

Radioisotope Detection with Accelerators

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ABSTRACT

High energy mass spectrometry is a new and very sensitive technique of measuring rare radioisotopes. We describe the techniques used to select and identify the individual radioisotope atoms in a sample and the status of the radioisotope measurements and their applications.

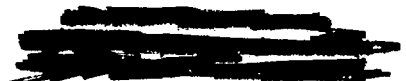
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1. HIGH ENERGY MASS SPECTROMETRY

A new method of measuring radioisotope concentrations is beginning to have a major impact on the field of radioisotope detection. The method consists of directly counting individual radioisotope atoms that have been accelerated to high energies. The high energy allows the identification and counting of the individual atoms in the beam. As a result, this method has much greater sensitivity than the standard method which detects only the tiny fraction of atoms that decay during the counting period. The improved sensitivity allows one to use much smaller samples and to measure much greater ages than were possible with the decay-counting technique.

Radioisotope dating for many isotopes is performed using proportional counters to detect the electron (beta particle) emitted from radioactive decay in the sample. The sensitivity of this method is limited in practice by radioactive backgrounds caused by penetrating cosmic rays, by the radioactivity of the materials used in the construction of the detector and also by the statistics of the finite number of detected atoms. The measured radioactivity is then used to infer the radioisotope abundance, often with respect to a standard. This fact can then be

used to interpret the age or history of the sample, or alternatively, if the age is known, to measure such things as the past cosmic ray flux that created the isotope. However, it has been long recognized that counting the radioisotope atoms directly, rather than waiting for their decay, is potentially a much more sensitive means to determine their concentration.

Table 1

isotope	halflife	atoms to produce one decay/hour	terrestrial abundance *	interfering isobars
^{10}Be	1.5×10^6	2×10^{10}	10^{-8} to 10^{-12}	^{10}B
^{14}C	5.7×10^3	7.2×10^7	10^{-12} to 10^{-15}	^{14}N
^{26}Al	7.3×10^5	9×10^9	10^{-12} to 10^{-14}	^{26}Mg
^{32}Si	2.8×10^2	3.5×10^6	10^{-15} to 10^{-17}	^{32}S
^{36}Cl	3×10^5	3.8×10^9	10^{-12} to 10^{-14}	$^{36}\text{A}, ^{36}\text{S}$
^{39}Ar	2.7×10^2	3.4×10^6	10^{-15}	^{39}K
^{41}Ca	1.3×10^5	1.6×10^9	10^{-14} to 10^{-15}	^{41}K
^{53}Mn	3.7×10^6	4.7×10^{10}		^{53}Cr
^{59}Ni	7.5×10^5	9.6×10^9		^{59}Co
^{81}Kr	2.1×10^5	2.7×10^9	10^{-12} to 10^{-13}	^{81}Br
^{129}I	1.6×10^7	2×10^{11}		^{129}Xe

* compared to the stable isotope

The detection by radioactive decay of the isotopes in table 1 requires long counting times and/or large samples. The efficiency of decay counting can be defined as the number of hours of counting multiplied by the chance that the beta particle is detected (often close to 1) divided by the number of atoms required to produce one decay per hour (third column

table 1). Even if the efficiency of direct counting is low (10^{-3} - 10^{-5}) it is still orders of magnitude better than the efficiency of decay counting, especially for the longer half-lives. A larger efficiency allows the same number of atoms to be detected with a smaller sample. The time required for one measurement depends on the magnitude of the beam current that can be extracted from the ion source. For instance, for ^{14}C the samples can typically be a 1000 times smaller than is required for conventional decay counting and a measurement can take less than an hour.

For some radioisotopes (especially those used in isotope geology) this direct detection has been achieved with conventional mass spectrometers. This has been possible when the potential background atoms (stable isotopes of the same or similar mass to the radioactive ones) have a sufficiently low concentration in the sample. However for many radioisotopes of interest the natural concentrations are low, 10^{-12} to 10^{-16} . The very low peak of the radioisotope in the mass spectrum is obscured by the abundant stable isotope distributions and conventional mass spectrometry cannot be used.

