

A STRATEGY FOR ADOPTION OF ACCELERATOR MASS SPECTROMETRY
BY THE EARTH SCIENCES

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Summary of Conclusions

1. Applications of accelerator mass spectrometry to investigations in the earth sciences are sufficiently advanced to:
 - a. continue technical development, and
 - b. warrant a transition in support from the nuclear physics community to the earth science community.
2. The most convenient accelerator for analyzing a wide variety of terrestrial cosmogenic radionuclides at a precision of about 5% is the tandem Van de Graaff accelerator at 6-10 MV in the first stage.
3. The minimum acceptable level of support needed is the current level, i.e., three facilities at 20% of full-time or \$200K per facility per year including salaries.
4. Three tandem Van de Graaff accelerators should be upgraded to measure isotopic ratios to a precision of 1%. This upgrading would cost about \$570K per facility including salaries for a total of \$1.7M. Ideally, funding would come in fiscal year 1985.
5. In fiscal year 1986, the earth science effort is expected to increase to half-time use of three tandem Van de Graaff accelerators at \$600K per year per facility. The other half-time use of these facilities might still be utilized in nuclear physics by experimentors for tuning up experiments preparatory to going on scarce higher energy machines.
6. The Cockroft-Walton principle used in the 2-2.5 MV Tandetron may be ideal for particle induced X-ray emission (PIXE) and perhaps radiocarbon dating. The energies available are not sufficient as yet for sensitive analysis of other terrestrial cosmogenic

radionuclides such as beryllium-10, chlorine-36, calcium-41, or iodine-129. The cost of a completed Tandetron is about \$2M, but the upgrading of existing tandem Van de Graaff accelerators may be more cost effective.

7. A sum of \$200K per year is needed to explore instruments alternative to existing machines. The two most promising possibilities are the cyclotrino and resonant ionization spectrometry using laser light.
8. A facility dedicated to accelerator mass spectrometry should be targeted for fiscal year 1989. The details of this facility will depend upon what is learned about accelerator mass spectrometric methods in the next few years.

I. INTRODUCTION

In mass spectrometry, four steps are involved in the analysis of the isotopes of an element: volatilization and ionization of the element, accelerating the ions through a potential field, separation of the isotopes usually by passing the ions through a magnetic field and measuring the intensity of the isotopes. In conventional mass spectrometry, the energies given ions are usually a few kilovolts (~10 kv). Accelerator mass spectrometers (AMS) produce ions with much higher energies in the megavolt range -- three to four orders of magnitude greater than in conventional mass spectrometry. Nuclear physicists have been examining applications of AMS to geology in collaboration with earth scientists, primarily over the last five years. The cyclotron was the first accelerator used for radioisotope dating (Muller 1976, 1977) but it was soon followed by measurements with tandem Van de Graaff accelerators (Nelson et al., 1977; Bennett et al., 1977). Although extensive use of the cyclotron was made by Raisbeck for earth sciences purposes (Raisbeck et al., 1978a,b, 1979, 1980, 1981a,b) access to these large machines proved too limited to be of general use. Decrease in the use of lower-energy tandem Van de Graaff accelerators for nuclear physics stimulated their use in investigations of applications to earth science. The tandem Van de Graaff actually has a number of advantages as a mass spectrometer, as is discussed in Section III. Funding for the modification and operation of these machines has come almost exclusively from the Nuclear Physics Division of the National Science Foundation. Progress in the application to the earth sciences (Section II) has been rapid, and a strategy for the transition of funding to the earth science community is needed (Section IV).

As the result of a recommendation from a subcommittee of the Geological Sciences Board of the National Academy of Sciences, an intersocietal committee on instrumentation in geochemistry and cosmochemistry was formed in 1983 with representatives from the Geochemical Society, the section on Volcanology, Geochemistry, and Petrology of the American Geophysical Union and the Mineralogical Society of America. In a preliminary analysis of frontier areas of isotopic geochemistry, accelerator mass spectrometry was seen to be in a particularly precarious state. Because of the great potential of the use of terrestrial cosmogenic radionuclides -- some of which can only be practically determined by AMS -- in dating old ground waters and determining ages and rates involved in ground instability, this study was funded with Bruce R. Doe as the Principal Investigator by the USGS program in Radioactive Waste Disposal that is much concerned with these topics. The authors of this report met on January 10 and 11, 1984, to develop a strategy for adoption of accelerator mass spectrometry by the earth sciences. This report is the summary of that meeting.

II. APPLICATIONS FOR EARTH AND PLANETARY SCIENCES

The cosmogenic nuclides are produced by the interaction of cosmic rays and their secondaries with terrestrial and planetary matter. Up until recently, the only cosmogenic radionuclide that was being extensively applied to terrestrial radiometric dating or natural tracer problems was radiocarbon. Measurements were made by the anticoincidence beta-counting technique that applies only to about the last 40,000 yrs B.P. (see Table 1 for half-lives), and several grams of carbon are usually required. Following the discovery of ^{10}Be as a naturally produced isotope in pelagic sediments by Arnold (1956), a small but significant effort on beryllium-10 in establishing

growth rates of manganese nodules and determining deposition rates of pelagic sediments was carried out in 1970's mainly by scientists of India and Japan. A problem in the utilization of beta-anticoincidence counting of such long-lived isotopes is the scarcity of decays. Until recently, however, little could be done with species having much longer half-lives, especially beryllium-10, aluminum-26, and chlorine-36, owing to the large amounts of sample material required to furnish an adequate number of atoms to count by observing their decay. The difficulty can be appreciated by noting that of 10^9 atoms of beryllium-10, the quantity present in a gram of typical terrestrial sediment, only one decays in a day.

