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Radioisotope Dating with the LBL 88" Cyclotron

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Since July 1976, our group at the Lawrence Berkeley Laboratory has been studying and developing the potential of the 88" cyclotron for radioisotope detection and dating[1,2,3]. This cyclotron is much larger and more powerful than required for dating, but its flexibility and ability to tune rapidly to accelerate almost any isotope has enabled us to study several different applications. We now have demonstrated the sensitivity of using the cyclotron for the detection at natural concentrations of tritium, beryllium-10, carbon-14, and chlorine-36.

The key feature that allows direct detection of low-level radionucleids is the high energy of the emerging beam, which allows particle-identification techniques to be used on an atom-by-atom basis. Without such particle identification, direct counting is impossible since unstripped background ions with the same charge-to-mass ratio always appear in the beam. For carbon-14 the main background is nitrogen-14. In order to apply the particle identification techniques the nitrogen beam intensity must be reduced to the level which will not damage or cause pile-up in the silicon detectors. When tandem accelerators are used for radiocarbon dating[4] this reduction occurs in the negative ion source. For the cyclotron we have developed a technique that achieves the required reduction after acceleration. We have built and used for this purpose a simple gas cell which completely stops the nitrogen beam while allowing the carbon-14 atoms to pass.

The separation technique makes use of the fact that the range of carbon-14 atoms is approximately 30% longer than that of nitrogen-14. Uniformity of the stopping material is essential to ensure the minimum range-straggling for the nitrogen ions. The best material we have found to meet this requirement is gas, separated from the cyclotron vacuum by a thin window. We found that 1/3 micron of gold foil will support an atmosphere of pressure difference if supported by a grid with gaps no larger than 1/3 mm (fig. 1). Gold and platinum are convenient window materials because their high atomic number Z inhibit nuclear charge-exchange interactions which can generate spurious carbon-14 atoms. For the same reason, xenon was chosen for the gas.

With the xenon gas cell we were able to eliminate completely 10 nA of ^{14}N ions. A plot of the nitrogen penetration as a function of xenon pressure is shown in figure 2, measured using a single particle detector. The curve shows discrimination to a level of 10^{-9}; in several hours of coincident detection using both ΔE and total E detection, not a single nitrogen

event penetrated the xenon cell, implying a discrimination factor for the coincident system better than 10^{-14}.

For the range separation performed in the original tritium experiment[1] a single piece of aluminum foil was used. Because of the low level of background ^3He and its much shorter range than ^3H, non-uniformities in the foil were not critical. For beryllium-10 a solid foil is probably acceptable; for our measurements the xenon cell was used. For chlorine-36 the high uniformity of the xenon cell is essential. The "thickness" of the cell in milligrams per cm^2 can be adjusted remotely by changing the pressure of the xenon gas. The range-separation technique should prove to be valuable for those using tandems as well as cyclotrons.

Except for the case of carbon-14, the levels of background radioisotopes are extremely small; the production of the radioisotopes in real time from nuclear charge exchange reactions may, however, prove to be the ultimate limit to the cyclotron approach. For carbon-14 our sensitivity at present is limited by a high level of carbon-14 within the graphite-lined tank of the 88"-cyclotron produced from years of scraping deuteron beams. The level of this carbon which finds its way into the ion source during a run has varied from 1/3 to several times the level of carbon-14 in the sample being measured. We have made dates in spite of this background by rapidly switching sample gases to allow the comparison of the unknown with a blank and a reference standard. The results of a blind measurement[2,3] of a sample are shown in figure 3. In this run the measured age (in radiocarbon years) was 5900 ± 800, with the large error dominated by the statistics of the background subtraction. The Rochester tandem group has a similar background; their ability to measure dates as old as 40 thousand years is a reflection of the fact that their background is about a factor of ten lower than ours.

In the carbon-14 measurements the carbon was conveniently introduced to the ion source in the form of CO_2 gas. The efficiency for acceleration of the $^{14}C^{3+}$ ions could be varied between 10^{-5} and 3×10^{-4}; in a typical run it was 3×10^{-5}.

We have made nearly a dozen successful "nonblind" tests, and the successful "blind" test shown in the figure. More recently, however, we made a blind measurement in which our answer differed greatly from the answer achieved by decay-dating: we misestimated the age of an 8,000 year old sample dated by Rainer Berger to be 18,000 years old. This was the only colligen sample we have measured, and we suspect that impurities in the CO_2 affected the performance of the ion source, changing the level of background appropriate to subtract. The best hope for the elimination of such systematic errors lies in the elimination of the background. The easiest way to achieve this will be to use an external ion source which can pre-accelerate the ions to tens of keV before they enter the contaminated tank of the cyclotron. The design and

construction of such a source is now underway by R. Gough and the staff of the 88" cyclotron, and we expect it to be operational later this year.

