the $^{12}CH_2^-$ beam component reaches a maximum radius $r = 2.79$

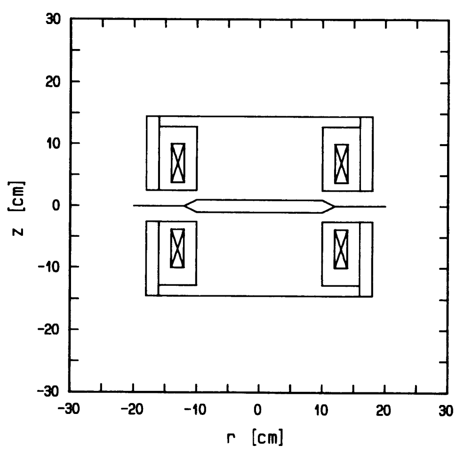

Fig 1. Schematic representation of the superconducting mini-cyclotron dedicated to AMS analysis

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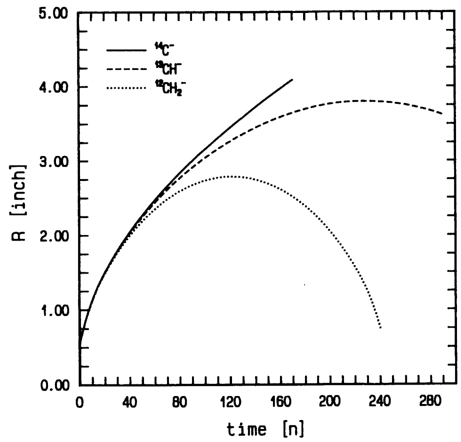

Fig 2. Changes of the orbit radii for interfering atomic and molecular species, as a function of the executed number of turns. The field parameters and harmonic numbers employed are the parameters of the Berkeley AMS cyclotron (Welch *et al* 1987; Clark 1984).

in after 121 turns, while the $^{13}CH^-$ beam component will not experience deceleration before reaching radius r = 3.8 in after 230 turns, so that ^{14}C cannot be extracted before achieving r value of 10.16 cm, *ie*, after executing 163 turns. Maximum ^{14}C energy will then be 42keV, *ie*, 3keV/nucleon. Due to low particle energy, the problem of particle detection and identification is not trivial. Various schemes of ion identification, depending on ion energy achieved and extraction procedure assumed are needed for careful analysis.

Cyclotron-based techniques achieving the highest possible level of selectivity, should be able to solve the problems of 1) rare particle identification and counting, 2) attainment of the required accuracy in determining relevant isotopic ratio at ^{14}C dating with AMS.

It is important to note that, due to low ion energies, the standard techniques of ion identification and counting cannot be applied when a small cyclotron is used at a field level of 1 T. An inexpensive low-energy ion detector concept that we intend to use, was recently developed by the Berkeley group (Friedman *et al* 1988). At this successfully tested detection system, the 36keV C^-(3keV/nucleon) ion strikes a grazing-incidence AL_2O_3 conversion dynode ejecting a few tens of secondary electrons which enter separate pores of a micro-channel plate and are independently multiplied. The output signal is proportional to the number of secondary electrons. The satisfactory background discrimination and counting efficiency at ion identification are observed. However, the operational field of the superconducting mini-cyclotron can be increased to the level of 5 T, allowing a corresponding increase of the C ion energies to the level of 1 MeV, where the standard detection methods can be applied.

The Berkeley group (Welch 1987, in press) also propose to obtain the ratio $^{14}C/^{12}C$ by tuning the cyclotron to ^{12}C and measuring the current at the detector position with a movable Faraday cup, replacing the Faraday cup with the ion detector and tuning the cyclotron to the ^{14}C ions. The ^{12}C current intensity may then be controlled, positioning the appropriate sensors along the axial injection line. The orbit off-centering techniques (Milinkovic *et al* 1988) can also be considered a possible solution in determining the $^{14}C/^{12}C$ ratio.

Raising the magnetic field (up to a level of 5 T) gives the theoretical opportunity to increase the resolution to a factor equal to the field change ratio, to move the critical radii toward the machine center and to increase the ion energy to a level of easy detection and identification. The eventual use of a stripper canal beyond the highest critical radius can also substantially improve the beam extraction efficiency.

Cost Estimation. We estimate that hardware costs could be kept within a budget of $80,000. It is also important to note that such a dedicated type of the cyclotron also could allow us to enter the field of heavy ion decay (asymmetric fission).

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