The revival of accelerator mass spectrometry in 1977 altered the observational situation so radically that completely new fields of investigation have been added to the revitalized older ones (Lal & Peters, 1967; Arnold, 1956) that employ cosmogenic isotopes. Accelerator mass spectrometry overcomes this decay problem by directly counting numbers of atoms of the isotope that are many orders of magnitude greater than the number of decays over short-time intervals such as days or weeks. Thus cosmogenic radionuclides of longer half-lives may be precisely measured and smaller sample sizes are now amenable for study than was previously possible. Among the longer half-lives are beryllium-10 (Raisbeck et al., 1979) and chlorine-36 (Elmore et al., 1979). Milligram rather than gram-sized samples of carbon may now be dated (Bennet et al., 1978). Over a thousand samples have already been analyzed for beryllium-10 and nearly 1000 analyses exist for chlorine-36 in a broad variety of materials such as soils, pelagic sediments, lake sediments, ice, petroleum, volcanic rocks, and tektites. Lesser but still significant uses were found for cosmogenic isotopes of shorter half-lives than carbon-14, such as hydrogen-3 (tritium), and beryllium-7.

Considerable science has already been accomplished by the accelerator method with beryllium-10, carbon-14, aluminium-26, and chlorine-36. Indeed a recent review (Brown, 1984) cites 49 reports of original investigations using the technique, a large fraction of which was accomplished during the last year or two. The method's developers had carbon-14 primarily in mind and employed it in their initial experiments. Because of a data span of only three orders of magnitude in the isotope ratios and severe competition from the older analytical technique, the accelerator method required special development. On the other hand, an improvement of seven orders of magnitude in detection efficiency of beryllium-10 over beta-counting was obtained with minor modifications of existing accelerators and their peripheral equipment. These improvements coupled with the rare cosmic abundance of beryllium-9, the only stable isotope, completely altered the scientific use of beryllium-10. Similarly, chlorine-36 has found important application in studies of terrestrial water and ice, where it is not overwhelmed with stable chlorine. Even aluminum-26 -- an isotope measured in only a handful of samples prior to 1982 -- has now been observed in manganese nodules by using the accelerator method.

The advances that have accrued during the brief period of application are quite varied. The results have contributed to research on the atmosphere, the history of polar ice, manganese nodules, and pelagic sediments, the evolution of soil and its erosion, the formation of continental margins, the dynamics of aquifers, the origin of tektites, the nature of island-arc volcanism, the dating of earthquakes, and the irradiation or exposure history of lunar and meteoritic material. This work now reflects the efforts of those who were struggling with the experimental difficulties in 1977 and of many more who previously never dreamed that their research would involve a particle

accelerator. A complete summary of this effort is out of place here but a few examples can illustrate the method's strengths.

Ages measured by accelerator mass spectrometry are beginning to be reported for radiocarbon (Tucker et al., 1983). Measurements on carbon-14 at Arizona (Taylor et al., 1983) and at Chalk River (R.M. Brown et al., 1983) seem to have settled a long-standing dispute by setting an age of about 4000 yrs for American Indian skeletons for which another technique had given a much older age, one which would have had sensational consequences for archaeology had it been true. Measurements in polar ice (Raisbeck et al., 1981b; Beer et al., 1983) have disclosed a threefold increase in beryllium-10 concentration for ice precipitated during the most recent ice age, and Beer et al. (1983) have even observed variations correlated with the 11-year solar cycle. The slow accretion rate of manganese nodules (mm/Ma) seems to be well substantiated by beryllium-10 measurements (Turekian et al., 1979; Krishnaswami et al., 1982; Ku et al., 1982). Evidence that tektites have their origin in terrestrial sediment has come from the discovery of Pal et al. (1982) that they have beryllium-10 concentrations comparable to these of continental sediments. Later Raisbeck et al. (1983) showed the aluminum-26 concentration in tektites is incompatible with a lunar origin. Measurements of chlorine-36 by Nishiizumi et al. (1979, 1981, 1983) on Antarctic meteorites identified many stones with terrestrial ages much older than the maximum previously known. Numerical age differences for Antarctic ice samples have been established by their beryllium-10/chlorine-36 values. Chlorine-36 has been applied recently to the dating of saline deposits at Searles Lake, California (Phillips et al., 1983). Speculation about the mechanism by which

subducted material is incorporated into the magmas of island-arc volcanos must conform to new constraints imposed by the observation of beryllium-10 in the lavas from some magmatic arcs (Brown et al., 1982).

Some measurements show the possibilities of developing cosmogenic radionuclides into new fields of research. There are indications that beryllium-10 may be a particularly useful tracer in studying the recent history of soil development (Monaghan et al., 1983; Pavich et al., 1984) and of soil erosion (Klein et al., 1983) by comparing the amount of the isotope that is incident on the drainage basin with that which is borne to sea in the sediment load. Beryllium-10 has been observed in a variety of organic materials: the organic component of lake sediments (Lundberg et al., 1983), petroleum (Yiou et al., 1983), wood (Middleton, 1981), humus and lignite (Carnegie Institution of Washington, and University of Pennsylvania, unpublished data). One need not be considered an uncontrolled optimist to believe that these new fields of study will prove of great value.

The high sensitivity of accelerator mass spectrometry can be exploited to measure stable isotopes present in concentrations at or below the part-per-billion level (Muller, 1977; Sie, 1983). Such information may find use where elemental compositions carry special significance, for example as in the calculation of partition coefficients.

