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APPLICATIONS OF ACCELERATOR MASS SPECTROMETRY IN ANALYSIS OF TRACE ISOTOPES AND ELEMENTS

(Technical Report)

Prepared for publication by

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Applications of accelerator mass spectrometry in analysis of trace isotopes and elements (Technical Report)

Abstract: Accelerator mass spectrometry (AMS) is mainly employed to measure very low concentrations of a number of long-lived radioisotopes and stable isotopes with very high sensitivities. The predominant application of AMS is in the field of geosciences and is based on chronological evaluations and dating principles. This paper deals with AMS applications other than dating problems, aiming at the analytical aspects of the trace isotopes and trace elements. It addresses non-dating applications in the fields of nuclear physics, geology, hydrology, environmental sciences, material sciences and, in recent years, biomedical sciences which have received increasing attention. Comparison of AMS with other analytical techniques with respect to detection limits and other functions is also discussed.

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I. GENERAL OVERVIEW

Accelerator mass spectrometry (AMS) is a ultra-sensitive technique, using a tandem Van de Graaff or other accelerators, that makes possible the measurement of several long-lived radioisotopes and certain stable isotopes at very low abundances (10^{-12} - 10^{-15}) with very high sensitivities (down to 10^3 - 10^5 atoms/sample).

Although the potential of the accelerator as a sensitive mass spectrometer was recognized early in 1939[1,2], this method was forgotten for almost 40 years. In 1977 it was simultaneously revived by a few laboratories[3-8] using a cyclotron or tandem accelerator. After more than 10 years of continuous development, AMS has become one of the most important tools for detection of a number of cosmogenic long-lived radionuclides, such as ^3H , ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{41}Ca , ^{53}Mn and ^{129}I . At present, more than 30 AMS facilities throughout the world are in operation or being built[9]. The current world-wide capacity of AMS measurements is probably close to about 15,000 samples per year.

Most of the applications of AMS so far have been in the earth sciences[10-12], including geochronology, sedimentology, petrogenesis, hydrology, volcanology, glaciology, oceanography and cosmology. In these fields, AMS has been applied predominantly on the basis of chronological evaluations and dating principles. By the same principles, AMS has also been successfully applied in

archaeology[13] and anthropology[14,15]. The non-dating applications of AMS have also played a contributive role in many fields, such as nuclear physics, geology and hydrology. Some new non-dating applications in environmental, material and biomedical sciences, have got rapid developments in recent years.

Symposiums on AMS have been organized every three years since 1978. Proceedings of these meetings provide the references and up-to-date information of the recent efforts and developments in this field[16-20].

A tandem based AMS system is basically composed of the following components[10]:

(i). A caesium sputter ion-source for producing negatively charged atomic or molecular ions. The typical beam currents are 1-20 μA , depending on the source type, specific element to be measured and the composition of the target.

(ii). An ion-injector consisting of a magnetic analyzer and sometimes a pre-acceleration system for negative ion beam.

(iii). A tandem accelerator including one section for negative ion acceleration and another section for positive ion acceleration, and an electron stripper in between. A total acceleration of heavy ions to a few tens of MeV in energy is achieved.

(iv). A positive ion analysis system for removing scattered particles, molecular fragments and unwanted charge states from the selected ion beam. A magnetic analyzer alone is not sufficient for that purpose and electrostatic analyzer or velocity selector is additionally used.

(v). An ionization detector for measuring the amounts of individual ions of different atomic number and mass. Sometimes a time-of-flight detector is also used.

The AMS facility at Peking University is schematically demonstrated in Fig. 1.

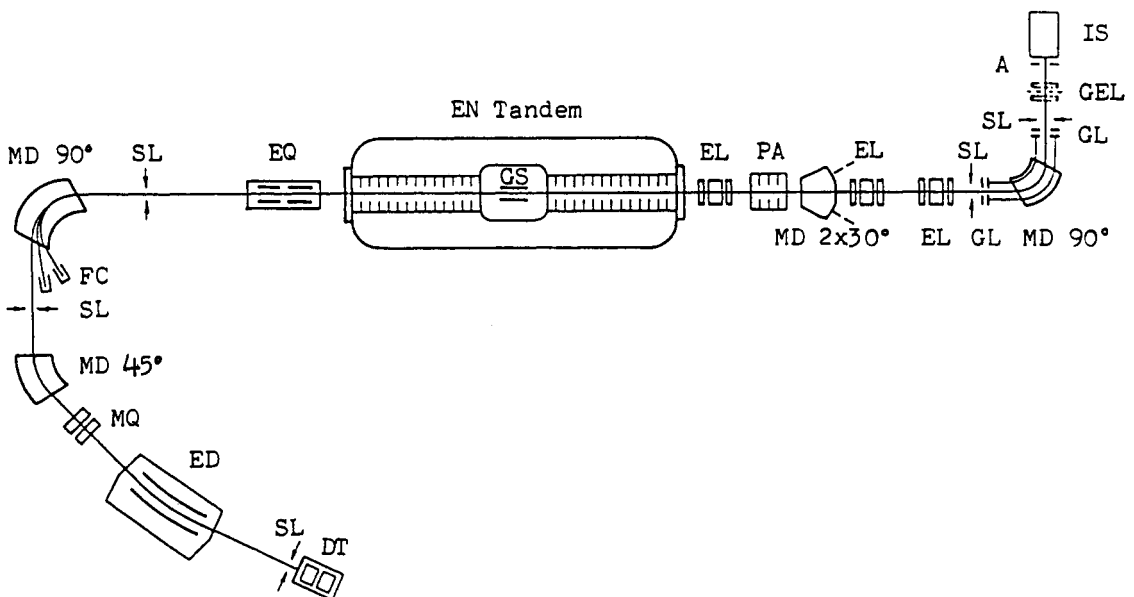


Fig. 1. Schematic layout of the AMS facility at Peking University. A aperture; DT ΔE -E detector; ED electrostatic deflector; EL einzel lens; EQ electrostatic quadrupoles; FC Faraday cup; GEL grid einzel lens; GL gap lens; GS Gas stripper; IS dedicated sputtering ion source for AMS; MD dipole magnet; MQ magnetic quadrupoles; PA preacceleration section; SL slits.

AMS has distinct advantages in comparison with conventional mass spectrometry and low level beta-counting methods[10,21]:

(i). Ultra-sensitivity

The AMS measurement is principally based on single atom counting, allowing detection of a very small isotope ratio of a long-lived nuclide or a very low concentration of a stable isotope. The very high sensitivity results from the high discriminatory power of the facility against the interferences of molecular species, scattering particles, isotopes and isobars, which makes the sensitivity of AMS several orders of magnitude higher than the conventional mass spectrometry. For most of cosmogenic radionuclides, especially when their half-life is greater than 10^5 years, the sensitivity of AMS measurements is much higher than that of β -counting method. For example, the lowest ^{10}Be concentration in a sample measured by β -counting is above 10^9 atoms/g, but AMS improves the detection sensitivity to 10^3 atoms/g or even less. The sensitivity limit, for example, for detecting $^{14}\text{C}/^{12}\text{C}$ ratio is down to 1.7×10^{-15} , and of the order of 10^{-15} for $^{10}\text{Be}/^9\text{Be}$, $^{26}\text{Al}/^{27}\text{Al}$ and $^{36}\text{Cl}/\text{Cl}_{\text{tot}}$. This rapidly growing technique increases the sensitivity of mass spectrometry by at least a factor of a thousand, and thus is often termed "ultrasensitive mass spectrometry" (see section III.A.).

(ii). Reduction of sample size and counting time

In conventional low level beta-counting, a large sample is required. For instance, for ^{14}C -counting in dating or tracer studies, several grams of a carbon sample are usually taken. For the longer-lived isotopes, even kilograms of samples are needed. AMS can often reduce the size of samples to a few milligrams and even to as little as 10 micrograms.

Also, AMS shortens the measurement time considerably. For example, measurement of 10 ppt (parts-per-trillion) content of Ir in an untreated mineral sample by AMS only took 10 minutes, whereas the time spent to gather such data for Ir by neutron activation analysis is in the order of a couple of days. In particular, when a series of tests are made, AMS gives shorter turnaround time in measurements.

On the other hand, there are some limitations in AMS measurements and applications[10,21].

(i). The obvious difficulty is the small choice of the appropriate elements to be studied, since only ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl and ^{129}I can be routinely measured, and the other radioisotopes including ^{32}Si , ^{41}Ca , ^{44}Ti , ^{53}Mn , ^{59}Ni , ^{60}Fe as well as some stable isotopes of Os, Pt, Ir, B, P, Sb, etc. are seldom used.

(ii). The overall efficiency of AMS is the ratio of the number of atoms detected to the number of the same kind of atoms in the sample. It depends on these parameters: the fraction of sample used, the efficiency of producing negative ions, the stripping yield in the particular charge selected and the transmission efficiency through the accelerator. The overall efficiency of AMS is lower than that of low-energy MS. The maximum efficiency among the accelerators ranges from 1.5×10^{-2} for ^{14}C to 10^{-6} - 10^{-5} for ^{53}Mn [17]. This variation reflects the different efficiency for forming negative ions in the caesium sputter ion source. The efficiency for forming a negative ion depends not only on the element, but also on the sample matrix.