In our ^{10}Be measurements no comparable background has been detected. We have measured $^{10}Be/^{9}Be$ ratios in beryllium metal at the 10^{-9} level, comparable to that expected in sea floor sediments. We observed 800 ^{10}Be per second in our detectors, and the background was less than one count in five minutes. Our sensitivity for ^{10}Be is better than 10^{-14}, justifying all the optimism expressed in reference 1. A comparable sensitivity was achieved when BeO was substituted for beryllium metal in our Penning Ion Gauge (PIG) sputter source, but we have not yet achieved an accurate date with BeO.

For ^{36}Cl our demonstrated sensitivity is 10^{-12} for the ratio $^{36}Cl/Cl$. This sensitivity is sufficiently good to make ^{36}Cl an attractive tool for measuring the age of water in potential nuclear waste storage sites, useful in determining the geologic isolation of the deep site from the surface. Further details of our work with ^{10}Be and ^{36}Cl will be presented at this conference by Terry Mast.

After only one and a half years of development, the accelerator approach has already proven its potential for several radioisotopes. Except for the case of carbon-14, there have been no unanticipated backgrounds, and we are optimistic that the full predicted[1] sensitivity of the accelerator approach will soon be utilized.

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Figure 1. The thin foil separating the xenon in the range cell from the vacuum of the cyclotron is supported on a tungsten grid, with hexagonal openings 0.33 mm in diameter. This grid is only 25 microns thick, and is supported in turn by a heavier 100 micron-thick tungsten grid with hexagonal openings 0.1 mm in diameter. The entire structure gives a clear aperature of 65%, over which 0.3 micron-thick gold foil can support a one atmosphere pressure difference.

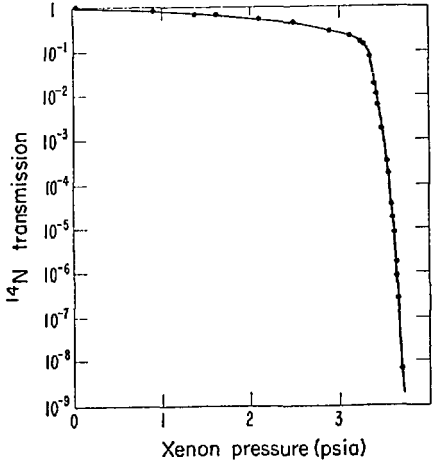

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Figure 2. Nitrogen-14 transmission through the xenon cell as a function of xenon pressure. The nitrogen-14 was detected with a single ionization chamber with a threshold energy loss of 0.8 MeV. When coincident detectors were used, the discrimination against nitrogen was found to be better than 10^{14}.

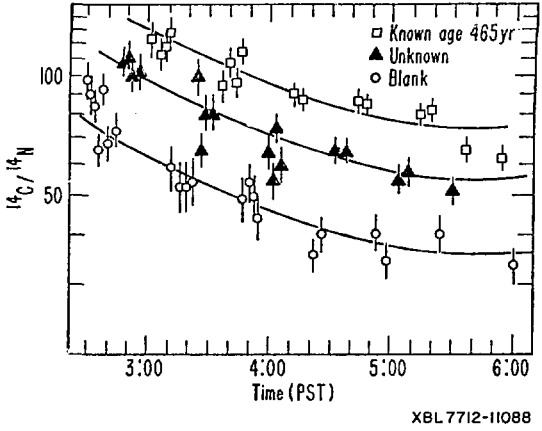

Figure 3. The $^{14}C/^{14}N$ ratio as a function of time, for a blind sample (2,3). The carbon-14 was measured with an ionization chamber, silicon-detector telescope. The nitrogen-14 was measured by integrating the current from slits which collimate the beam soon after it emerges from the cyclotron and well before it enters the xenon gas cell. Three samples were alternated: one of known age 465 years; a sample known to contain no carbon-14; and the unknown. The fit corresponds to an age of 5900 years, where the "standard" radiocarbon half-life of 5570 years has been assumed.

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Technical Considerations for Dedicated Systems—the Cyclotron*

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With the recent detection of low-level ^{3}H, ^{10}Be, and ^{14}C beams from cyclotrons [1—5], it became clear that the cyclotron is capable of measuring the radioisotope concentration in a naturally occurring sample. In these experiments, the radioisotopes were extracted from an ion source discharge containing material from the sample and accelerated as positively charged ions. The principal background was the simultaneous acceleration of ions with the same mass value, e.g., ^{3}He, ^{10}B, and ^{14}N. In each case, the beam was sent through an absorber that removed the background ions and allowed the radioisotope ions to pass through and be detected. By placing charged particle counters directly in the beam line behind the absorber, beams of radioisotopes were detected at levels as low as 1 part in 10^{14} of the primary cyclotron beam.