The analysis of platinum-group metals constitutes an area of active interest. Competing methods are subject to various interferences and hindrances and may require chemical separations. Rucklidge et al. (1982) have demonstrated the possibility of using accelerator mass spectrometry to measure platinum and iridium directly. Bulk samples of a few milligrams were analyzed in a matter of minutes. These results represent sensitivity gains of 10^4 and 10^5 for platinum and iridium, respectively, compared to alternative

techniques. The production of ions by sputtering, that constantly exposes fresh sample material, appears greatly to reduce problems of sample contamination. The work of Rucklidge and co-workers confirms the presence of large iridium anomalies in Cretaceous-Tertiary boundary deposits.

Recent studies of Luck and Allegre (1983) and Luck and Turekian (1983) using the ion-probe mass spectrometer have rekindled interest in radiometric dating with rhenium-187/osmium-187. For certain applications, the determination of rhenium-187/osmium-187 ratios with a precision of 1% may suffice (Luck and Turekian, 1983), a figure that lies within the reach of current methods of accelerator mass spectrometry.

Other possible applications not now under active development at a U.S. laboratory may include studies of lithium and boron isotopic abundances and of the minor isotopes of other elements.

Accelerator mass spectrometry has opened up a whole new vista of opportunities for solving aspects of societal concerns that were previously not approachable, as well as contributing much to basic earth science. Examples include dating of old ground waters -- a problem given the highest priority in geochemistry related to radioactive waste disposal; hydrologic tracers; ages and rates of sedimentation for solving times of faulting, buckling, and the like. Measurements of cosmogenic radionuclides in target rocks on the earth's surface may also lead to the possibility of determining tumbling ages of rocks and boulders at the Earth's surface, thus providing the opportunity for dating land slides, terrestrial flooding, vertical faulting, and other processes. Establishing the ages and rates of hydrologic flow are important to nuclear and toxic waste disposal because non-leaking aquifers with long, slow moving flow paths of 10's of thousands of years are desirable to inhibit return of any waste to the biosphere. Ages of faulting,

bulging, etc., are also important to waste disposal, not so much because waste will be lifted to the surface, but because waste may be introduced into a potable-water source or introduced into the biosphere by alteration of hydrologic flow paths. A unique opportunity is afforded us by the unfortunate anthropogenic contribution from the marine nuclear tests of the mid-1950's that put substantial chlorine-36 into the stratosphere. This chlorine-36 gradually has settled to the Earth and pretty well was cleaned from the stratosphere by about 1975 (Elmore et al., 1982). This pulse of chlorine-36 is entering the water table, providing a splendid opportunity to follow this pulse with time while determining common chlorine build-up, leaking aquifers, etc. Hopefully, this kind of opportunity will not arise again for humanity, but a very useful tracer has been provided, nonetheless. In addition, iodine-129 was added to the stratosphere by all atmospheric nuclear tests and is continuously added to the troposphere by nuclear power plants. Iodine-129 thus is continually entering the water table affording the possibility of distinguishing pre-nuclear device water from post-nuclear device water. Nuclear-waste disposal deals fairly comprehensively with natural-hazards concerns, and various aspects of that program apply to other programs as well, e.g., reactor hazards, earthquake hazards, volcano hazards, etc. Few frontier areas of earth science show such promise for applications of earth science as accelerator mass spectrometry and terrestrial cosmogenic radionuclides.

Following sections will expand on the capabilities of accelerator mass spectrometers and recommendations of the working group.

III. ACCELERATOR CAPABILITIES

A. Tandem Accelerator Mass Spectrometry

The tandem Van de Graaff accelerator is a constant voltage, electrostatic accelerator used widely for fundamental studies in nuclear physics. This accelerator was the first type used in 1977 (Bennett et al., 1977; Nelson et al., 1977) for the detection of carbon-14 at natural levels, and this measurement was accomplished without any modifications of the instrument. The tandem accelerator and its associated beam-transport system has many features that make it ideal for ultra-sensitive mass spectrometry. It is a direct current machine known for its energy stability and ease of changing its operating characteristics. Beam injection and extraction are at ground potential and use straightforward beam optics. Isotope ratios are obtained by the standard mass-spectrometry technique of magnetic-field tuning. The fact that negative ions are accelerated in the first stage leads to important advantages in isobar separation (isobars are isotopes of different elements having the same mass number). As negative ions are not formed from nitrogen, magnesium, or argon, interferences are minimized between nitrogen-14 and carbon-14, magnesium-26 and aluminum-26, and argon-36 and chlorine-36. Molecules are completely eliminated by choosing a charge state of 3 or greater from the terminal stripper because such highly charged molecules are highly unstable and would not survive the trip to the detector. Tandem mass spectrometers capable of 2-3 MV terminal voltages are necessary to strip efficiently to the 3^+ charge state.

The tandem-accelerator system can be considered a general-purpose mass spectrometer except for heavy noble gases (only helium forms negative ions). The radioisotopes beryllium-10, carbon-14, aluminum-26, chlorine-36, and iodine-129 have been detected at the University of Rochester, for example.

However, each isotope is a special case, requiring individual ion source and detection techniques. In particular, isobar separation is a non-trivial problem requiring different solutions in each case. Various techniques, to be described below, have been developed for separation of isobars at the 10^6 to 10^{10} level for isobars below mass 40. Separation of roughly equal intensities is possible at mass 100, but separation is not practical for the heaviest elements (except where stable isobars are noble gases, as with iodine-129 and xenon-129).

The system in use at Rochester (Fig. 1) is similar to that used at other laboratories and is a good example of the use of the principles of ultrasensitive mass spectrometry. Although the large electrostatic analyzer is the only major addition to the standard apparatus used in nuclear physics, many smaller additions and refinements are necessary for the most sensitive and precise isotope ratio measurements. The selection achieved by the various components is summarized briefly below.