(iii). Background in AMS may arise from contamination and interference species. The contamination can be introduced into a sample from either the environment or other residual samples in the ion source, and the interferences are unresolved molecular fragments, isotopes and isobars. Cross-contamination of different targets varies between 10^{-5} and 10^{-3} , depending on the geometry of the ion source. Special attention should be paid to the contributions by isobaric interferences from ^{10}B to the background of ^{10}Be , from ^{36}S to ^{36}Cl , and ^{32}S to ^{32}Si .

(iv). Factors that affect precision are counting statistics, isotope fractionation effects, frequency of cycling, and the effectiveness of blank and standard correction. Generally, for the long-lived radionuclides the precision attained in AMS is comparable to or even higher than that of radioactive counting of natural samples when the counting time available is not long enough, but lower than that of conventional MS.

Table I. Information about radioisotopes measured routinely with AMS

| | ¹⁰Be | ¹⁴C | ²⁶Al | ³⁶Cl | ¹²⁹I |
|---|---|---|---|---|--|
| Half-life(yr) | 1.6x10 ⁶ | 5730 | 7.05x10 ⁵ | 3.0x10 ⁵ | 1.57x10 ⁷ |
| Cosmogenic origin | N, O spallation | ¹⁴ N(n,p) ¹⁶ O(p,3p) | ⁴⁰ Ar spallation | ⁴⁰ Ar spallation ³⁵ Cl(n, γ) ³⁶ Ar(n, p) | Xe spallation U fission |
| Anthropogenic origin | - | ¹⁴ N(n,p) nuclear weapons tests nuclear reactors | - | ³⁵ Cl(n, γ) nuclear weapons tests | U, Pu fission nuclear weapons tests nuclear reactors |
| Stable isotope | ⁹ Be | ¹² C, ¹³ C | ²⁷ Al | ³⁵ Cl, ³⁷ Cl | ¹²⁷ I |
| Stable isobar | ¹⁰ B | ¹⁴ N | ²⁶ Mg | ³⁶ Ar, ³⁶ S | ¹²⁹ Xe |
| Appropriate chemical form of sample | BeO | C graphite | Al ₂ O ₃ | AgCl | AgI |
| Sample size, mg (elemental, needed for < 10% precision) | 0.2 | 0.25 | 3 | 2 | 2 |
| Background level | ¹⁰ Be/ ⁹ Be 10 ⁻¹⁵ | ¹⁴ C/ ¹² C 10 ⁻¹⁵ | ²⁶ Al/ ²⁷ Al 10 ⁻¹⁵ | ³⁶ Cl/Cl _{tot} 10 ⁻¹⁵ | ¹²⁹ I/I _{tot} 10 ⁻¹⁴ |
| Overall efficiency | 10 ⁻⁵ - 10 ⁻² | 0.5%-5% | 10 ⁻⁴ -10 ⁻¹ | 10 ⁻³ -2% | 10 ⁻³ -10 ⁻² |

Some fundamental information and data in AMS measurements of ¹⁰Be, ¹⁴C, ²⁶Al, ³⁶Cl, and ¹²⁹I are listed in Table I[10,22].

II. APPLICATIONS

This paper deals with the applications of AMS in analysis of trace isotopes and trace elements. In other words, we only aim at the analytical problems in various scientific fields, not at the dating problems which have predominated most studies of the routine applications of AMS. The following discussion will focus attention on the problem "What can we do with AMS?" and will not go far with the data interpretation, which usually needs to combine the AMS results with too much expertise descriptions.

A. NUCLEAR PHYSICS

AMS has the potential to be used for a number of fundamental nuclear physics applications[23,24] including nuclear reaction cross section and half-life measurements, and searches for free quarks, exotic particles, super-heavy elements and solar neutrinos.

1. Long half-lives of radionuclides

In the past, long half-lives of isotopes have been measured by Low Energy (keV) Mass Spectrometry (LEMS) via the well-known relation

$$\frac{dN}{dt} = -\lambda N = -N \frac{\ln 2}{t_{1/2}}$$

where N is the absolute number of radioactive atoms and dN/dt is the decay rate. This has been carried out only when a sufficiently large number of atoms was available to overcome the eventual interference

from the stable isobar background, and N is often calculated from the production cross sections of the radioactive atoms with rather large uncertainties. AMS can allow one to measure an absolute radioisotope to stable isotope ratio with high precision. But also the absolute decay rate has to be measured with high accuracy.

So far, five half-lives have been redetermined with AMS: ^{32}Si , ^{44}Ti , ^{60}Fe , ^{10}Be and ^{41}Ca .

a) ^{32}Si

Early AMS measurements of the ^{32}Si half-lives (101 ± 18 yrs[25] and 108 ± 18 yrs[26]) agreed remarkably well, but they were clearly lower than the values from geophysical measurements (230 ± 40 yrs[27] and 276 ± 32 yrs[28]) and decay measurements (172 ± 4 yrs[29]) which were made over a period of four years. Recently a new half-life value of 133 ± 9 yrs was redetermined by improved AMS measurement[30]. These authors produced ^{32}Si -samples by proton activation of ^{37}Cl (as LiCl) by $^{37}\text{Cl}(p, 2p\alpha)^{32}\text{Si}$ reaction and by neutron activation of ^{31}P (as $\text{Mg}_2\text{P}_2\text{O}_7$) through $^{31}\text{P}(n, \gamma)^{32}\text{P}(n, p)^{32}\text{Si}$ reaction sequence. After chemical separation, purification and conversion to K_2SiF_6 , the specific activity of ^{32}Si was measured by liquid scintillation counting. The $^{32}\text{Si}/\text{Si}_{\text{tot}}$ ratio was measured by AMS. Four silicon isotopes ^{28}Si , ^{29}Si , ^{30}Si and ^{32}Si were measured quasi-simultaneously under as identical conditions as possible to determine quantitatively the mass fractionating in the complex tandem accelerator system. The estimated overall uncertainties is $\pm 7\%$ for the determination of both the specific activity and the isotopic ratio.

b) ^{44}Ti

The half-life of ^{44}Ti has been measured to be 54.2 ± 2.1 yrs[31] by AMS, somewhat higher than the previously reported values of 46.4 ± 1.7 yrs[32] and 48.2 ± 0.9 yrs[33]. ^{44}Ti was produced by means of $^{45}\text{Sc}(p, 2n)^{44}\text{Ti}$ reaction. This new value was obtained from the specific activity and the radioisotope concentration $^{44}\text{Ti}/\text{Ti}_{\text{tot}}$ of several TiO_2 samples. An overall sensitivity of 10^{-10} for $^{44}\text{Ti}/\text{Ti}_{\text{tot}}$ ratio measurement was achieved in this work.

c) ^{60}Fe

The only measurement of the half-life of ^{60}Fe in the past was performed in 1957[34] giving a value of 3×10^5 yrs with an uncertainty of a factor of three. In this work the number of ^{60}Fe atoms was estimated from spallation cross section systematics of high-energy protons in copper.

A new, longer half-life of ^{60}Fe has been measured by AMS with the Argonne FN type tandem-superconducting linear accelerator[35]. ^{60}Fe was again produced by spallation reactions of copper. A value of $(1.49 \pm 0.27) \times 10^6$ yrs was obtained from measurements of the specific activity and the absolute $^{60}\text{Fe}/\text{Fe}_{\text{tot}}$ ratio.

d) ^{10}Be

The previously reported values for the ^{10}Be half-life ranged from 1.48×10^6 yrs to 1.9×10^6 yrs with an uncertainty from 10% to 24%. [36-38] Hoffmann et al. [39] presented a more accurate value of $(1.51 \pm 0.06) \times 10^6$ yrs with a smaller uncertainty of 4% for the ^{10}Be half-life with AMS measurement.

The key to improvement in this work was adoption of a very well standardized ^{10}Be solution as a starting material. The standardized solution of ^{10}Be called ORNL-MASTER solution was originally made from BeO at Oak Ridge National Laboratory. The $^9\text{Be}/^{10}\text{Be}$ weight ratio of BeO was precisely measured as 0.576 ± 0.006 by the secondary ion mass spectrometry (SIMS). A complicated procedure for dilution of ORNL-MASTER solution with water and inactive ^9Be to make a working solution was described. Two independent dilution series produced new AMS-standards with $^{10}\text{Be}/^9\text{Be}$ ratios of the order of 10^{-10} and 10^{-11} . Specific activity of the working solution was measured by liquid scintillation counting.

e) ^{41}Ca

In 1974, a rather precise half-life of $(1.03 \pm 0.04) \times 10^5$ yrs was reported[40] as the result of a specific activity measurement. Using recent information on neutron capture cross-section and X-ray fluorescence yields, and a more realistic estimate of the uncertainties, this half-life has been corrected to $(1.13 \pm 0.12) \times 10^5$ yrs[41].

In 1991, Kutschera et al.[42] reported on an AMS measurement of the half-life of ^{41}Ca . They used a sample of enriched calcium material which ^{41}Ca abundance (1.237%) was determined directly by AMS. The activity was measured with a Si(Li) detector via the emitted 3.3 keV X-rays. The resultant $t_{1/2} = (1.01 \pm 0.10) \times 10^5$ yrs is in excellent agreement with the very recent AMS measurement value, $t_{1/2} = (1.03 \pm 0.07) \times 10^5$ yrs, of Klein et al., who determined the half-life from the decay of ^{41}Ca relatively to ^{36}Cl in Antarctic meteorites of different terrestrial ages[43].

From these three independent half-life measurements, a weighted mean of $t_{1/2} = (1.04 \pm 0.05) \times 10^5$ yrs is calculated, which is recommended as a new accurate value of the ^{41}Ca half-life.