The chief obstacle to radioisotope dating by conventional mass spectrometry has been the elimination of background [6]. In addition to stable isotopes of the same mass, these backgrounds have included molecular ions and particles of other masses scattered from residual gas in the spectrometer. These backgrounds can be eliminated with the cyclotron because of two features. First, the mass resolution of the cyclotron [7] is sufficiently narrow that the extracted beam is composed purely of atomic ions of the desired mass. During acceleration, ions must travel for several hundred turns in phase with the oscillating dee voltage in order to reach full energy and be extracted from the machine. Ions of different masses or ions created by collisions in the residual tank gas drop quickly out of phase and are lost from the beam. For radioisotopes other than tritium, the high charge state of the beam prevents the formation of a molecular component. Thus with the cyclotron, the only significant backgrounds are stable isotopes with the same mass.

The second feature of the cyclotron, the high energy of the emerging beam, can be used to remove the stable isotope background. The energetic ions from the beam are sufficiently stripped in passing through matter so that differences in their nuclear charge creates differences in their rate of energy loss. Ion counter telescopes based on this principle can distinguish adjacent elements with some sensitivity [8]. For more intense backgrounds of elements whose proton number is greater than that of the radioisotope, the background may be removed in a sufficiently uniform absorber [1—5, 9].

The discussion here will be concerned primarily with the extension of the results [5, 9] for ^{14}C-dating on the Berkeley 88-inch cyclotron

to the dating of other radioisotopes. Design features for a cyclotron dedicated to radioisotope dating will be presented. The discussion will be organized around two important problems: elimination of the stable isotope background and the calibration of the radioisotope beam current.

Above some threshold energy for each radioisotope, the separation of background becomes easy. With some estimate for that energy, the specifications of the cyclotron and ion source required to date samples are clear. The size of the cyclotron and ion source can be described by one quantity, Kq^2, where K, the cyclotron constant, is given by $B^2r^2/2$ and q is the ion charge. The quantities B and r are the average magnetic field and radius at beam extraction. This size parameter is equal to the product of the energy and mass of the ions in the beam [7]

$$Kq^2 = Em.$$

For example, the Berkeley cyclotron has K = 140 MeV-amu. The production of ^{14}C beam is intense up through the 3^+ charge state, giving Kq^2 = 1260 MeV-amu. This corresponds to a maximum ^{14}C energy of 90 MeV, much more than the 40 MeV actually used to obtain a date [5]. This experiment could be duplicated on a cyclotron of Kq^2 = 560 MeV-amu. With an ion source that produced intense beams on even higher charge states, an even smaller and less expensive cyclotron would be sufficient.

The present work [9] on ^{14}C-dating with a cyclotron used a beam energy of 40 MeV and a xenon gas cell absorber. At this energy, the range [10] of ^{14}C is about 30% longer than that of ^{14}N. Range straggling of the nitrogen ions reduces this difference to about 20% [ref. 9]. The residual ^{14}C energy is about 4 MeV, sufficient to be counted in a gas ionization counter telescope [9]. Fig. 1 shows how the conditions change with lowered beam energy. The ratio of the carbon range and the nitrogen range falls [10]. Operating with an absorber thickness greater than the carbon range will substantially reduce the counting efficiency. At the same time the range which is unavailable for counting because of nitrogen straggling increases [11]. The curve in fig. 1 is an upper limit, since it may be possible to further reduce the straggling with more uniform gas cell windows. Because the nitrogen background is reduced by such a large factor (10^{14}), a small reduction in the nitrogen background will not substantially affect the straggling. With reduced beam energy, the residual ^{14}C energy shown in fig. 1 drops. Much below 1 MeV, it is difficult to identify the Z of these ions unambiguously. It will be difficult to conduct effective range separation for ^{14}C at energies much below 20 MeV (Kq^2 = 280). The cyclotron required for this task is modest (K = 30, q = 3) and available commercially [12].

An upper limit on the range separation is set by the energy at which the ions in the beam begin to react with absorber nuclei. This energy is denoted by the vertical line in fig. 1, and bounds a trianglar region in which useful measurements are possible. As the mass of the radioisotope increases, the energy of this operating triangle goes up and its size decreases.