Chemical separation is usually made prior to insertion of the sample into the ion source. This separation ensures the maximum current of the ions of interest and reduces the concentration of isobars, where required. A cesium-sputter ion source is used (Middleton, 1977). Basically this is a secondary-ion mass-spectrometry (SIMS) instrument that has been refined to produce high currents of negative ions. Generally, solid samples are analysed although it is possible to use gas if "memory" of previous samples can be reduced sufficiently. Typically, samples of 1-10 mg are used and beam currents of 1-20 μA are possible, depending on the element (see Table 1).

Prior to acceleration, the 90° inflection magnet selects the mass of interest with a resolution of about 200. Molecules and isobars that form negative ions are still present after this magnet. The ions are then

accelerated to the terminal which is held to a constant positive voltage in the range 5-12 MV by a generating volt-meter stabilization system. Here several electrons are removed in a gas stripper (or sometimes a thin carbon foil is used) and accelerated further to ground potential as positive ions. The name "tandem" is derived from this dual acceleration concept. The magnetic analyzer and magnetic switcher serve as a second mass spectrometer that removes molecular fragments. The electrostatic analyzer or a Wein filter (consisting of crossed electric and magnetic fields) is necessary to remove particles that have different mass but possess the correct momentum to pass the magnetic analyzers. These particles are differentially accelerated following multiple charge exchange in the accelerator and are effectively removed by combined electrostatic and magnetic analysis.

The heavy ion detection system (Fig. 2) provides extra valuable information and sometimes allows the conclusive identification of the isobar of interest. The system consists of several parts. The stable-isotope beam current that is used to optimize previous ion-optical elements passes through an aperture and is measured in a removable Faraday cup. A pair of time-of-flight detectors determines the beam velocity to a fraction of one percent. This determination is required for resolving heavy isotopes such as iodine-129 and stable isotopes of the platinum-group elements from their neighboring isotopes. A combined foil and gas-cell absorber following the time-of-flight detectors stops boron-10 but allows beryllium-10 to pass because beryllium-10 has a longer range. The final ionization detector measures the total energy and the rate of energy loss. The latter distinguishes the atomic number and is most useful for separation of chlorine-36 and sulfur-36.

Comments will now be made on the operating characteristics and capabilities of tandem-accelerator mass spectrometry for measurements that

have been well developed for several long-lived radioisotopes. Details for typical routine performance are presented in Table 1. Beryllium-10 is accelerated as BeO^- because the samples are difficult to reduce to metallic form and the BeO^- beam is much more intense from the sputter source than is Be^- . Because more than half of the energy at the terminal goes into the oxygen atom, a terminal voltage of at least 7 MV is needed to get optimum yield in the 3^+ charge state. However, boron-10 and beryllium-10 can be separated with terminal voltages as low as 2 MV with greatly reduced yield.

Radiocarbon is perhaps the simplest of the listed radioisotopes to detect and separate from interferences. A terminal voltage of 2 MV is adequate (Donahue et al., 1983) although several existing accelerators have used 8-10 MV. The difficulty with carbon-14 lies in the necessity of forming a graphite-like substance that produces the best negative-ion beams, and in achieving the precisions of 1% or better to which the radiocarbon community has grown accustomed to in dating larger samples by radioactivity counting. Several successful sample preparation techniques are discussed in the literature and new methods are being developed.

Aluminum-26 was first detected with tandem-accelerator mass spectrometers in 1978 (Kilius et al., 1980) but useful studies with natural samples waited for a new high-intensity ion source (Middleton 1977; Middleton 1983a) because it is particularly difficult to form negative ions of aluminum. Because the stable-isobar magnesium-26 does not form negative ions, the detection difficulty is the separation from aluminum-27. This separation can be done even at low energies with the 3^+ charge state if sufficient magnetic and electrostatic analysis is used.

Chlorine-36 detection (Elmore et al., 1979) requires the difficult separation from the isobar sulfur-36. Even though the sulfur-36 abundance is

TABLE 1: TYPICAL PERFORMANCE FOR TANDEM ACCELERATOR MASS SPECTROMETRY
(MINIMUM SAMPLE FOR 10% PRECISION)

radio-isotope	half-life (10 ⁶ yr)	chemical form of target ^{a)}	terminal voltage (MV)	sample size (mg) ^{b)}	lab ^{c)}	reference
⁰ Be	1.6	BeO	7	1	Pennsylvania	Klein et al., 1982
⁴ C	0.0057	C	6	1	Rochester	Bennett et al., 1978
⁶ Al	0.72	Al ₂ O ₃	7	2	Pennsylvania	Middleton et al.?
⁶ Cl	0.30	AgCl	10	4	Rochester	Elmore et al., 1979
²⁹ I	15.9	AgI	5	2	Rochester	Elmore et al., 1980

radio-isotope	ion source current (μA)	accelerator ^{d)} efficiency (%)	isotope ratio (10 ⁻¹⁵) ^{e)}	run time (hr) ^{f)}	atoms per sample
⁰ Be	4	14	10	1.6	6x10 ⁵
⁴ C	2	40	50 ^{g)}	0.3	3x10 ⁶
⁶ Al	1	13	100	0.7	4x10 ⁶
⁶ Cl	15	10	10	0.6	6x10 ⁵
²⁹ I	6	4	1000	0.1	1x10 ⁷

Numbers are not necessarily the best obtained independently for each parameters but are, wherever possible, a consistent set derived from actual measurements of natural samples.