2. Cross sections of nuclear reactions

Many production cross sections of radionuclides can be measured by radiochemical techniques. However, when the half-lives become too long for an efficient decay measurement, it is appropriate to count the atoms directly by AMS, which is independent of the half-life. Of particular interest are cross-section measurements relevant for cosmic-ray production of very long-lived radionuclides in lunar rocks, meteorites, and terrestrial materials. These cross sections are critical for interpreting the information obtained from concentration measurements of the same radionuclides in the mentioned materials.

In an early measurement with AMS the cross section of the reaction $^{26}\text{Mg}(p, n)^{26}\text{Al}$ was measured[44] by bombarding ^{26}Mg foils with protons, adding ^{27}Al for normalization, and measuring $^{26}\text{Al}/^{27}\text{Al}$ ratios in the range of 10^{-12} . It was quite remarkable that in spite of $^{26}\text{Mg}/^{26}\text{Al}$ ratios of around 10^{10} no chemical separation of ^{26}Mg from ^{26}Al was necessary, because ^{26}Mg did not form stable negative ions in the ion source.

The spallation cross section of 165 MeV protons on nickel producing ^{60}Fe was measured[23], and a preliminary value of about 0.2 mb was found. Spallation reactions are of interest for the cosmic ray production of ^{60}Fe in iron meteorites which typically contain about 10% nickel. A main technical problem in these experiments is the separation of ^{60}Fe from a very strong, ever-present ^{60}Ni background. Since the two ion species are not discriminated in the acceleration process, they arrive with the same energy of about 360 MeV at the final detection system. By replacing the Al foil stack by a vacuum chamber of a magnetic spectrograph filled with nitrogen gas of a few torr pressure, the ions experienced many charge changes and different energy losses during their 3m flight path through the spectrograph. Thus, ^{60}Ni was obviously suppressed by this arrangement. The $^{60}\text{Fe}/\text{Fe}_{\text{tot}}$ ratio was decreased from 1×10^{-7} to 5×10^{-9} . In an ^{60}Ni enriched sample, the $^{60}\text{Fe}/\text{Fe}_{\text{tot}}$ ratio of 7×10^{-12} could be observed.

The cross section for ^{26}Al produced through $^{27}\text{Al}(p, pn)^{26}\text{Al}$ reaction was determined[45]. From the measured $^{26}\text{Al}/^{27}\text{Al}$ ratio of 1.61×10^{-13} at a bombarding proton energy of 159 MeV, the resulting cross section value is 43.9 mb.

The cross sections of proton-induced spallogenic ^{41}Ca of elemental Ti, Fe and Ni targets were measured for energies ranging from 40 to 600 MeV[46]. Recently the thin-target cross sections of ^{10}Be produced by proton induced spallation reactions from the elements O, Mg, Al, Si, Mn, Fe and Ni were measured[47]. The irradiations were carried out with energies between 100 and 2600 MeV using various accelerators. $^{10}\text{Be}/^9\text{Be}$ ratio was measured at the ETH AMS-facility in Zurich.

Using a 4MV tandem accelerator at the University of Tokyo, the neutron induced formation cross sections up to $En = 38$ MeV for ^{14}C from oxygen and ^{26}Al from silicon were measured[48]. The excitation functions for the $^{16}\text{O}(n, x)^{14}\text{C}$ and nat. $\text{Si}(n, x)^{26}\text{Al}$ reactions (here, x means that the reaction channel is not well defined) were obtained from the neutron flux distribution and the measured number of atoms.

In order to accurately model ^{41}Ca production from solar cosmic ray proton interaction on Ti at the lunar surface, the nat. $\text{Ti}(p, 3pxn)^{41}\text{Ca}$ excitation function for $Ep = 35\text{-}150$ MeV was measured[49]. Proton excitation curves over the above energy range for ^{42}K , ^{43}K , ^{44}g , ^{44}m , ^{46}V , ^{47}V , ^{48}Sc and ^{48}V were also measured. The reaction cross sections for K, Sc, and V radionuclides were compared to previous measurements and agreed favorably at $E < 50$ MeV but were larger by 10-30% for the higher energies.

The $^{36}\text{Ar}(n, p)^{36}\text{Cl}$ reaction has been suggested as a source of the important cosmogenic radioisotope ^{36}Cl in the atmosphere. For the study of the ^{36}Cl production mechanism the slow neutron cross section of $^{36}\text{Ar}(n, p)^{36}\text{Cl}$ was determined[50]. An upper limit for the thermal neutron cross section of 1.5 mb was obtained. This value is three orders of magnitude lower than the value previously assumed.

3. Exotic particles

Kutschera published a good review paper on the application of AMS for the detection of exotic or hypothetical particles[51]. The term "exotic" characterizes particles which were previously not found in measurable quantities. This means that every search is probably bound to end in a negative result. The exotic particles mentioned here include free quark, superheavy elements, anomalously heavy isotopes, etc..

a) Free quark

The existence and confinement of quarks in hadrons is one of the most fascinating aspects of modern particle physics. When the first reports from Stanford were published[52] showing evidence for fractional charges on superconducting niobium spheres there was great hope of finding them elsewhere too. However, neither in accelerator experiments nor in cosmic rays nor in stable matter searches has any further evidence for free quarks been found. The main problem is that the mass of hypothetical free quarks is unknown. In AMS techniques, a fractionally charged particle has a unique behavior in accelerator and gives a specific signature in the beam analyzing processes, and a purely electrostatic system will bring all particles, independent of their mass from the ion source, to the final detector.

AMS experiments at the University of Toronto[53], the California Institute of Technology[54] and the University of Rochester[55] were performed for more general searches of fractionally charged particles in matter. In some cases[54] sensitivities of around 10^{-18} fractionally charged particles per normal atom were reached. So far no positive evidence for free quarks has been found.

b) Anomalously heavy isotopes [56]

The term anomalously heavy isotope is used for stable isotopes of known elements with masses far beyond what can be formed by simply adding neutrons. The anomalous mass results from binding a hitherto unknown heavy particle to the nucleus. These particles may be neutral or integrally charged with masses up to 10^5 amu. AMS searches at the FN tandem of the University of Pennsylvania, Philadelphia on anomalously heavy isotopes of O[57], He, Li and Be[58] have been done. The AMS searches for anomalously heavy isotopes of H at MeV energies with a cyclotron and a Van de Graaff accelerator were also performed[4,56,59]. These searches have established limits well below the abundance estimated from cosmological models. However, it is difficult to predict whether these primordial abundances will be preserved on earth. It is conceivable that depending on mass an appreciable depletion of these heavy isotopes may have occurred through their geophysical and geochemical history. The uncertainty as to where to search for these species seems to have discouraged further AMS attempts in recent years.

c) Superheavy elements and extinct radionuclides

Superheavy elements are expected to occur around $Z=114$ and $A=300$, with predicted half-lives varying by many orders of magnitude. AMS techniques require half-lives of at least 1×10^8 yrs for search of primordial superheavy elements. The most thorough experiment of this type was performed with the FN tandem in Philadelphia in a search for naturally occurring element $Z=110$, $A=294$ in its supposedly chemical homolog platinum[60]. Using a Pt nugget from Alaska as the source material and a time-of-flight (TOF) technique for identifying mass 294, an upper limit of 10^{-11} for $^{294}110$ in Pt was determined. In order to check an improved TOF technique for this superheavy mass region, an AMS experiment was suggested to confirm the existence of ^{244}Pu ($t_{1/2}=8.5 \times 10^7$ yrs) in nature originally detected by Hoffman et al.[61]. A ^{244}Pu concentration of 2.4×10^6 atoms/kg of Precambrian bastnatite, a rare earth fluorocarbonate mineral was found. The unexpectedly high ^{244}Pu concentration is very important for our understanding of the possible origin of very heavy elements.

4. Solar neutrinos [62]

The well-known solar neutrino problem is based on the discrepancy between the neutrino flux predicted by the standard solar model[63] and the result of the 20-year effort of Davis and collaborators[64] to measure this flux on earth. The prediction is high by about a factor of three and cannot be explained within our current knowledge of the sun.

A common problem in all solar neutrino detection experiments is the extremely low reaction rate. Only a few atoms are produced by neutrino interactions per year in one ton of target material. The exploratory AMS experiments for the detection of ^{98}Tc [65] ($^{98}\text{Mo}(\nu_e, e^-)^{98}\text{Tc}(4.2 \times 10^6$ yrs)) and ^{205}Pb [66] ($^{205}\text{Tl}(\nu_e, e^-)^{205}\text{Pb}(1.5 \times 10^7$ yrs)) have been reported. The long-lived radioisotopes ^{98}Tc and ^{205}Pb can accumulate over millions of years in stable minerals. ^{205}Pb , in particular, has a very low production threshold and a very long half-life. One kg of a suitable thallium mineral may well contain 10^5 ^{205}Pb atoms after a 10^7 years accumulation time[67]. A very efficient suppression of neighboring lead isotopes of about $1:10^{16}$ was found due to the combined selectivity of injection resolution, rf selectivity of linac, and beamline and spectrograph resolution. However, for real samples, much has to be improved for the isobar separation, since the $^{205}\text{Pb}/^{205}\text{Tl}$ ratio in the thallium mineral is estimated to be around 10^{-19} . Extensive chemistry will have to be done to bring this ratio to the level accessible by AMS.