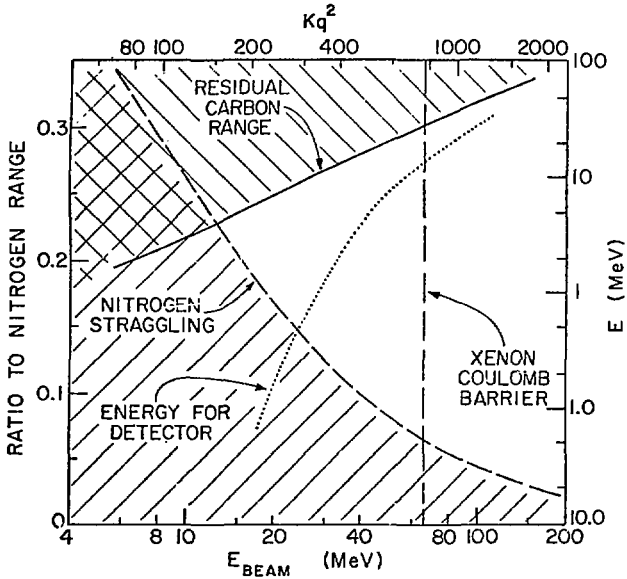

Fig. 1. Constraints on range separation of ^{14}N from ^{14}C as a function of beam and energy, E_{BEAM}, and cyclotron system size, K_q^2. Limits are normalized to the nitrogen range at each energy. The upper limit is set by the carbon range remaining after the nitrogen has stopped, and the lower limit is set by the straggling of nitrogen ions. As operating conditions determined by beam energy and absorber thickness enter the shaded regions, separation becomes more difficult. The energy loss of carbon ions travelling between the two limits is given by the dotted line. At beam energies above the vertical dashed line, the N + Xe coulomb barrier, background will appear from reactions in the absorber.

The next natural radioisotope whose proton number is less than its stable counterpart is ^{39}Ar. If the background at this mass is about as intense as the ^{14}N component in the ^{14}C beam, then the operating triangle has nearly vanished. However, it is likely that the background from ^{39}K is much smaller, and it may be possible to make the separation directly in a counter telescope without reducing the background intensity in an absorber.

A counter telescope distinguishes ions of different Z by measuring differences in their stopping power dE/dx. Fig. 2 shows values of the stopping power [10] as a function of ion energy for elements between Z = 10 and Z = 30. For low energies, the average ionization in passing through matter is low. The nucleus is well screened, stopping power differences among elements are reduced, and the curves tend to cluster along one locus. As the Z increases, greater energy is needed to operate in a region where the separation of neighboring elements can be made effectively. The Kq^2 required to reach points of increasing energy and mass is indicated in fig. 2. Dating with heavier radioisotopes requires more powerful cyclotrons. A cyclotron with $Kq^2 = 300$ is still adequate for ^{26}Al. The Berkeley cyclotron can handle elements well beyond Z = 30, especially since the ion source production [13] is still intense for heavier elements at charge states of 4^+ or 5^+, giving $Kq^2 > 2000$.

To achieve separation in an ion counter, the count rate for background cannot greatly exceed that of the desired radioisotope. The internal ion source [14] in use at the Berkeley cyclotron is exposed to an environment that is difficult to clean and, because of bombardment by energetic particles, also contains substantial concentrations of radioisotopes [9]. This background is likely to be substantially less in an external heavy-ion source where the environment can be more rigidly controlled.

Once the radioisotope beam is detected unambiguously, dating depends on relating the radioisotope count rate to the isotope ratio in the original sample. The cyclotron accelerates only one mass at a time, and direct measurement of an isotope ratio at the cyclotron exit means varying either the main magnetic field strength or the dee frequency to measure individually each beam current. Even so, the acceleration efficiency is mass dependent [7] and must be calibrated with a known sample under similar ion source and accelerator conditions. Where fast sample changing is possible [5], a direct comparison of the radioisotope count rate for known and unknown samples can be made without reference to another beam. With either method, sufficient measurements must be made to unfold the time variation of the ion source output.

A direct measurement of the isotope ratio in a sample involves changing the cyclotron resonance and measuring the external beam current for each isotope. Care must be taken in shaping the main magnetic field so that both beams are accelerated efficiently [15]. Since external beam focusing and steering are usually magnetic, it is easier to hold the main field constant

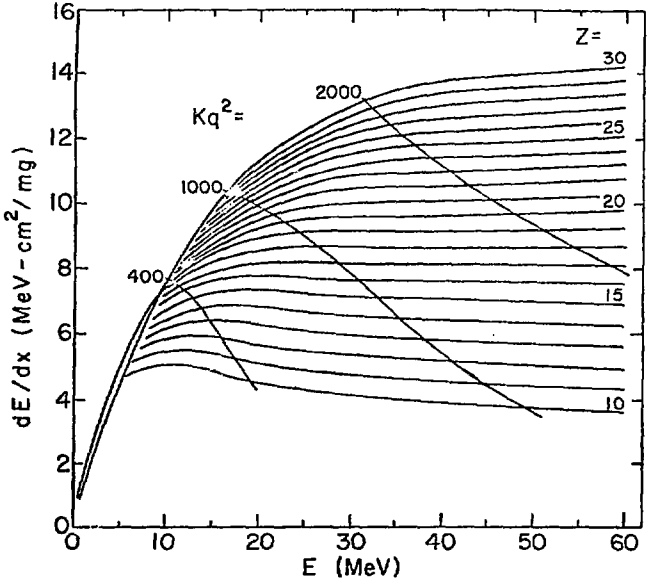

Fig. 2. Relationship between stopping power and energy in a charged-particle counter for elements between Z=10 and 30. The energy required for good separation of adjacent elements increases with Z or mass. The size of the cyclotron system required to reach points of good separation is given by contours of constant Kq^2.