The key idea in the new technique which allows the isotope separation is the acceleration of the sample atoms to high energy (multi-MeV). With the high energy beams provided by accelerators the separation and identification of the isotope of interest is readily made using standard techniques developed for nuclear physics. The ionization loss (dE/dx) of a high energy ion in matter provides a measure of the nuclear charge Z . When combined with magnetic or electrostatic analysis the required radioisotope separation and identification can be made.

In principle this new "high energy mass spectrometry" can be used for all isotopes (Purser 1977). In practice, however, its most interesting application is to radioisotopes which occur with low natural concentrations. For most stable isotope measurements (and some special radioisotope samples) the less complex and fully developed conventional mass spectrometry is satisfactory.

Many groups have begun developing high energy mass spectrometry since the realization of the idea three years ago. Descriptions of the work leading to the first measurements are given in recent reviews by Muller (1979), Bennett (1979), Purser (1979) and by Mast and Muller (1980). There has been a topical conference on the new method (Gove 1978). Already nine groups have made sensitive measurements with virtually unmodified existing accelerators (table 2). Two kinds of accelerators have been used to provide the high energy: cyclotrons and tandem van de Graaff accelerators. The initial research applications of the technique have now produced measurements with samples over a thousand times smaller than previously required. The development work has primarily used ^{10}Be , ^{14}C , ^{26}Al , and ^{36}Cl with the first actual research applications using ^{10}Be and ^{36}Cl . The isotope separation and identification techniques used for each radioisotope depend on the background atoms, and these in turn depend on the specific accelerator which is used.

Table 2

isotope	groups	sensitivities achieved
^{10}Be	W, Y, R, G, B	10^{-14}
^{14}C	W, A, O, R, C, B, S	3×10^{-16}
^{26}Al	A, R, G	10^{-14}
^{32}Si	A	10^{-14}
^{36}Cl	A, G, R, B	2×10^{-16}
^{129}I	R	11^{-11}

A Argonne National Laboratory
B Lawrence Berkeley Laboratory
C Chalk River
G Orsay-Grenoble
O Oxford
R Rochester, General Ionex, Toronto collaboration
S Simon Fraser-McMaster
W University of Washington
Y Yale

2. TECHNIQUES, LIMITATIONS AND ERRORS

The operation of the high energy mass spectrometer involves the following basic stages.

- ionization of the sample in an ion source
- acceleration of the isotope of interest
- separation in the high energy beam of the radioisotope from intense stable backgrounds
- identification of individual atoms

These stages provide a measurement of the radioisotope atoms. To convert this number to a concentration in the sample it must be normalized with respect to a stable isotope in the sample. The techniques for

making this normalization are described below for each type of accelerator.

A schematic of the basic components of a spectrometer using a cyclotron is shown in Figure 1. The components for a spectrometer with an electrostatic accelerator are shown in Figure 2.

The cyclotron both accelerates the ions and also serves as a high energy mass spectrometer (Stephenson 1977). The cyclotron resonance condition gives a mass resolution function with exceedingly low tails. Tuning the dee voltage in frequency with a fixed magnetic field shows a remarkable mass selection. For example only 1% in frequency away from the resonance producing a 10 microampere beam of $^{12}\text{C}^{3+}$ there are no accelerated particles observed in an hour. This clean mass selection is a consequence of the 100 to 200 turns made in the cyclotron during acceleration. Ions with the wrong charge to mass ratio (either from the source or from charge exchange in the residual gas) drop quickly out of phase and are lost from the beam. Thus when the frequency is tuned for $^{14}\text{C}^{3+}$ the only other ion accelerated is $^{14}\text{N}^{3+}$; no high energy ^{12}C , ^{13}C , ^{15}N or other ions appear in the beam. The simultaneous measurement of the total energy of the particle and its energy loss per unit path length through matter which depends on the nuclear charge Z , enables one to distinguish between ^{14}C and ^{14}N . However, the ^{14}N background is too intense for the detectors and the ^{14}N has to be filtered out before the beam enters the detector. This can be done for backgrounds which have a higher nuclear charge because they have a shorter range. For example ^{14}N can be removed from the ^{14}C beam in this way, but not ^{26}Mg from ^{26}Al .