a) Atomic ion is accelerated except for BeO⁻.

b) Elemental weight includes carrier added, if any. (Carrier is material of same element with negligible radioisotope content used to dilute the sample.) Samples larger than that shown are easier to handle but do not necessarily produce better results. In some cases a binder such as Ag powder is mixed with the sample (this is not included in the weight given).

c) These labs were chosen because results were available to the authors; others may perform as well or better.

d) Efficiency includes all beam losses between ion source and detector.

e) The isotope ratio used for the final two columns is taken as ten times the value for a blank. Blank here means a sample with negligible radioisotope that is prepared identically to the unknown. Since backgrounds are often uncertain by a factor of two, ten times consistently gives 10% precision. For samples having a higher isotope ratio: more carrier can be used, the time can be reduced, and/or better precision can be obtained.

f) The run time (time to collect 100 counts at given isotope ratio) has been multiplied by a factor of two to allow for measuring the stable isotopes. Multiply by 10 for 3% statistics (more sample may be required).

g) This ¹⁴C blank times ten corresponds to a 25,000 year age. The blank results from ¹⁴C contamination in the ion source and sample, not from interfering beams. Cleaner sources can solve this problem.

1.4×10^{-4} of the sulfur and chemistry can reduce the sulfur concentration to 1 ppm, the interference rate is 100-5000 counts-per-second. Several energy loss measurements in the gas counter are used to separate this isobar sufficiently to "date" several hundred-thousand-year-old ground water (chlorine-36/chlorine = $5-10 \times 10^{-5}$). Up to now, published results at this level have been made only at 10 MV using the Rochester accelerator.

Iodine-129, both the heaviest and longest lived isotope detected so far by tandem-accelerator mass spectrometers, requires a different detection technique. Other typical beam transport magnets following the accelerator pass iodine-127 tails at the iodine-129/iodine-127 = 10^{-10} level. Time-of-flight detectors have been used successfully to separate these isotopes (Elmore et al., 1980).

B. Cyclotron and other methods

One feature of the tandem Van de Graaff that makes accelerator mass spectrometry successful is the high energy of the emerging beam. This high energy allows techniques developed in nuclear physics for "particle identification" to be used to separate and identify background ions that accompany the ions of interest. Although tandem Van de Graaff accelerators have figured prominently in high-energy mass spectrometry, other devices have been proposed and used. The Berkeley 88-inch cyclotron was used for early measurements of hydrogen-3 by Muller (1977) who also discussed how beryllium-10 and carbon-14 might be analyzed. Measurements of carbon-14 by Muller (1978) were reported the following year. Extensive use of a cyclotron at Grenoble was made by Raisbeck and Yiou (Raisbeck et al., 1978a,b) in their pioneering measurements on a cyclotron at Grenoble of over 100 samples containing Be-10. However, neither the Berkeley nor the Grenoble cyclotron is currently employed for radioisotope measurements. In the case of Berkeley,

the machine proved cumbersome to modify to accelerator mass spectrometry, and, in the case of Grenoble, although the machine did not require extensive modification (it already had an external ion source) it was simply not made available for further measurements. Raisbeck had proposed a combination of cyclotron and linear accelerator for measurements of aluminum-26, but was never given the opportunity to test his method experimentally.

The current extensive use of tandem Van de Graaff accelerators was facilitated by several factors, including the ready availability of existing machines (there are 31 tandem Van de Graaff accelerators currently installed in the United States alone), the accessibility of the ion source (which is external, and at ground potential), and its use of negative ions (which suppresses certain backgrounds, such as nitrogen-14 and magnesium-26). Given the present operating capabilities, it makes most sense to give the primary support to tandem-accelerator programs.

Despite the present dominance of the field by the tandem Van de Graaff mass spectrometer, it is worth continuing the consideration of other devices, and basic research and development in such systems should be supported. For many ions, (such as beryllium-10) the tandem offers no intrinsic advantage, and for some which form no negative ions (such as argon-39 and krypton-81) it simply cannot be used.

We know of two alternative systems presently being developed, the small cyclotron ("cyclotrino") and a laser resonant-ionization technique. If either technique proves feasible, even for a limited range of radioisotopes, it may offer advantages in cost and accessibility over the tandem Van de Graaff mass spectrometer. They are also the only techniques at present that offer hope of measuring the noble gases (krypton-81).

Cyclotron

The cyclotron is a small (10-inch diameter) low-energy (40 keV) cyclotron being developed at Berkeley (Muller et al., 1981) using either negative or positive ions. The goal is to take full advantage of the mass resolution obtained in a cyclotron (Muller, 1977) made possible by 100-200 turns during acceleration at relatively high beam current (one microampere). It uses several innovative features, including high harmonic operation (31st) needed to obtain high resolution with 100-200 turns, and electrostatic focusing. It is currently undergoing testing, but has not yet been used for radioisotope detection. If it works, it may bring the cost of owning a machine down below \$100K, but because of limited current, it may not reduce the cost per sample below that obtainable by a large dedicated machine.

Laser Based Methods

Resonant-ionization spectroscopy (RIS) using tunable laser light affords the possibility of elemental and isotopic identification, obviating the needs for the high energy - particle telescope arrangement (Hurst, 1979). The combination of this selectivity with conventional mass analysis offers high abundance sensitivities. Development of these schemes is being vigorously pursued in several laboratories.