5. Neutron dosimetry

$^{36}\text{Cl}/\text{Cl}_{\text{tot}}$ in mineral samples (i.e., concrete, granite, tiles) were measured using AMS with sufficient precision to quantify the thermal neutron activation levels at distances between 1000 and 2000 m from the hypocenter in Hiroshima[68]. The initial measurements showed that the activity of ^{36}Cl induced by bomb neutrons disagreed with calculations based on a specific A-bomb dosimetry system D586. This work has been carried on by a joint US-Japanese team aiming at reconstruction of the thermal neutron fluency as a function of distance from the hypocenters in both Hiroshima and Nagasaki.

B. GEOLOGY

1. ^{10}Be in ice cores

Any application of the cosmogenic nuclides as either tracers or dating tools requires the knowledge of the nuclide production rate variations through time. The production of cosmogenic nuclides on the earth is primarily a function of the galactic cosmic ray intensity, which is modulated by the variation of the solar activity and the earth's magnetic field. ^{10}Be concentrations in ice cores are extremely useful to study the historical changes of solar activity and the geomagnetic field.

The earliest AMS measurements on ^{10}Be concentrations in ice cores were carried out by Raisbeck et al.[69] and Beer et al.[70] at the Dome C site in the Antarctic and the Dye 3 site in Greenland, respectively. Variations of ^{10}Be concentrations, showing an 11-year periodicity and correlated with

sunspot activity, were observed. A series of ice cores from both the Antarctic and Greenland has been measured since then. Beer et al.[71] made ^{10}Be measurements in the Camp Century ice core, Greenland covering the past 10,000 years, and compared the ^{10}Be data with $\delta^{18}\text{O}$ and ^{14}C records. They concluded that short-term ^{10}Be fluctuations were mainly caused by changes in the production rate but not by climatic effects.

However, over the long-term, the ^{10}Be concentration variations can be seen to reflect climatic changes. A dramatic change, corresponding to the Younger Dryas Glacial cold phase/Holocene transition that occurred about 10,000 years ago, has been found at 250 m above bedrock in the Camp Century ice core. The ^{10}Be concentration in the Younger Dryas glacial period is considerably greater than that in the Holocene period, reflecting less dilution of ^{10}Be deposited during the cold and dry intervals.

2. ^{10}Be in seawater and pelagic sediments

A knowledge of the marine geochemical cycle of ^{10}Be is important in applying ^{10}Be as a geophysical tracer or geochronometer. Kusakabe et al.[72] measured the water column profiles of ^{10}Be at the San Nicolas Basin station in the Pacific. The data show a profile from a depth of 20-50 m (with ^{10}Be concentrations of less than 500 atoms/g) to deep water between 100 and 500 m (with a constant ^{10}Be concentration of ~1200 atoms/g). But relatively higher ^{10}Be concentrations of ~1100 atoms/g were observed close to the water surface, which may have resulted from the input of ^{10}Be from the atmosphere or from river water. The maximum values (1500-2000 atoms/g) reached near the sediment-water interface indicating the refluxing of ^{10}Be from the sediments into the water column. The residence times of ^{10}Be in the San Nicolas Basin (ranging from 0.5 yr in the surface layer to 30 yrs at the bottom) were also estimated.

Raisbeck et al.[73] has measured ^{10}Be in a marine sediment core sample, V16-58, taken at $46^{\circ}30'\text{S}$, $31^{\circ}16'\text{E}$ at a depth of 4731 m and having a sedimentation rate of -2.5 cm/kyr. They reported a ^{10}Be concentration increase by a factor of ~ 2 near the geomagnetic reversal boundary of Brunhes-Matuyama 730,000 years ago.

3. ^{10}Be in soil and continental sediments

^{10}Be is readily adsorbed on soil and other surficial materials after landing, preferentially on clay minerals and organic matter. Pavich et al.[74] measured a residual weathering profile in the Virginia Piedmont, USA. The vertical distribution of ^{10}Be shows a large enrichment in the top 320 cm of soil, followed by an exponential decrease to a depth of about 15 m. This result indicates that ^{10}Be is fixed by soil and subsequently has low mobility in the weathering profile. This affinity of ^{10}Be for near-surface soil particles enables its use as a tracer for soil erosion and sediment transport studies. Assuming that the site is in dynamic equilibrium of ^{10}Be deposition, radioactive decay and erosion, the denudation rate at the surface has been estimated as 0.9 mg/cm².yr (or 0.45 cm/kyr) from the measured ^{10}Be inventory.

The status of erosion can be evaluated using the ^{10}Be erosion index, namely, the ratio between number of atoms at output and input. Brown[75] has shown the results of a study of basin erosion in the eastern United States. The indices of Coastal Plain basins are all less than unity with an average value of 0.26, but the Piedmont samples have an average almost ten times higher. This can be explained from the fact that the large amount of soil set in motion in the Piedmont by intemperate farming in the preceding two centuries has not yet gone far into the Coastal Plain through the river water flow.

Valette-Silver et al.[76] measured ^{10}Be concentrations in cores collected in undisturbed estuarine sediments of three tributaries of the Chesapeake Bay and observed two increases in ^{10}Be concentrations coincident with the transition from forested land to European style farming. These features were interpreted as a result of the erosion, transport and sedimentation of ^{10}Be -rich soils in the mouths of the rivers.

^{10}Be in lake sediments was first studied by Raisbeck et al.[77] in Lake Keilambete, Australia and Windermere, England. They found both lakes to have concentrations in excess of 10^9 atoms/g, which they attributed to erosional transport. Later, Wahlen et al.[78] reported that ^{10}Be deposition rates for Green Lake, New York from the Period 1200 to 1490 AD were low and comparable to atmospheric production rates with no erosional component. They found a value three times higher for the period that followed, corresponding to the time of the Maunder Minimum in solar activity (1645-1715 AD). They also found higher ^{10}Be deposition associated with the erosion effects due to the introduction of European farming techniques during and after the 18th century.

4. Subducted sediments and island-arc lavas

It became clear during the 1960s that a cause-effect relation existed between subduction of lithospheric plates and island-arc volcanism, but the origin of the lavas of these volcanoes remained controversial. A layer of pelagic sediments, usually a few hundred metres thick, accompanied the plate during the early stages of subduction. Tera and Sacks suggested in 1978 that the high concentration of ^{10}Be known to exist in pelagic sediments (typically 5×10^9 atoms/g) might be utilized as a sediment tracer for figuring out the origin of the island arc lavas. The half-life of ^{10}Be is perfect for this study because it is long enough to trace the tectonic activity. Brown[75] reported on AMS measurements of 26 samples of non-island-arc volcanoes and mid-ocean ridge basalts, as a control group, showing no values greater than 10^6 atoms/g; but much higher concentrations (sometimes higher than 10^7 atoms/g) were encountered from lavas of volcanoes from 6 Pacific Ocean arcs. The results provided a direct evidence of incorporating of subducted pelagic sediment into island-arc lavas.

5. Origin of tektites

Tektites are silica-rich glassy objects found in Australia and Southeast Asia with the same age of 7×10^5 yrs, also in Europe (Moldawites, 5×10^6 yrs) measured by the K-Ar method. Most of tektites are a few centimetres in dimension and show evidence of at least two stages of melting. Of the various origins proposed, two are extraterrestrial: (i) they are the result of some object striking the Earth; (ii) they are in some way ejected from the Moon. But, a large majority of geochemists accept a terrestrial origin primarily because tektites are chemically and isotopically similar to the Earth and not the Moon. Tera et al.[79] measured the ^{10}Be concentration in 42 tektites by AMS and gave a range of $(1-3) \times 10^8$ atoms/g when corrected for their age of 7×10^5 yrs. Such concentrations are about two orders of magnitude lower than found at the surface of the Moon but similar to continental margin sediments. The $^{26}\text{Al}/^{10}\text{Be}$ ratio is a more critical and reliable parameter to distinguish between terrestrial and extraterrestrial substances. When ^{26}Al and ^{10}Be are produced in the atmosphere, the $^{26}\text{Al}/^{10}\text{Be}$ ratio is about 1000 times smaller than that resulting from production in solid materials and hence in space. Middleton and Klein[80] measured five tektites giving a limit for the $^{26}\text{Al}/^{10}\text{Be}$ ratio of less than 0.11. If tektites were produced in space, the $^{26}\text{Al}/^{10}\text{Be}$ ratio would have to have been in the range of 2.7-5.4 previously measured on some extraterrestrial bodies. Thus, the measured $^{26}\text{Al}/^{10}\text{Be}$ ratios are consistent with a terrestrial origin of tektites.

6. Platinum group elements

a) Examination of the hypothesis "dinosaurs extinction"

An Ir anomaly at the Cretaceous-Tertiary (K-T) boundary was initially found by Alvarez et al.[81] using neutron activation analysis (NAA) in 1980. They have ascribed it to the impact of an asteroid on the Earth about 6.5×10^7 yrs ago, which caused an unusual climate and the extinction of the dinosaurs. By NAA it is difficult to reach ppb (parts-per-billion) levels for Pt because of the small neutron capture cross section and the interference from trace amounts of Au.