The normalisation of the counting rate either to a concentration or to an isotopic ratio with respect to the appropriate stable isotope can be performed by repeatedly interchanging the sample with a standard at the ion source, without changing any of the cyclotron and detection parameters. In conventional decay counting the counting rates are also referred to a standard.

The sample ionization for the cyclotron takes place in a Penning Ion Gauge (PIG) source which produces positive ions. The plasmas in these sources are hot enough (1kV) to dissociate all molecules; they supply large currents, and can be used with either solid or gaseous samples. Gaseous samples were used for ^{14}C dating in the form of CO_2 . For ^{10}Be detection the samples are solid pellets of compressed and sintered BeO (Raisbeck 1978). The pellets are placed in the plasma and sample atoms are sputtered by the plasma ions. Typically the cyclotron is tuned to accelerate the 2+ or 3+ charge states which are copiously produced by these sources.

The overall efficiency (atoms detected per sample atom consumed) of the cyclotron spectrometers has varied from 10^{-5} to 10^{-3} depending on the isotope and operating conditions. The ionization efficiency of the source depends strongly on the arc conditions and sample flow rate. The transmission through the long acceleration path depends on the quality of the tank vacuum since charge exchanges in the residual gas cause a loss of beam particles from resonance. Finally there is the efficiency of the absorber detector system. Improvements in the overall efficiency can be made with better detection geometry, improved vacuum, and optimized source conditions. These improvements are part of the continuing

development of the technique.

Spectrometers using tandem van de Graaff accelerators achieve a very powerful selection in the initial stage of ionization from their use of negative ion sources. If the radioisotope forms stable negative ions but one of the background ions does not, then this background is eliminated even before acceleration. (Some radioisotopes, including ^{39}Ar and ^{81}Kr , do not form negative ions and are difficult to measure with the tandem spectrometers.) The negative ions are produced in sputter sources (Middleton 1974, Brand 1977) in which a fine beam of Cs^+ ions is used to sputter negative ions from the sample. The sample is in solid form and a variety of sample preparations have been used. For ^{14}C dating pulverized charcoal is compressed with an equal volume of KBr. Samples using other matrix material and also pure graphite prepared by pyrolysing acetylene have been used. Ions for ^{36}Cl dating are sputtered from AgCl and NaCl, and compressed BeO is used for ^{10}Be measurements.

The cesium sputter sources provide large beam currents (20-30 microamps of $^{12}\text{C}^-$, 300 nanoamps of $^{9}\text{BeO}^-$) and they are highly efficient. Tests of the ion source output of carbon beams show that perhaps as much as 10% of the sample may be convertible to useful beam. Along with the radioisotope ion there are a number of other atomic and molecular ions produced by the sources and these are then separated in the acceleration and analysis stages. With carbon samples intense beams (3 nanoamps) of $^{12}\text{CH}_2^-$ and $^{13}\text{CH}^-$ are produced. In addition $^{14}\text{N}^-$ is produced, but since it is unstable it doesn't survive to the detectors. Production of $^{10}\text{BeO}^-$ is accompanied by a large current of $^9\text{BeO}^-$, and $^{36}\text{Cl}^-$ ions are produced amongst strong beams of $^{35}\text{Cl}^-$, $^{37}\text{Cl}^-$, and H^{35}Cl^- .

The tandem then serves as both an accelerator and a "molecular disintegrator." Molecular components in the beam are broken up upon passing through the stripper at the tandem terminal. Both thin carbon foils and gas strippers have been used. The terminal voltage is chosen to make the disintegration and also to optimize the output at the desired charge state.

The final stages of beam analysis vary considerably from group to group; undoubtedly because they have so far been based simply on the availability of existing equipment. In addition to a stage of high resolution magnetic analysis most include some additional dispersion. Three types of additional beam dispersion are possible: velocity ($E \times B$ filter), time of flight, and energy/charge (electrostatic filter). Each of the three have been used.