Versions of this scheme are being developed at the National Bureau of Standards (RIMS-Resonant Ion Mass Spectrometry; Fassett et al., 1983) and at Atom Sciences Incorporated (SIRIS-Sputter Initiated Resonant Ion Spectrometry). Atoms from a thermal or sputter ion-source are first resonantly excited by a laser (single or multiple photon). These atoms are then laser ionized, mass separated, and detected. Pulsed-laser operation provides the high power needed for ionization or two-photon resonant

excitation. This procedure affords time-of-flight rejection of background, but may involve line-broadening difficulties in distinguishing isotopes. Several improvements and variations on this basic scheme have been demonstrated or are under consideration. A thirty-fold improvement in sample utilization has been obtained at low duty-cycle with a pulsed thermal-atom source (Fassett et al., 1984). Further selectivity (with some loss of sensitivity) could be obtained after mass analysis by observation of laser-induced resonance fluorescence of the ions or of neutralized atoms, detecting photon bursts from multiple excitation under conditions of small Doppler spread (Lewis et al., 1979).

Potential atom-abundance sensitivities of 10^{13} (strontium-90) to 10^{14} (carbon-14) have been projected (Lucatoro, 1984), but present achievements fall short by several orders of magnitude. Limitations include low duty-cycle (although pulse rates of several thousand Hertz are foreseen), losses from resonant charge and excitation exchange (source vapor density), and two-photon produced background of atomic and molecular ions. The ideal of an efficient direct-current system conflicts with high laser power (pulsed operation). For very high sensitivities, the laser scheme does not presently appear generally competitive with accelerator-based mass spectrometry, but at intermediate sensitivities (ppb), it may be of considerable value, especially in elemental analyses.

IV. RECOMMENDATIONS

Accelerator mass spectrometry (AMS) is now in a critical transitional stage. Developed by physicists who were mindful of its possibilities and intrigued by the technical challenges it presented, its support has come almost entirely from the funding for the nuclear-physics laboratories where it

was nurtured. This tentative and sometimes informal basis for its funding will not continue much longer now that the techniques for measuring beryllium-10, carbon-14, and chlorine-36 have become sufficiently routine so that much of the work is no longer developmental. Thus, just when the method is beginning to be of real importance as a tool in earth science, the grounds for support from funds for nuclear science are waning. Furthermore, there are trends in the support of nuclear science which can altogether cut off the possibility of continued accelerator mass spectroscopy at laboratories where it has thrived--i.e., some of these laboratories are losing their nuclear funding; in others, the accelerators are being upgraded to energies and configurations suitable only for the nuclear research. Accordingly, it is important that the support of existing AMS facilities be derived from the user community. In principle it would be possible to support the isotope-geology applications from a multiplicity of sources, possibly by payment for use. This mode of funding is inefficient, however, and not conducive to the systematic operation of an expensive facility. A national strategy is needed for the operation of AMS facilities in order to match projected demand for measurements with a reasonable level of funding. In addition, because the operation of an accelerator is expensive, it is necessary to inject the proposal for funding into the budget of the prospective funding agency with sufficient lead time to be included in the agency's funding cycle. Prompt action now could introduce the request for funding into the FY 86 budget.

Existing Facilities

In order to foster further development and application of accelerator mass spectroscopy in the United States as a research tool, we recommend support by agencies concerned with earth science at a level somewhere in the following range:

Lowest - Continuance at the level of activity previously maintained by sources of funding for nuclear physics. This level is equivalent to 20 percent of the machine time at three university laboratories equipped with tandem Van de Graaff accelerators in the 6 to 10 MV range. Since the operating budget of a typical tandem Van de Graaff accelerator is \$1M, just to maintain the current level of activity -- without major instrument upgrades -- requires at least \$600K in current dollars. This amount, it must be stressed, provides no more than what is currently being done, and, as we have pointed out above, the present activity in AMS will diminish unless new funds are obtained to replace decreases in nuclear physics support.

Highest - For the fiscal year starting in October 1985, support should be provided for the expansion and upgrading of the accelerator mass spectroscopy program in three university laboratories equipped with Van de Graaff accelerators in the voltage range 6 to 10 MV. Funding for this year would depend upon the specific needs of the individual laboratories but should include approximately \$70K at each institution for postdoctoral salaries and like amounts, on the average, for equipment to accomplish the upgrading. It is probably not realistic to suppose that the scientific requirement for AMS will be static; especially because the technique is at the point of rapid expansion of

applications. We estimate that demand may soon be the equivalent of 3 facilities working at about 50% capacity.

A possible alternative to participation of three laboratories in the 20 to 50 percent range would be a lesser number of laboratories involved at a higher level of activity. There is a tradeoff involved between efficiency (for the high level of activity) and a healthy diversity of approaches and competition (for the more numerous laboratories). There are advantages where a machine is shared with nuclear physicists--their continued association with an accelerator is a highly valuable resource for the work in AMS.

Therefore, for the fiscal year starting in October 1986, and in following years, support should be provided for the equivalent of a 50 percent program at each of three laboratories at an annual level of \$600K each. The laboratories so supported would be recognized national centers for accelerator mass spectroscopy and would be broadly accessible to U.S. investigators without cost. Users should usually be required to do their own chemistry, however, and to assist in the mass spectrometry runs. Samples for this research program, together with the chemical processing required to prepare them for analysis, will continue to be part of programmatic research at user institutions.

An advisory committee is needed to oversee the procedures whereby users apply for machine time, although every effort should be made to apply their controlling function so as to least impede the informal atmosphere in which such collaborations usually thrive.

If this new branch of science is to advance, more is needed than provision of data. Tandem accelerators in use in AMS were originally designed for nuclear physics and such characteristics as high and stable transmission was not an important consideration. The first step toward improving the

quality of the isotopic data is to upgrade selected existing instruments to optimize performance within the restraint of reasonable cost. We estimate that the cost per machine would be on the order of \$500. This amount need not necessarily be provided in one payment, however.