Rucklidge et al.[82] measured Pt and Ir in samples of both marine and nonmarine origin at the K-T boundary by AMS. Ir was shown to be concentrated within the region of the marine K-T boundary with an abundance of the Cretaceous chalk. In the most Ir-enriched clay layer the Pt was also found to be

enriched, but at levels 10 to almost 100 times more than expected if both Ir and Pt were of meteoritic origin. Similar measurements on material from a nonmarine origin of the K-T boundary at Hell Creek, Montana has shown no matching anomaly in Ir and Pt. Chew et al.[83] extended AMS measurements to Os and isotopic ratios of platinum group elements(PGE). The measurements indicated an anomalous increase of ^{189}Os , ^{193}Ir and $^{194}, ^{195}\text{Pt}$ for the sample from the Danish K-T boundary at Stevns Klint, but the isotopic ratios of $^{194}\text{Pt}/^{195}\text{Pt}/^{196}\text{Pt}$ measured for the samples from Danish and Israel K-T boundaries, and $^{191}\text{Ir}/^{193}\text{Ir}$ and $^{188}\text{Os}/^{189}\text{Os}$ for the Stevns Klint K-T samples all agree with normal terrestrial composition. The results of AMS measurements have shown that the situation of "dinosaurs extinction" studies is more complex than originally speculated.

b) Re/Os ratio and mantle-crust differentiation

Another interesting case was the detection of the ^{187}Re - ^{187}Os isotope system to study extraterrestrial and crustal materials. ^{187}Re is a very long-lived radioisotope with a half-life of 4.6×10^{10} yrs which constitutes 62.6% of the abundance of natural rhenium, and its decay product ^{187}Os is a stable isotope, having a small abundance of 1.6% in nature. As a result of the decay of ^{187}Re , the primordial $^{187}\text{Os}/^{186}\text{Os}$ ratio changes in the planetary system as a function of time and the starting $^{187}\text{Re}/^{186}\text{Os}$ ratio. For a value of $^{187}\text{Re}/^{186}\text{Os}$ ratio of about 3.15[84], typical of meteorites and the Earth's mantle, the present $^{187}\text{Os}/^{186}\text{Os}$ ratio is about 1. The Earth's continental crust has an estimated $^{187}\text{Re}/^{186}\text{Os}$ ratio of about 400, so that for a mean age of the continent of 2×10^9 yrs, a $^{187}\text{Os}/^{186}\text{Os}$ ratio of about 10 is expected[85].

Researchers at the University of Rochester presented the results for the isotopes ^{187}Os , ^{188}Os , and ^{189}Os measured in sub-ppb level samples by AMS[86,87]. Measurements on extraterrestrial and crustal materials with Os concentrations between 2 ppm (parts-per-million) and 0.006 ppb showed a detection limit of 0.01 ppb, when Os samples were preconcentrated with a nickel sulfide method. The $^{187}\text{Os}/^{186}\text{Os}$ ratios measured for samples from the Canyon Diablo meteorite and from the East Clearwater crater in Quebec, Canada were close to 1, typical of extraterrestrial material. In contrast, ratios in samples from the Ries crater, Germany were around 10, which is evidence of predominantly crustal osmium in these rocks.

c) Trace element concentrations in minerals

Rucklidge et al.[88] have measured PGE in a variety of rocks and sulfide concentrates using a MP tandem accelerator. In quantitative analysis of standards, levels as low as 10 ppt (10^{-11}) of PGE could be measured in about 10 minutes, but the reproducibility and stability of the data has proved to be a problem. This might be attributed to inhomogeneous samples and could presumably be improved by using a microprobe primary ion beam with which individual grains can be studied[89]. Several years later, Rucklidge et al.[90] have used AMS for the microanalysis of trace elements in minerals. The *in situ* measurement of PGE, Au and Ag, down to the sub-ppb level could be performed on milligram-sized grains of minerals, but only a larger microarea of $\sim 400 \mu\text{m}$ was detected.

C. HYDROLOGY[10,91]

For groundwater management it is important to study the hydraulic characteristics, flow paths, and solute transport. Many regions of the world rely upon groundwater for drinking water, agricultural activities, and industrial uses. Since the first AMS measurements of ^{36}Cl in 1978, this radionuclide has proved to be a versatile tracer of hydrologic processes. ^{129}I produced naturally also appears to be very useful for studying hydrologic processes, although sample collection, preparation, and interpretation are much more complicated than in the case of ^{36}Cl .

The range of hydrological applications demonstrated for ^{36}Cl and ^{129}I mainly covers: delineation of water flow and infiltration, evaluation of ion filtration and salt dissolution as sources of groundwater salinity, estimation of environmental impact of bomb-produced ^{36}Cl and industrial discharges of chloride, and investigations of nuclide migration behaviour relevant to radioactive waste repository siting. The typical applications are introduced below.

1. Delineation of water flow by tracing

Elmore et al.[10] postulated to use ^{129}I can be used for tracing the slow movement of seawater through sediments in response to hydrothermal convection in the underlying oceanic crust. Determination of flow rates in long-lived, low-temperature convection cells is essential for understanding the elemental budgets of the oceans.

2. Ion filtration

^{36}Cl was used to determine that the solutes in the distal end of the Milk River Aquifer, Alberta, Canada originated by infiltration rather than retention of initial ancient water[92]. Ion filtration is a process whereby water leaks out of the system through compacted geological formations but leaving part of its salts behind. The increase in the ^{36}Cl concentration in the aquifer may be explained by ion filtration by which the total chloride concentration is reduced. These are persuasive arguments against a mixing with the initial ancient chloride which would leave the ^{36}Cl concentration unaffected. Similarly, the significant increase of ^{36}Cl along the flow path of the groundwater in the aquifer formation of Southern Maryland, U.S.A.[93] is also interpreted as a result of ion filtration.

3. Salination

The process of salination of water systems was studied using ^{36}Cl by AMS[94]. Chlorides content originating from young rainwater and from subsequent evaporation were distinguished from those generated by the leaching of ancient rocks. Results for the Jordan River/Dead Sea system showed that the amount of chloride leached from rocks ranged from 70% in source springs to >90% in water bodies downstream. The amount of water left after evaporation decreased from 50% in the source spring to 20% in the intermediate Lake Kinneret. In the terminal Dead Sea 99% of the chloride originated from ancient rocks and evaporite formations while 80% of its ^{36}Cl content was of meteoric origin.

4. Bomb ^{36}Cl as tracers

Thermonuclear weapon tests at ocean sites in the mid-1950s produced a large amount of ^{36}Cl through neutron capture in ^{35}Cl in seawater. The fallout pulse of bomb ^{36}Cl released to the environment peaked at 2-3 orders of magnitude above cosmogenic levels[91], and returned to near pre-bomb levels in the mid-1960s. This man-made pulse of ^{36}Cl is an excellent tracer for measuring various hydraulic characteristics. A prominent bomb-pulse was detected at a depth of one meter in a sandy loam in arid Socorro, New Mexico, indicating a net salt infiltration from groundwater to the depth of 2.5 mm/yr[95]. An apparent hydrodynamic dispersion coefficient can also be estimated from the spread of ^{36}Cl concentrations in this soil profile.

5. Nuclide migration studies for siting of nuclear waste repositories

AMS using ^{36}Cl and ^{129}I has played a unique role in nuclear waste disposal studies. Such studies include the evaluation of waste repository siting by assessment of nuclide migration by groundwater from the proposed repository site, and the study of natural analogues imitating the natural conditions on a large scale and very long periods.

The migration of ^{36}Cl and ^{129}I from uranium minerals to groundwater along the fractured system of the Stripa granite in Sweden has been extensively studied[91,96]. Chloride concentrations in the groundwater increased with depth down to about 815 m, where a peak of 630 mg/l was observed. Meteoric recharge contained less than 5 mg/l, thus this Cl must have been derived from the higher neutron flux associated with uranium minerals within the granite. $^{129}\text{I}/\text{I}_{\text{TOT}}$ ratios in Stripa waters were enriched by 30 to 200 times over that in meteoric recharge.

A recent report by Fabryka-Martin et al.[97] indicated that the accumulation of ^{129}I in the water of Stripa was attributed to subsurface production by spontaneous fission of ^{238}U in micro-fractures in the granite matrix, and subsequent ^{129}I transfer by diffusion into fractures constituting the groundwater flow system of the granite. The authors expected that it would be an analogue for the leaching of such fission products from buried nuclear waste.

Fabryka-Martin et al.[98] reported on a natural analogue study of ^{129}I migration in uranium ore deposits at the Alligator Rivers region, Northern Territory, Australia. At the Koongarra deposit, the uraninite is hosted in quartz-muscovite-chlorite schists. The groundwater flows downwards to the schists from sandstones. From 1983 to 1985, iodine was extracted from groundwater through sampling boreholes in the vicinity of the deposit. The contents of ^{129}I were analyzed by AMS. The ^{129}I concentrations in the ground water decreased with increasing distances from the deposit due to by sorption. The results showed an apparent sorption distribution ratio (R_D) of 300 ml/g which was much higher than the ratios reported in laboratory studies.

Researchers at the Lawrence Livermore National Laboratory[99] reported on measurements of low concentrations of ^{129}I and ^{36}Cl in groundwater samples used to study nuclide migration at the Nevada Test Site (NTS), a potential disposal site for high-level nuclear wastes in U.S. The characterization of water movement in an unsaturated tuff zone at Yucca Mountain, Nevada was made by ^{36}Cl measurements. The unexpected result of this work was the observation of bomb pulse ^{36}Cl as deep as 152-153 m below the ground surface, which was interpreted as evidence for the presence of modern water flow through faults and fractures.