(preserving the beam rigidity) and to vary the dee frequency [7]. This technique has been successfully employed in the development of ^{3}H-dating and ^{10}Be-dating [2–3].

With gas samples, rapid sample changing has been used [5,9] to compare the radioisotope count rate for known and unknown samples. When the perturbations from sample changing were made sufficiently small, the ion source conditions remained stable during the change. Care was taken in the design of the gas manifold to reduce cross-sample contamination by keeping working volumes small and free of dead space, and by flushing with each sample change. A similar technique could be developed for solid feed sources, but would probably require the unrestricted space of an external ion source.

In addition to background reduction, an external ion source would permit direct monitoring of the ion source output. Aside from occasional discharges at the dee, variations in the external beam current are due almost entirely to variations in the beam from the ion source. With magnetic analysis, isotopes from the sample other than the radioisotope could be separated and their current measured. This would avoid problems associated with monitoring the background beam [5,9], which may be uncorrelated with the beam from the sample.

The elements of a cyclotron based radioisotope detection system are shown in fig. 3. An external Penning Ion Gauge source [13] is used because it produces high intensities of high charge state beam for reasonable operating times. The magnetic field required by the Penning Ion Gauge source can also analyze the extracted beam for monitoring. Although somewhat more complex to operate, this external ion source configuration provides ease of access and minimal background. Transmission efficiency into the cyclotron can be significantly improved by bunching the beam so that it is in phase with the dee frequency. The size of the cyclotron is determined by the maximum charge state obtained in the ion source and the mass of the radioisotope being detected. The bending magnet after the cyclotron serves to remove any beam scattered from the deflector channel at the cyclotron exit. The radioisotope beam is focused onto a detector which may be preceded by an absorber to remove any stable isotope background. The initial cost of such a facility is dominated by the cost of the cyclotron, and would be substantially lower if such a system were added to an existing machine.

The sample size for cyclotron dating is substantially less than that required for decay counting. Consider the ^{14}C case an an example [5,9]. The ion source feed was about 1 cc/min, or 3×10^{7} atoms/min of ^{14}C at the natural level. The count rate for ^{14}C was about 10^{2}/min, giving an efficiency of 3×10^{-6}. A factor of 10 was lost both in the ion optics and in multiple scattering from the absorber. In addition, the beam current was suppressed to reduce background. Thus a better design of the system with present techniques can raise the efficiency to nearly 10^{-3}. If the sample is examined

CYCLOTRON RADIOISOTOPE DETECTION SYSTEM

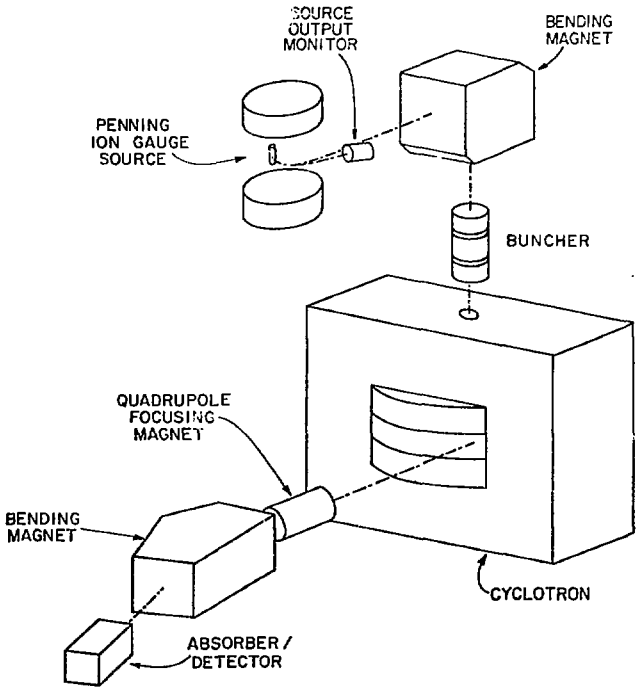

Fig. 3. Elements of a radioisotope dating system employing a cyclotron. The function and properties of the various elements are discussed in the text.

for 1 hr, 30 mg of carbon are consumed, less than 1% of the amount required by decay counting techniques. Sample sizes and efficiencies for other low mass radioisotopes should be similar. The first ^{14}C date with a cyclotron was obtained in 3.5 hr of running time. Automation of data collection can substantially reduce this time, so it is reasonable to consider a system capable of dating more than 1000 samples/yr.