The AMS field is presently dominated by tandem Van de Graaff accelerators. There may be other ways to make AMS measurements, however, and it is important for rational funding of the field that a reasonable fraction of the total funding be devoted to instrument development; perhaps 10% of the total budget for AMS would be appropriate, or \$200K. This fraction would be used to support two areas of research. First is the investigation of optimum conditions for the measurement of new isotopes. Those conditions are important for the design of new smaller machines optimized to AMS operation that might ultimately be introduced. Second is support of research on highly innovative instruments that may have a lower probability of success, but which offer a large benefit if brought to fruition. Of special interest are techniques which could reduce the cost and complexity that presently characterize accelerator mass spectrometry and limit its applicability in geological research.

New facilities

By 1988, accelerator mass spectrometry should have evolved to the point where planning for a new facility will be required. Simple and relatively inexpensive techniques may emerge for handling routine measurements in one or more of the basic nuclides -- beryllium-10, carbon-14, and chlorine-36 -- and application of these techniques may come into more general use at programmatic laboratories. It seems likely, nevertheless, that the present trends in mass spectrometric techniques applicable to measuring heavy nuclides will persist

in the direction of their becoming more complex, more diverse, and continuing to require accelerator ion beams of 6 to 8 MV or more. We recommend, therefore, that planning for a new national user's facility be undertaken and that a preliminary target date for this new facility be 1990. There are currently perhaps 10 or 15 under-utilized FN and EN class tandem accelerators in the U.S., and until the introduction of a new generation of machine, it may be most cost-effective to rebuild one of these as a machine dedicated to AMS. The modification would involve improved transmission, lower operating voltages and fully automated operation. At an institution where an accelerator hall exists, the cost of refurbishment would be equivalent to a new accelerator of the Tandetron type, i.e., about \$1.5M. In order to justify such a large expense, very significant improvement in quality of measurement (i.e., sensitivity, precision) and substantial cost benefit in terms of lower operating expenses would need to be demonstrated.

New low-energy machines of the Tandetron type are applicable to carbon-14 and perhaps beryllium-10 or determination of trace element concentrations by PIXE. The installation of such a machine may become justified by a large regional demand in an area not served by other AMS facilities; or by a populous agency that requires its own in-house capability for vast quantities of data on a limited range of isotopes. It would be expected, however, that such installations would be funded by the interested parties because these would not represent national facilities.

Possible Funding Sources

In order to achieve a dependable level of support, we reiterate that the funding for regular operation must be provided from a single non-volatile source: the NSF is clearly the most appropriate agency for this funding.

Because of the budget cycle, such a large amount probably would not be available until FY 1986. For specific applications and one shot support for re-equipment, other sources of money may be:

- USGS - basic and applied geological science
- DOE - basic and applied energy sciences, energy resources, reactor siting, nuclear waste disposal
- NSF - basic and applied earth and marine science, anthropology, archeology
- NBS - standards for ultra-pure materials
- NASA - planetary science and applications

- Smithsonian - basic and applied geological and planetary science; archaeology.

- National Endowment for
the Humanities - archaeology

We recommend that this document be circulated to these agencies and that the possibility of both diverse and joint funding be pursued.