The distribution of anthropogenic ^{36}Cl in the unsaturated aquifer zone was used to estimate the water flux at a study site in the Chihuahuan Desert, Texas for a potential low-level radioactive waste disposal site[100].

D. ENVIRONMENTAL SCIENCES

Nuclear weapon tests at ocean sites and in the atmosphere in the mid-1950s produced a large amount of ^{36}Cl in seawater as well as ^{14}C via $^{14}\text{N}(n, p)^{14}\text{C}$ reaction in the atmosphere. The ^{14}C activity in the northern-hemispheric atmosphere is now about 100% higher than its natural level. It is decreasing gradually in response to the incorporation of CO_2 into carbon reservoirs of the oceans and biosphere.

Both bomb-produced ^{36}Cl and ^{14}C are useful tracers in environmental sciences including oceanic and atmospheric studies.

1. ^{10}Be in atmosphere

^{10}Be is mainly produced in the upper atmosphere by cosmic rays and then delivered to the Earth's surface primarily by rain and snow. Raisbeck et al.[101] have measured its concentration at latitude 65°N in both stratospheric and tropospheric air. Stratospheric concentrations are 7×10^6 atoms/ m^3 at an altitude of 10.7 km, increasing to 1.3×10^7 atoms/ m^3 at 19.2 km, whereas the tropospheric concentrations are lower by more than two orders of magnitude. Measurements of the $^{10}\text{Be}/^7\text{Be}$ ratio suggest that the residence time of Be-bearing aerosol particles is 350 days in the stratosphere and 20-35 days in the troposphere.

^{10}Be is removed from the atmosphere by precipitation scavenging of aerosol particles. Monaghan et al.[102] collected precipitation in continually open containers for about one year at seven sites around the United States. After correction for recycled ^{10}Be , which is introduced by soil particles contamination (8-35% of the total ^{10}Be), the global-average ^{10}Be production rate was assessed at 1.21×10^6 atoms/ cm^2yr . Brown et al.[103] reported that the weekly rain collections in Mauna Loa, Hawaii where the concentration by dust was negligible showed a sufficiently great variation in ^{10}Be concentrations from 1.90×10^3 to 8.94×10^4 atoms/g rain water. It is evident that a few high-

concentration rains dominate and large uncertainties occur. The researchers demonstrated that the mean value of the ^{10}Be deposition rate in temperate latitudes is 1.53×10^4 atoms/g rain water with an uncertainty of 20%.

2. CO_2 transfer from atmosphere to ocean and biosphere

One of the important environmental problems facing mankind today is the global warming that is imposed by a green-house effect caused by the rise of the CO_2 concentration in the atmosphere. The capability of the ocean to remove excess CO_2 from the atmosphere is a factor against this danger. Thus a study of the exchange of CO_2 labeled with bomb ^{14}C between atmosphere and ocean by means of AMS is very valuable.

^{14}C concentrations of annual tree rings (1945-1983) of a Kiso hinoki tree grown in the Gifu prefecture, and of acid-insoluble carbonaceous compounds extracted from sediment cores of Lake Biwa in the Shiga prefecture, Japan have been measured with AMS techniques[104]. An increase in ^{14}C resulting from bomb ^{14}C was found for both, tree rings and cored sediments. A primitive model of carbon exchange between the troposphere and surface ocean water or the biosphere was applied to the annual changes in ^{14}C of the hinoki tree rings and also of coral rings from Florida, U.S.A.. The time required to transfer total bomb ^{14}C from the troposphere to surface sea water and to the biosphere was estimated to be about 11 and 23 years, respectively.

French researchers reported [105] that 34 measurements of $\Delta^{14}\text{C}$ have been performed on water samples of 100 ml which was collected in the tropical Indian Ocean in 1986. This work has provided a water column inventory of the bomb ^{14}C from atmosphere (7.7×10^9 - 1.6×10^{10} atoms/cm²) and an estimate of ^{14}C penetration into the ocean during the last eight years which exhibits a maximum value around a depth of 300-500 m.

3. Ocean circulation

Since the pattern and rate of the ocean circulation are strongly coupled to climate, understanding of ocean circulation is of great practical importance. The radiocarbon in dissolved carbonate is an excellent tracer to gain information on the long time scales of transport processes in the bulk ocean. For example, the inventory of ^{14}C in the Atlantic[106] has shown that the upper waters carrying the bomb ^{14}C are pushed to the north, sinking in the Nordic Seas, then moving out southward at depth towards the Atlantic. However, the large volume sampling of deep seawater is a slow and very expensive procedure. Using AMS, the seawater sample size has been reduced from 250 liters for decay counting down to 250 ml or even less with nearly the same accuracy, $\pm 0.3\%$.

Recently an AMS facility dedicated to oceanography has been established at the Woods Hole Oceanographic Institution, U.S.A.. An ability of measuring up to 4500 ^{14}C samples per year with the accuracy of 0.3-0.5% for modern seawater samples is expected for the two international projects WOCE (World Ocean Circulation Experiment) and GOFS (Global Ocean Flux Study)[107].

4. Source of atmospheric methane

CH_4 is a strong infrared absorber and an important greenhouse gas. The direct contribution of methane to the anthropogenic greenhouse effect was estimated to 15%, second only to CO_2 , 60%[108]. Lowe et al.[109] measured the ^{14}C of atmospheric methane samples, which were obtained from clean air sampling stations sited on a south-eastern point of New Zealand's North Island, and concluded that about one fourth of atmospheric methane may be fossil in origin. Wahlen et al.[110] measured the ^{14}C concentrations in atmospheric CH_4 and showed that 21% of the annual input is derived from fossil fuel sources and the remainder from biogenic sources including 25-26% from wetland, peat bogs and tundra, 23-24% from rice production, 19-22% from ruminants and 9-10% from biomass burning.

5. Source of atmospheric aerosols

Source apportionment of atmospheric aerosols is important from the perspective of regional health, visibility impairment and climate. Health effects arise from mutagens and carcinogens, i.e., polycyclic aromatic hydrocarbons (PAH) in aerosols; visibility and climatic effects relate to light scattering and absorption as well as cloud nucleation by aerosols[111]. The source of combustion carbon include natural fires, agricultural burning, land clearing, residential wood combustion, and the combustion of fossil fuel. AMS ^{14}C measurements by Currie et al.[112] have yielded direct and reliable discrimination of the fossil and contemporary carbonaceous sources. The techniques related to the employment of 10-100 micrograms carbon samples and special Standard Reference Materials for atmospheric urban particulate were presented. In order to obtain the information on the particulate source signatures and a quantitative apportionment of source intensities, a method combining ^{14}C data with multivariant chemical data has been used[111].

6. ^{36}Cl fallout in Arctic ice

Recent ^{36}Cl measurements have been made in an Arctic ice core drilled near the Dye-3 site (65 11'N, 43 50'W) by Woelfli et al.[113]. The samples analyzed cover the period between 1945 and 1985 with a resolution of one year. Due to the release of bomb ^{36}Cl into the atmosphere, the data show a peak in the late fifties with ^{36}Cl fallout rates about three orders of magnitude higher than expected from cosmic ray production. A stratospheric residence time for bomb ^{36}Cl of 2 ± 0.3 yrs was derived from the data.

7. ^{129}I from Chernobyl accident

The Chernobyl nuclear reactor accident has injected into the atmosphere a pulse of a large number of radionuclides. About 10-20% of the volatile radionuclides, present in the fuel, mainly iodine (2.6×10^{17} Bq for ^{131}I), caesium and tellurium were released. Concentrations of ^{129}I in rainwater samples collected after the accident in some areas were measured by AMS[114]. They amounted in the Munich area and in Israel during the fallout period to 2.6×10^{10} and 1.2×10^9 atoms/l respectively, while in 1982 a rainwater sample from Israel showed a ^{129}I concentration of 8.2×10^7 atoms/l.

8. Leakage of FeCl_3 wastes to fresh water

The application of the AMS technique has been extended to water pollution problems. The leakage of industrial acidic wastes containing FeCl_3 to an overlying fresh water aquifer in western Tennessee was investigated by ^{36}Cl measurements[115]. No contamination by acidic chlorides was found.

9. ^{36}Cl in reactor water

The cooling water of reactors and ion exchange resins in the water purification system, contain fission and neutron activation products. Among them are long-lived nuclides such as ^{14}C , ^{36}Cl , ^{79}Se , ^{99}Tc , ^{129}I , and some transuranium elements which play a prominent role in radioactive waste disposal. Because of their low concentrations and low decay energies, quantitative measurements of these nuclides are very difficult. So far accurate measurements of these nuclides in reactor cooling water and in ion exchange resins were lacking.

Liu et al.[116] have used two methods, AMS and liquid scintillation counting, to measure the concentrations of ^{36}Cl of reactor cooling water and ion exchange resins in nuclear power plants. The results from these measurements were several ppt of ^{36}Cl in cooling water and several ppb in ion exchange resins. The data obtained were in consistence, within an order of magnitude, with the predicted value for a nuclear power plant.

E. MATERIAL SCIENCES

1. Trace elements in semiconductors

Semiconductor devices are playing an increasingly important role in various fields of modern life. As the size of semiconductors decreases, the purity of semiconductor materials becomes more important. Dopants and impurities in semiconductors at very low concentration levels are commonplace. Thus, there is strong need for a quantitative assessment of the contents of the trace impurities and their effects on the electronic performance of the semiconductors.