Experiments with low-level radioisotope beams have shown that the cyclotron can be used as the key element in dating systems for a broad spectrum of radioisotopes. For the lighter masses, the size and complexity of the machine can be substantially reduced from those on which initial studies were done. The resonant acceleration of the cyclotron eliminates all backgrounds from the beam except other stable isotopes with the same mass. The remaining background can be removed by selective absorption or discrimination in a charged particle counter, leaving an unambiguous radioisotope signal. The radioisotope count rate is converted into a concentration, or date, by comparing it sequentially with beams of other isotopes or the radioisotope beam from a known sample. The cyclotron system can be developed as an efficient detector of radioactive atoms, useful in dating small samples or samples many half-lives old.

The development of radioisotope dating with accelerators has occurred at a time when the relevant ion source, accelerator, and detector technology existed as part of the heavy ion research effort in nuclear physics. As the heavy ion technology improves, the scope of radioisotopes easily accessible for dating will enlarge. Of particular interest for the future are EBIS [16] and ECR [17] ion sources that offer beams on very high charge states, and cyclotrons using superconducting magnets [18] that will accelerate the heaviest ion efficiently. Within the next few years, these improvements should become available for radioisotope dating.

Footnotes and References

*Work supported by the U. S. Department of Energy.

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Plans for Radioisotope Dating With the Lawrence Berkeley Laboratory 88-inch Cyclotron

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The use of the Berkeley 88-inch cyclotron as an ultra-sensitive mass spectrometer for radioisotope dating has been discussed in other papers of these proceedings by Muller and Stephenson and in References 1-3. At the present time our plans for the future are not well defined. We have been making a series of exploratory tests on a number of different isotopes; ^{14}C, ^{10}B, ^{36}Cl, and ^{81}Kr. The future direction of our program will depend on the results of these tests. I will describe here some of the results obtained and indicate briefly the work we see needed to make the cyclotron a useful analytical tool for measuring these isotopes. I want to emphasize the preliminary nature of the results; most of the conditions have not been optimized. We are anxious to hear from others about the avenues they think would be worth exploring.

^{14}C. The use of the cyclotron as a mass spectrometer has been discussed by Ed Stephenson. Our detection scheme and results for ^{14}C were summarized by Richard Muller. As they indicated we are waiting the construction of an external ion source to continue with radiocarbon dating. It is expected the source will be finished this year. In addition to the elimination of the background ^{14}C we foresee a number of other benefits the source can provide. It will allow an independent monitor of the source output and perhaps a monitoring of the ^{12}C and ^{13}C currents. An improved vacuum in the ionization region will reduce the ^{14}N current and thus remove the possibility of ^{14}N interactions in gas cell impurities giving rise to a very low level ^{14}C signal. The external ion source may also allow recycling of un-ionized CO_2. Improvements will be made in the gas handling system which will allow measurements to be made with 10 mg samples. Finally a continuous monitor of the ^{14}N current measured at the gas cell along with the monitor of the source output will provide means for removing the systematic effects of variations in source output and acceleration efficiency. Without the presently large ^{14}C background a precise measurement could be made in 10 to 20 minutes. Thus the long term variations of the source output that we've seen in the past will be reduced considerably and compensated more accurately.

^{10}Be. We have made a number of tests of the acceleration and detection of ^{10}Be. With a half-life of 1.5×10^6 years ^{10}Be will be useful for studying sedimentary rocks, manganese nodule formation, and other geochemical problems. Measurements of marine sediments, manganese nodules, meteorites, and lunar rocks have yielded $^{10}Be/^{9}Be$ ratios in the range from 10^{-8} to 6×10^{-7}. Measurements requiring smaller sample sizes will allow more detailed studies; for example of variations in sediments during a single magnetic field reversal and correlations with climatic variations.

In the tests thus far we have accelerated $^{10}Be^{2+}$ to about 40 MeV. Although there are a large number of possible contaminating isotopes with charge to mass ratios close to 2/10 the frequency resolution of the cyclotron is sufficient to eliminate all but ^{10}B. If the levels of ^{10}Be and ^{10}B were comparable they could also be separated although the fractional mass difference is 1 in 17000. (Figure 1 shows the ^{10}Be and ^{10}B intensity as a function of cyclotron frequency.) However, the ^{10}Be is many orders of magnitude less abundant in the beam than ^{10}B and is thus obscured by the tail of the ^{10}B distribution. We have completely eliminated the ^{10}B using the same range separation technique developed for $^{14}C/^{14}N$. The separation is easier here since the ^{10}Be range is 50% longer than that of ^{10}B. When the gas cell pressure is set midway between the two ranges the ^{10}B is completely absorbed and only beryllium is observed in the detector telescope.