A number of different ions do reach the detectors due to scattering and charge exchange in the beam. The isotope of interest is finally separated and identified by the ion detector. The detector system consists of one or two transmission counters to measure the stopping power dE/dx and a final absorption counter to measure the residual energy. Since the beam is high energy the ions passing through the transmission counter are partially stripped of electrons and their effective nuclear charge allows their elemental identification. The radioisotope is cleanly distinguished from other ions.

The normalization of the radioisotope count rate to that for a stable isotope is achieved by comparing the count rate with that from samples of known concentration. The transmission of the system can be measured continuously by using a Faraday cup following the inflector (or

analyzing magnet) to detect a stable isotope. Both the Yale group and the Rochester collaboration have monitored the ion source and the accelerator transmission by periodically tuning the inflection and analysing magnets for a stable isotope.

The statistical error in a final age estimate is determined by the Poisson statistics on the number of atoms detected. The relationship between the magnitude of the fluctuations in the detected number of atoms and the resulting error in the estimated age is readily derived (Muller 1977). The conclusion is that a fractional error "f" in the measured rate (due to either systematic or statistical errors) will yield an error in the estimated age of $f \cdot T$, where T is the mean-life of the radioisotope. Thus a 10% error in the measurement of the ^{14}C concentration in a sample will produce an 800 yr error in the estimated age. Although such an error is intolerable for young samples, it may be acceptable for samples many mean-lives old (such as the 40,000 yr samples measured at Rochester). Even the measurement of only a single atom of the radioisotope gives an age determination statistically accurate to one mean-life.

In the accelerator measurements made so far, systematic errors have dominated. The sources of systematic errors include changes in the ion source efficiency and changes in the acceleration efficiency with time. The effect of these errors is that the accelerator method is at present not very precise, typically about 10-50%. There are also systematic effects arising from the contamination of samples and the existing spectrometers. This contamination limits the sensitivity of the method. It is expected that the construction of dedicated tandems and external ion

sources for cyclotrons will largely eliminate the spectrometer contamination. The small sample size requirement will allow investigators to isolate small chemical or mechanical fractions and obtain results for each of them. This may improve problems of sample contamination.

The accelerator method works best for the lighter elements. For $Z > 40$ the fractional difference in range becomes small and the unequivocal identification of different isobars more difficult. For the cyclotron the ranging out of the higher Z abundant stable isobars also presents more problems.

Costs do not appear to be a limiting factor. The typical operating cost of an accelerator is about 200 dollars/hour. In many cases the cost of sample collection and preparation far outweighs the cost of measurement.

3. FUTURE PROSPECTS

High energy spectrometry is still a very new technique and we have yet to learn the limitations and their relative importance. Likewise the long term successes and their real impact will not emerge for many years. The applications in the immediate future will undoubtedly be extensions and improvements of those discussed above. We can expect many more laboratories to develop spectrometers capable of rapidly making precise and accurate measurements.

We have seen in the last three years a trend in the development of radioisotope dating from the light isotopes to the heavier ones. For the lighter isotopes a lower energy is required to make use of the ioni-

zation loss and the difference in dE/dx for adjacent elements is greater. In addition most of the existing accelerators can accelerate the lighter isotopes. Higher energy machines capable of accelerating heavier isotopes have been built only recently and they are still few in number. techniques such as isotope enrichment.

Mass spectrometers of course have a very broad range of applicability and it is difficult to foresee all types of applications. The use of high energy spectrometry to search for new kinds of particles was the stimulus for the development of radioisotope dating in Berkeley and these searches have been continued at other laboratories (Middleton 1979, Boyd 1978). Another type of application, the measurement of low concentrations of nuclear reaction products in a target is being pursued by the group at Argonne National Laboratory (Kutschera 1979).