The detection of trace elements in semiconductors by AMS technique has been initiated by Anthony et al.[117]. They examined several impurities, i.e. B, Be, Si, Nb, Sb and Te in Si and GaAs. However, the background level for most of these impurities was also a trouble problem. This may be due to contamination in the ion source from other previous measurements. For Nb, the background level was quite low and the detection limits of 0.1 ppb resulted. In another study[118], the tandem AMS at the University of Arizona was used to detect dopants and other impurities in Si and GaAs crystals. B-, P-, As- and Sb- doped Si crystals as well as B- and Cr- doped GaAs crystals were examined. The results showed that the background ranged from 0.5 ppb for B in Si and in GaAs, and 40 ppb for Cr in GaAs; the background for other impurities lay in the 1-10 ppb range. This experiment demonstrated again that the major constraint on the sensitivity of the system was the ion source contamination. With some improvements, absolute detection limits for elements in a "clean" ion source will reach the ppt range for most impurity-substrate combinations.

A tandem AMS system designed for analysis of electronic materials was developed and installed at the University of North Texas[119,120]. The ultimate goal of the project was to provide a detection capability at the ppt level for most elements. For this purpose, a system with a custom-built "clean" ion source, incorporating high-resolution magnetic mass analysis ($M/dM > 350$) and electrostatic analysis ($E/dE > 700$) of the sputtering beam, ultraclean slits, and a surface barrier detector was constructed.

Using this AMS system, Anthony et al.[120] have first presented the multi-element mass scans from bulk materials of graphite Si and CdZnTe. In this study, Ti, V, Cr, Fe, Co, Ni in bulk CdZnTe, O, F, Na, Al, Si, Cl, K, Ti, V, Cr, Fe, Ni, Cu in bulk graphite and Li, B, C, O, F, Na, Al, P, Cl, K, Ti, Cr, Fe, Ni, Cu, Ge, As in bare Si were analyzed.

Some other impurities such as nitrogen and chlorine may also affect the electronic properties of semiconductors. Chlorine may diffuse into the silicon substrate from an insulating surface layer of borophosphosilicate glass (BPSG), which was produced from boron trichloride. The presence of chlorine may also arise from sources such as oxidation with dilute HCl gas and from cleaning procedures involving a mixture of acids including HCl. Very little is known about the processes by which chlorine or nitrogen diffuses into silicon, and the effects of such elements on the device performance need to be explored[121,122]. Gove et al.[123] have applied the combination of neutron activation (NA) and AMS to detect quantitatively the depth distribution of chlorine and nitrogen in the silicon wafers. NA of the silicon wafers converted stable chlorine and nitrogen to the radioisotopes ^{36}Cl and ^{14}C , which were then measured by AMS. The combination of NA and AMS resulted in a good sensitivity for both Cl and N measurements comparable to the very best achieved by secondary ion mass spectrometry (SIMS).

So far the application of AMS in material sciences is still in an exploratory stage but the tests have demonstrated the large potential.

2. Trace elements in photographic emulsions

Rhodium, osmium and iridium are sometimes added in concentrations of 50 ppb to 50 ppm to silver halides during the production of advanced photographic emulsions[124]. Both Rh and Ir at low

ppb levels in silver halides have successfully been measured[123]. AMS may also be useful in profiling these elements in single-crystal silver halides to aid in understanding of structure-property relationships.

F. BIOMEDICAL SCIENCES

The possibility of using AMS in biological research was noted[125] as early as the first AMS conference in Rochester (1978), and an overview of AMS and some primary applications to biomedical research has been reported and suggested by Elmore[21]. Direct application of AMS to biomedical sciences appeared in the past years[126], and only recently, at the fifth International Conference on AMS in Paris (1990) pioneering work was published, displaying the great potential of AMS in this field.

The necessities and advantages of the application of AMS to biomedical sciences compared to conventional radioisotope tracer technique can be summarized as follows[127,128]:

(i). Radioisotope techniques are not suitable when the half-lives of radioisotopes of interest are shorter than ~1 h and longer than ~100 yrs. Short-lived radioisotopes are inconvenient to use, and very long-lived radioisotopes have low sensitivities in decay measurement. Carbon, aluminium, silicon, and chlorine have no ideal radioisotopes for conventional use, but they are adequate for the AMS technique.

(ii). For long-term studies of metabolism and pharmacokinetics, the short-lived radioisotopes are not long enough accessible to be useful. Short-lived radioisotopes would have to be administered in high activity which would promote radiolysis on the labeled compound, and deliver a significant radiation dose to the body or material under investigation.

(iii). Use of much smaller amounts (10 μg or even less) of labeled material presents new opportunities for biochemical tracing and human research.

Isotopes that appear to be most useful for biomedical research[128-130] by AMS are listed in Table II.

Table II. Isotopes for Biomedical Research by AMS

| Isotope | Application and Advantage (current and potential) |
|------------------|---|
| ^3H | Double labeling, more labeled atoms/molecule Cheaper ,Low natural background $1/10^{15}$ |
| ^{10}Be | Berylliosis study |
| ^{14}C | Universal biochemical label(not exchangeable with C atom <i>in vivo</i>) |
| ^{26}Al | Alzheimer's disease Renal failure Bone disease |
| ^{36}Cl | Neutron dose determination Halide metabolism |
| ^{41}Ca | Bone metabolism Cell growth regulation, calmodulin(a Ca containing protein) |
| ^{55}Fe | Iron absorption and blood disorder Plant-soil relationship |
| ^{79}Se | Selenium containing enzyme Substitute for sulfur in protein labeling |
| ^{129}I | Protein labeling Thyroid function tests Increase in radioimmunoassay sensitivity |

Biomedical applications of AMS with the isotopes ^{14}C , ^{26}Al and ^{41}Ca are briefly described below:

1. ^{14}C

a) Carcinogen adducts to DNA

Felton et al. of the Lawrence Livermore National Laboratory, U.S.A. have performed very commendable work[129] using ^{14}C labeled compounds to show the effectiveness of AMS as a tool to quantify biologically important molecules at extremely low levels.

They have measured the adduction of an environmental carcinogen 2-amino-3, 8-dimethylimidazo [4,5-f]-quinoxaline (MeIQX) found at the ppb level in cooked meat, to DNA at concentrations relevant to human exposure. Currently, the most sensitive technique for adduct detection is the ^{32}P -postlabeling assay that has permitted quantitative measurement at levels of 1 adduct/ 10^8 nucleotides. AMS greatly enhanced the ^{14}C detection sensitivity, and thus enabled the measurement of ^{14}C -MeIQX to be made at a level of one adduct bound in 10^{11} to 10^{12} nucleotides in mice.

In a second set of studies 2, 3, 7, 8-tetrachlorodibenzo-*p*-dioxin (TCDD), an extremely toxic and potent carcinogen, was evaluated for its ability to form DNA adducts in mouse liver after a low dose exposure. TCDD used in this study was ^{14}C -labeled in all 12 of its carbon atoms providing over ten times more sensitivity than with the previous MeIQX-DNA adduct study. However, no TCDD related adducts are formed as measured by AMS, except for very small amount at the highest dose (which is close to lethal levels of TCDD for the species), suggesting that TCDD carcinogenesis is not mediated by formation of TCDD-DNA adducts.

b) Rare protein in sperm cells

In another experiment, AMS was used to detect the presence of rare proteins, a mutant form of protamines, in human sperm. This has important implications for fertility, because protamines "turn on and off" the expression of the sperm's genes by binding to DNA. Approximately 1-10 mutant sperm containing specific forms of protamine could be isolated from 10^6 - 10^7 total sperm by means of flow cytometry. This small number of isolated cells (100-1000) was post-labeled with ^{14}C , mixed with non-radioactive protamine, and separated by electrophoresis. The resulting bands containing protamine were identified by comparing for their very low level of ^{14}C with AMS. Thus, AMS could be used to verify the identity of an extremely small amount of biological material.

c) Other possibilities of application

In addition, studies are underway to measure the molecular targeting of cancer chemotherapeutics in human tissue and to pursue applications for receptor biology. In particular, studies on these very low levels of receptor proteins, will benefit from the high sensitivity and small sample size in AMS.

In these studies special precautions should be taken against the severe problem of contamination[127]. Most biochemical laboratories have used radioactive isotopes as tracers and their facilities have usually been contaminated with unacceptably high levels of these tracers, so that careful protocols and/or new facilities are required to solve the contamination problem.

2. ^{26}Al

Aluminum is suspected of being a toxic element, particularly in connection with renal failure[131,132] and possibly also Alzheimer's disease[133,134]. However, relevant research has been hampered by the lack of a suitable radioisotope for tracer studies. The isotope ^{28}Al has a half-life of only 2.3 minutes and is therefore too short-lived for biological studies. Besides, measurements of trace amounts of stable Al are difficult and liable to contamination since the Al content of many laboratory reagents is higher than that found in brain tissue (1-4 $\mu\text{g/g}$)[135]. Now, the use of ^{26}Al as a marker with the new technique of AMS has provided a new opportunity to enter the field.

Meirav et al.[136] collected urine and blood samples from a rat over a three week period following the administration of 0.35 ng of ^{26}Al . ^{26}Al concentrations in these samples as a function of time after injection were measured in terms of $^{26}\text{Al}/^{27}\text{Al}$ ratio at the AMS facility at the University of Pennsylvania. Similar experiments are expected to be feasible in humans since the administration of 30 pCi of ^{26}Al entails a radiation dose of less than 0.1% of the natural background[135].