Beryllium was introduced into the ion source (a PIG source) both as a metal and as beryllium oxide. In both cases the material was placed at the back of the source opposite the ion exit slit. Krypton gas was used to support the plasma arc. Atoms are sputtered from the sample by ions accelerated through the slit back into the source during the "wrong" polarity cycle of the r.f. and also by electrons in the plasma. The BeO samples were introduced in the form of pellets of compressed tantalum powder (90%) and BeO (10%).

The first tests were with samples of beryllium metal. An ordinary piece of beryllium metal gave no counts in 5 minutes. A piece which had been exposed to a deuteron beam gave 800 counts per second. The counts were normalized to ^{9}Be (about 2 microamps) obtained by changing the cyclotron frequency and exit deflector voltage. The ratio of $^{10}Be/^{9}Be$ was about 7×10^{-10} in agreement with that expected from the deuteron irradiation. The observation of no counts from the unirradiated sample indicates a potential sensitivity of $^{10}Be/^{9}Be \sim 10^{-14}$. No attempt was made to accurately determine the amount of sample used. However, after the measurement there was almost no visible change in the metal and we estimate less than 50 mg of Be were consumed.

In principle ^{10}B could be used instead of ^{9}Be to normalize the ^{10}Be counts. If the BeO sample were seeded with a known level of boron (which dominated the background boron)

then the ^{10}B current could be integrated simultaneously with the ^{10}Be counts. The advantage would be the ability to leave the cyclotron conditions completely unchanged during a measurement. The disadvantage is that the different source efficiency for boron and beryllium would have to be calibrated and may be larger than that difference for ^{9}Be and ^{10}Be. Although an initial attempt to use this technique has failed we feel further development work could prove it to be useful.

Further tests were made with BeO samples supplied by Professor J. Arnold of U.C. San Diego. These included an N.B.S. standard, the standard diluted by a factor of 10, and BeO from a meteorite. They had been measured in his laboratory by decay counting and contained $^{10}Be/^{9}Be$ levels of 7.3×10^{-8}, 7.3×10^{-9}, and 1.9×10^{-9} respectively. We observed the correct relationship among the samples to within 25%. However, the absolute $^{10}Be/^{9}Be$ ratios were all too low by a large factor of about 50. At the present time we do not understand this discrepancy and will be repeating the measurements with additional cross checks soon. Again no quantitative measurement was made of the amount of sample consumed. We estimate less than 5 mg of Be were used.

We find these initial results very encouraging. There is obviously much work to be done to turn the procedure into a reliable analytical tool. The initial work will be to improve the ease and speed of changing samples in the source so that knowns and unknowns can be readily compared. Ideally we would like to find a way to handle the beryllium in a gaseous form which is more reliably controlled in the ion source. We will soon add a direct continuous monitor of the ^{10}B current to provide a monitor of source variations. Then a large number of samples will be measured to establish the accuracy and reproducibity of the technique.

We are also encouraged by news that a group working at the Grenoble cyclotron[4] has successfully measured $^{10}Be/^{9}Be$ levels between 10^{-10} to 10^{-8} in samples of BeO. They have used the range separation technique to eliminate ^{10}B and have normalized to ^{9}Be.

^{36}Cl. During the past two weeks we have made some initial runs on ^{36}Cl. It has a half-life of 308,000 years and has been measured in meteorites at $^{36}Cl/Cl$ concentrations from 10^{-9} to 2×10^{-8}. Recently there is interest in measuring ^{36}Cl in ground water to study the hydrology of potential radiactive waste repositories. Concentrations of $^{36}Cl/Cl$ at levels of about 10^{-11} have been measured in a few samples of water.

We have introduced chlorine into the ion source in a number of different forms. Following earlier cyclotron experience BCl_3 and Cl_2 gas were used. Although microampere levels of ^{37}Cl beams were achieved we feel the low vapor pressure of BCl_3 and the corrosive nature of Cl_2 make them inappropriate for measuring a large number of samples.

We have also tried sputtering NaCl from a matrix of tantalum and have had difficulty so far achieving an intense beam. We have finally turned to Freon-12 which is relatively benign and can be readily controlled with the same gas system we use for CO_2. An alternative which is perhaps more easily produced from natural sources of chlorine would be methyl chloride.