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ROCHESTER TAMS APPARATUS

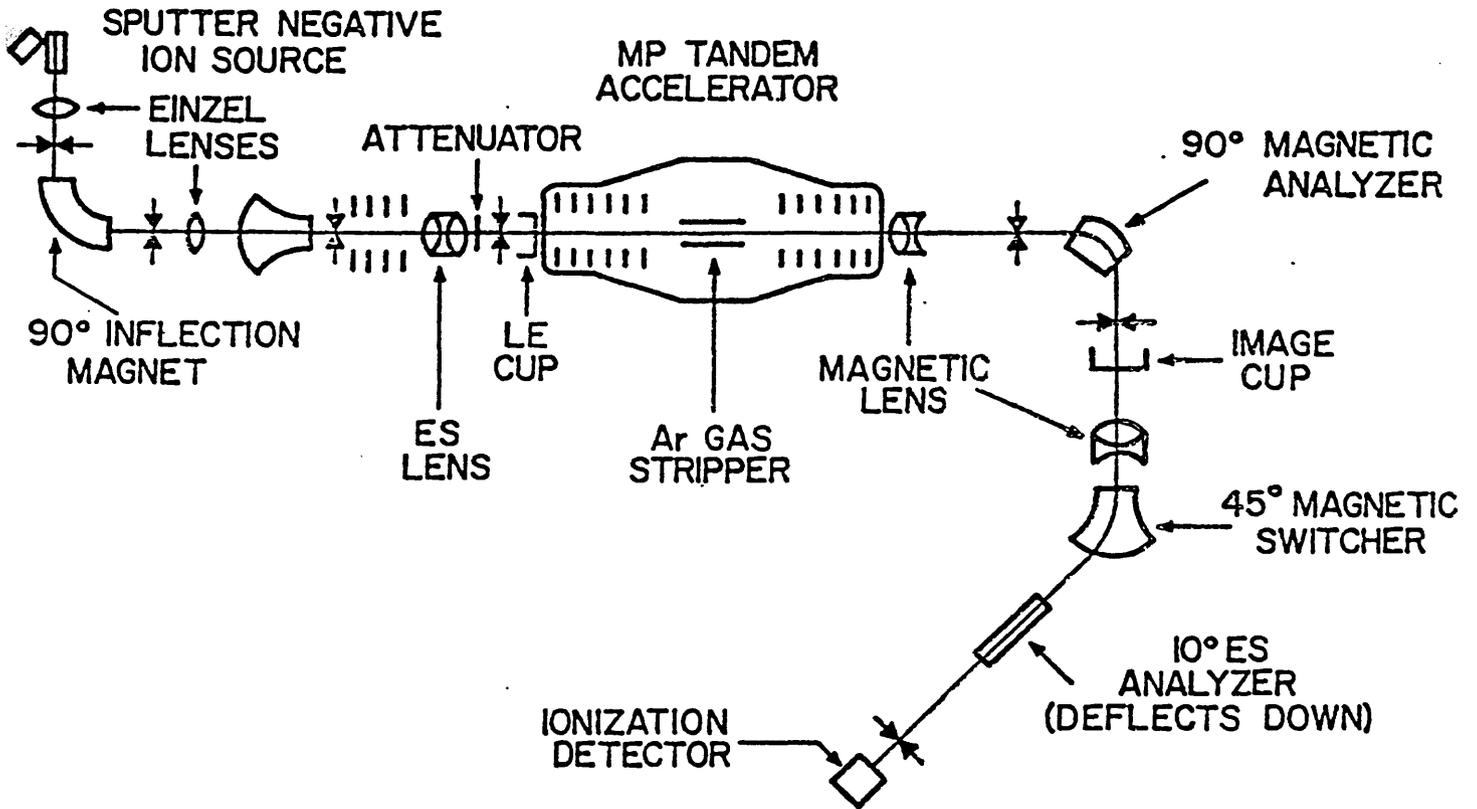


Figure 1

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TAMS DETECTOR SYSTEM

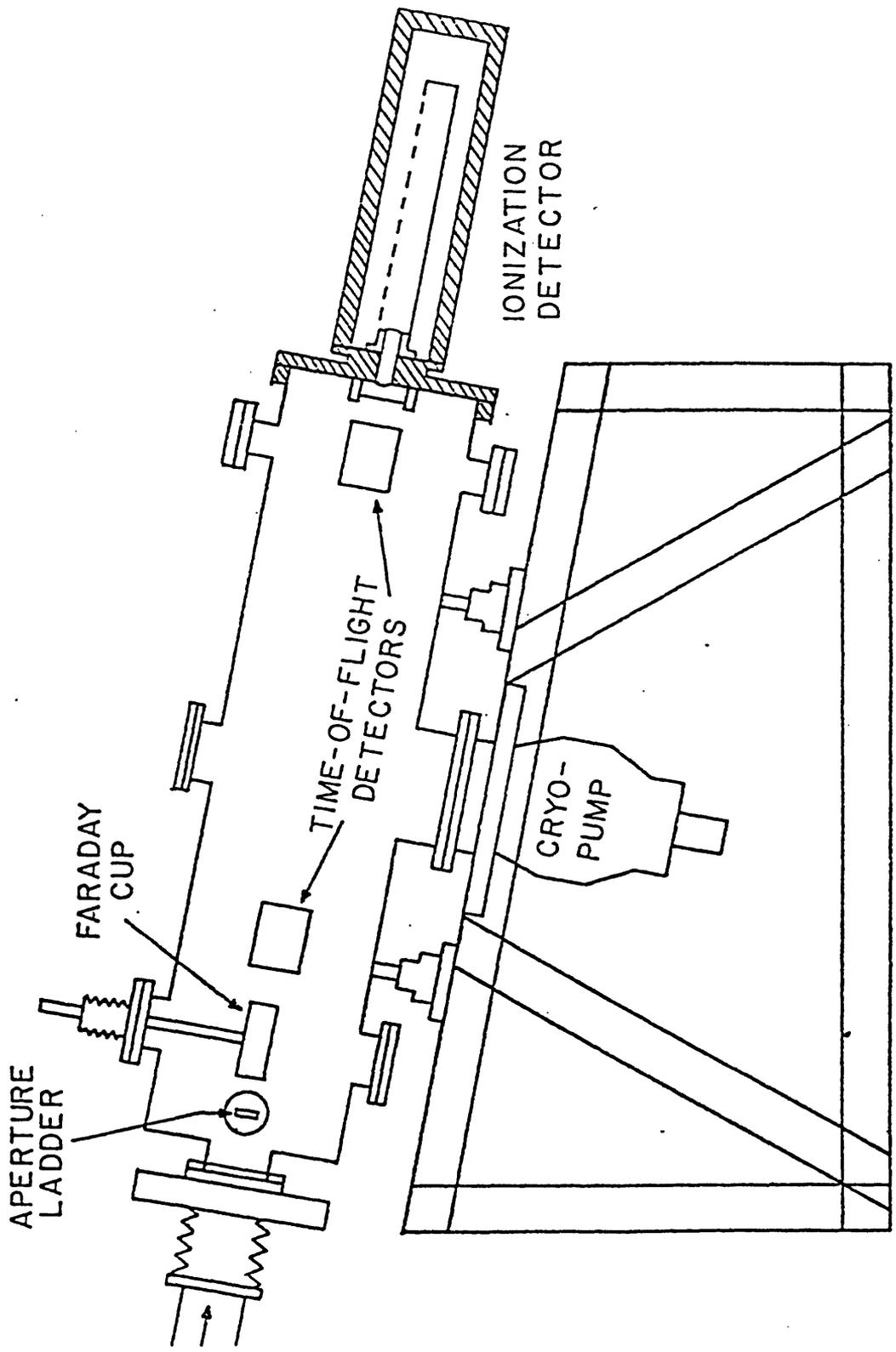


Figure 2