As more experience is gained with the present machines we can expect that high energy mass spectrometry will be combined with other analysis techniques. A program of isotope enrichment is being developed at Oxford in conjunction with the establishment of their high energy ^{14}C program (Hall 1978). At Rochester a successful test has been made of using an electromagnetic isotope separator to enrich a sample and simultaneously to implant the ^{14}C in a surface which is then sputtered with the tandem Cs sputter source (Currie, private communication). In another proposed hybrid application high energy spectrometry is combined with an area scanning Cs sputter source and used to analyse specific regions of a sample.

High energy mass spectrometry is now three years old. It was immediately recognized to have many exciting applications, and there was

an initial spurt of measurements proving the sensitivity of the accelerators. There has followed a period with perhaps fewer measurements than might have been expected from the initial optimism. During this period we have seen the hard and often slow work of trying to turn the technique into an analytical tool. This has included efforts to use smaller samples and to use them more efficiently, efforts to achieve precise, reproducible, and uncontaminated doubt that the optimism about the impact on many branches of science will prove well founded.

FIGURES

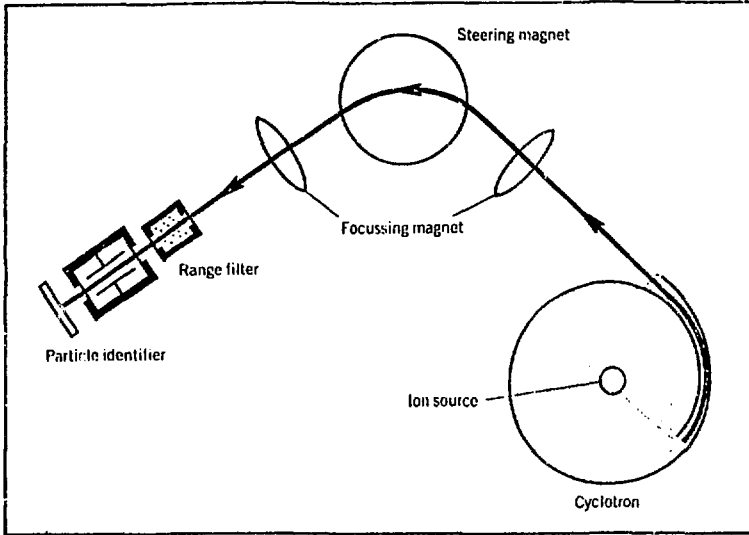


Figure 1

NBL 798-11179

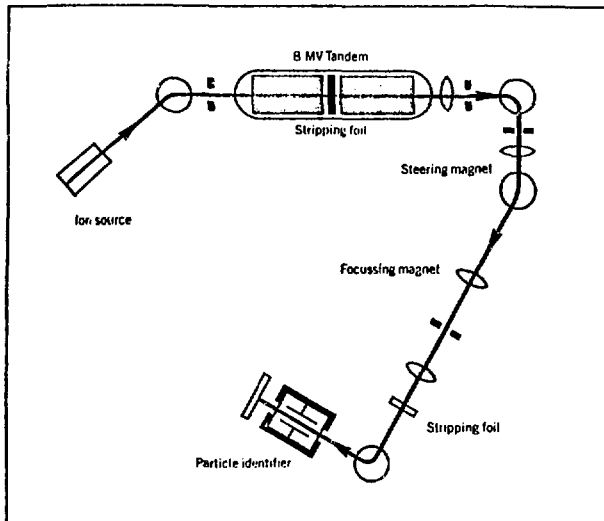


Figure 2

NBL 798-11178

FIGURE CAPTIONS

Figure 1. Schematic diagram of a high energy spectrometer at the Lawrence Berkeley Laboratory. Positive ions are accelerated by the cyclotron which also selects on charge to mass ratio. The range filter removes the background current of isobaric stable isotopes. The radioisotope atoms transmitted through the filter are then identified by their energy loss and total energy.

Figure 2. Schematic diagram of high energy spectrometer at the Rochester tandem Van de Graaff accelerator. Magnetic analysis is performed before and after acceleration. A final stage of stripping and magnetic analysis is used for further separation before the final identification of the radioisotope by energy loss and total energy.

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