There are two isotopes, ^{36}Ar and ^{36}S, which are not separated from ^{36}Cl by the cyclotron resonance condition. The high Z makes the separation difficult. We have accelerated $^{36}Cl^{5+}$ to 72 MeV and the range of the ^{36}Cl is only 6% longer than that of ^{36}Ar. The ^{36}S range is similarly longer than the ^{36}Cl range. Nonetheless the xenon cell has allowed separation of these isotopes. Instead of using the cell to completely absorb a species as was done for ^{14}C and ^{10}Be we simply use it to degrade the beam and allow the different stopping powers to separate the elements. Figure 2 shows a scatter plot of the events as a function of the energy loss and energy observed in the detector telescope. There is a large clustering of argon events, a small number of sulfur events, and a gap where ^{36}Cl is expected. This separation has been sufficient to look down to the level of about 10^{-12}. We have not detected ^{36}Cl in the samples measured thus far. The normalization was made by tuning to ^{37}Cl.

The future program will include the choice of a good sample gas, reduction of the argon level, improvements in detector resolution, and measurements of samples with known ^{36}Cl concentrations.

^{81}Kr. We have to date used the cyclotron as a mass spectrometer on krypton only to measure the isotopic abundances of the stable isotopes.[2] ^{81}Kr has a half-life of 200,000 years and occurs in pre-bomb testing air at a $^{81}Kr/Kr$ concentration of about 10^{-13}. This low concentration and the difficulty of separating ^{81}Kr from ^{81}Br mean the development problems will be more difficult for ^{81}Kr. We plan to make some tests to get a quantitative estimate of the level of the ^{81}Br contamination. However, further work will probably not continue until we have gained more experience with ^{10}Be and ^{36}Cl.

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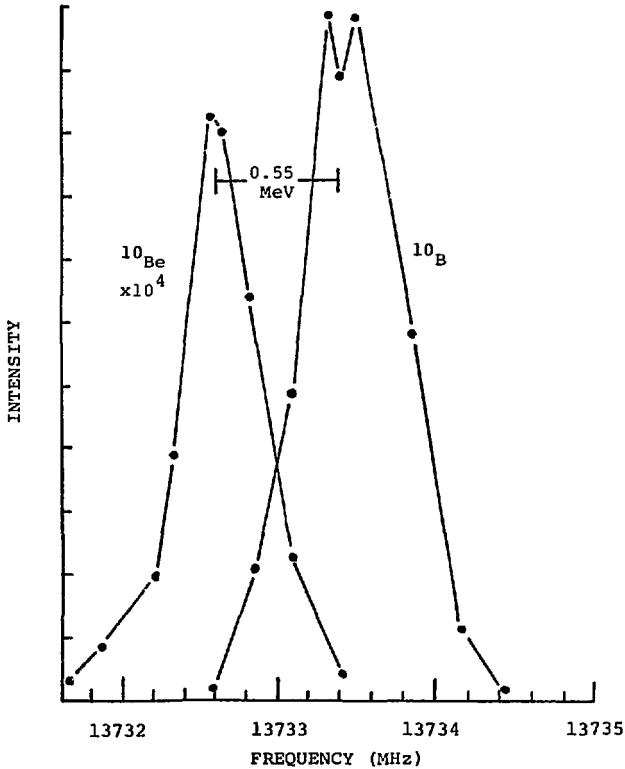

Figure 1. Intensity (arbitrary units) as a function of cyclotron frequency (MHz) for ^{10}B and ^{10}Be showing the 0.55 MeV mass difference. The ^{10}Be counts were obtained with the ^{10}B absorbed in the gas cell.

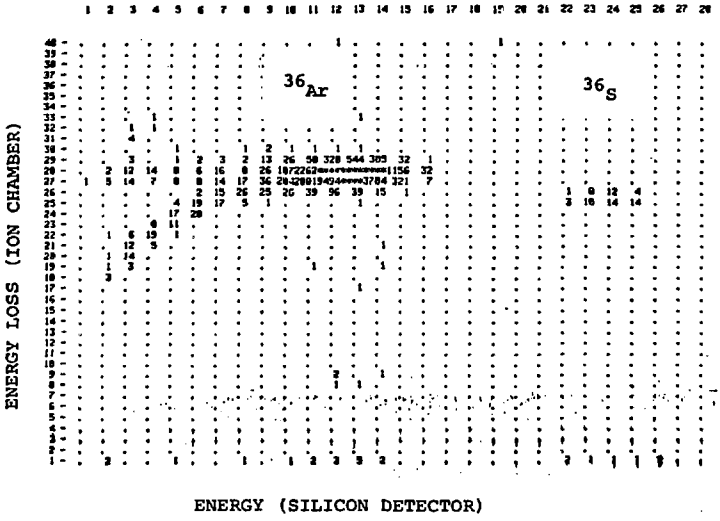

Figure 2. Events as a function of energy loss and energy measured in an ion chamber and silicon detector respectively. The original 72 MeV beam has been degraded by the gas cell and the different stopping powers have separated the ^{36}Ar and ^{36}S. If it were present ^{36}Cl would appear between the two clusters.