Barker et al.[137] have employed ^{26}Al to estimate gastrointestinal absorption of aluminum and characterize aluminum transporting proteins in blood. ^{26}Al was administered orally to a human subject. The presence of ^{26}Al in one particular blood protein, transferrin, has unambiguously demonstrated the nature of the aluminum carrier protein. The authors also applied ^{26}Al as a tracer to study the uptake of Al by human neuroblastoma cells in culture that might be related to Alzheimer's disease. This work was conducted at 20 MV tandem Van de Graaff accelerator at Daresbury, UK with fully stripped aluminum ions, Al^{13+} . Discrimination of ^{26}Al from ^{26}Mg is better than 1:10⁹. An estimated absolute detection limit for ^{26}Al was 10⁻¹⁸ g ($\sim 6 \times 10^4$ atoms) with a limit of $^{26}\text{Al}/^{27}\text{Al}$ ratio 10⁻¹³.

3. ^{41}Ca

Metabolism of calcium is currently of great interest because of the widespread incidence of the metabolic bone disease osteoporosis (loss of bone mass) and the effects of space travel on the bone loss of the skeletal system.

Elmore et al.[138] reported the use of ^{41}Ca as a tracer for studying calcium metabolism in dogs with AMS. Fourteen dogs were pre-labeled simultaneously with ^{41}Ca and ^{45}Ca (half-life 165 days), and both isotopes in blood, urine and feces were measured until ^{45}Ca became unmeasurable (about 6 months), and thereafter following ^{41}Ca in one dog for a year or more. It was demonstrated that ^{41}Ca behaved identically to ^{45}Ca *in vivo* and could be used as a tracer for calcium metabolism studies with an improvement in sensitivity of 10² over ^{45}Ca . The animal tests indicated the feasibility of using ^{41}Ca with AMS in studying human bone resorption over multi-year periods. Middleton et al.[139] presented the feasibility studies on ^{41}Ca concentrations in modern bones at the University of Pennsylvania. The concentrations of ^{41}Ca in bones of human, sheep, lions and zebra were determined. Furthermore, the variability of $^{41}\text{Ca}/\text{Ca}_{\text{tot}}$ ratios in contemporary bone samples in relation to species and location were studied.

III. COMPARISON OF AMS WITH OTHER ANALYTICAL TECHNIQUES

During the last couple of decades, a number of modern analytical techniques have been developed. Every technique has its characteristics which may meet the specific demands of an analytical task. In other words, none of these sophisticated techniques can solve all types of analytical problems. In order to assess the performance of AMS in inorganic analysis, we make a comparison of AMS with some other analytical techniques.

A. DETECTION LIMITS

The particles analyzed by AMS can be divided into three categories[140]: (i) stable isotopes (elements); (ii) radioisotopes; and (iii) exotic particles. The detection limits of the three categories vary greatly depending on the corresponding level of background. The detection limits of stable-isotopes can hardly be lower than the 10⁻¹²(ppt) level due to the difficulties of establishing an ultra-clean condition in an accelerator environment. For radioisotopes the sensitivities expressed as the concentration ratios of radioisotopes to their stable isotopes are in the range of 10⁻¹⁴-10⁻¹⁵(see Table I). The lowest detection limits of $\sim 10^{-19}$ can be reached in searches for exotic particles (see section II.A.3) since the parameter space of mass and charge (e.g. fractionally charged particles), where no known species exist, is scanned.

1. Long-lived radioisotopes analysis

Long-lived radioisotope analysis is based on the measurement of a very small isotopic ratio. Usually, small amounts of chemically prepared "pure" samples (in the form of element or compound) rather than the original samples are used in the ion source. The element-preconcentration is performed during the preparation of the AMS sample. Moreover, natural abundances of long-lived radioisotopes are usually very low in terrestrial materials. Hence, the contamination by the radioisotope itself from the environment is not a severe problem. The background may come from interferences of atomic and molecular species, stable-isobars and isotopes, which can be discriminated effectively by AMS.

Consequently, high sensitivities can be achieved in the AMS measurements of the radioisotopes (^{10}Be , ^{14}C , ^{26}Al , ^{32}Si , ^{36}Cl , ^{41}Ca , ^{129}I , etc.) for tracer studies. The equivalent detection limits for these elements are generally a few orders of magnitude lower than those obtained in other analytical techniques. In the ^{10}Be measurement, as a prominent case, the element beryllium (composed of 100% stable isotope ^9Be) has a low concentration in nature, and hence an extremely high equivalent sensitivity can be obtained. Kusakabe et al.[72] have detected ^{10}Be concentrations in seawater as low as -500 atoms/g (see section II.B.2), which is equivalent to a sensitivity of about 10^{-20} g/g. Also, an estimated detection limit for ^{26}Al of 10^{-18} g (6×10^4 atoms) was gained when Barker et al.[137] used ^{26}Al in medical tracer studies (see section II.F.2).

2. Elemental (stable isotopes) analysis

At present, AMS measurements of trace elements are concentrated on the studies of precious metals in minerals and impurities in semiconductors. The corresponding techniques are still under development.

a) Comparison with five analytical techniques

Table III lists the detection limits of different analytical techniques. In this table we select elements measured by AMS with well defined detection limits.

Table III. Comparison of detection limits (g/g) of different analytical techniques
in inorganic trace analysis

| | AMS | GF-AAS[141](m_o) | I-NAA[144] | PIXE | ICP-MS[149] | SIMS[150] |
|----|-------------------------|---------------------------|---------------------|--------------------------|---------------------|---------------------------|
| B | 10^{-12} - 10^{-9} | - | - | - | 8×10^{-11} | 9×10^{-10} |
| P | 10^{-12} - 10^{-9} | 3×10^{-8} | - | 2×10^{-6} [145] | 2×10^{-6} | 4×10^{-10} |
| Cr | 10^{-12} - 10^{-8} | 2×10^{-11} | 3×10^{-9} | 8×10^{-7} [146] | 2×10^{-11} | 2×10^{-10} |
| As | 10^{-12} - 10^{-9} | 1×10^{-11} | 4×10^{-9} | 1×10^{-7} [145] | 4×10^{-10} | 2×10^{-9} |
| Nb | 10^{-12} - 10^{-10} | - | - | - | 2×10^{-11} | 3×10^{-9} |
| Rh | 10^{-11} | 1×10^{-10} [142] | 1×10^{-9} | 1×10^{-6} [147] | 2×10^{-11} | - |
| Ag | 10^{-11} | 1×10^{-11} | 3×10^{-9} | 9×10^{-6} [148] | 4×10^{-11} | 1×10^{-9} |
| Sb | 10^{-12} - 10^{-9} | 2×10^{-10} | 9×10^{-10} | 1×10^{-5} [146] | 2×10^{-11} | 7×10^{-8} |
| Te | 10^{-12} - 10^{-9} | 1×10^{-10} | 6×10^{-9} | - | 4×10^{-11} | - |
| Os | 10^{-11} | 1×10^{-8} [143] | 5×10^{-10} | 1×10^{-6} [147] | 1×10^{-11} | - |
| Ir | 10^{-11} | 3×10^{-9} [143] | 9×10^{-12} | 1×10^{-6} [147] | 6×10^{-11} | - |
| Pt | 10^{-11} | 1×10^{-9} [143] | 5×10^{-9} | 5×10^{-9} [147] | 8×10^{-11} | 1×10^{-10} [151] |
| Au | 10^{-11} | 7×10^{-11} | 3×10^{-11} | 5×10^{-9} [147] | 8×10^{-11} | 1×10^{-10} [151] |

Note: GF-AAS: Graphite furnace atomic absorption spectrometry. Sensitivities are in terms of characteristic mass m_o , g/g per 1% absorption; I-NAA: Instrumental neutron activation analysis, detection limits are calculated for a thermal neutron flux is 4×10^{12} $\text{cm}^{-2} \cdot \text{s}^{-1}$ and an epithermal neutron flux is 8×10^{13} $\text{cm}^{-2} \cdot \text{s}^{-1}$. The sensitivities can be highly improved by radiochemical methods; PIXE: Proton induced X-ray fluorescence emission. * Detected with preconcentration by fire-assay; ICP-MS: Inductively coupled plasma mass spectrometry; SIMS: Secondary ion mass spectrometry.

Among the listed five other analytical techniques SIMS is a well developed technique with considerably high sensitivity in the analysis of trace impurities of the ultrapure materials[152]. However, the background problems limit the sensitivity of SIMS. The background arise mainly from the difficulty of eliminating molecular interferences from the atomic beam. AMS on the other hand has an effective molecular rejection through the charge changing and molecular dissociation accomplished during the stripping process at the tandem positive terminal[90].

For the purpose of comparing the sensitivities between AMS and SIMS, Donahue et al.[153,154] have detected a series of "standard" samples of different semiconductor materials containing known amounts of various impurities, including B in Si and GaAs; P, Ga, As and Sb in Si; Cr in GaAs, and Au in HgCdTe. The results show that the detection limits for AMS are lower than for SIMS by factors of 5-100. The AMS measurements at the University of North Texas[120] have demonstrated that molecular ions which are associated in SIMS with certain masses are completely removed in the AMS measurements.

b) Comparison with NTIMS and LRIS

Because of the lack of technical data, Table III does not include some other sophisticated analytical techniques, such as negative thermal ionization spectrometry (NTIMS) and laser resonance ionization spectrometry (LRIS or RIS).

The main advantages of NTIMS are high selectivities for various inorganic negative ions with very low interferences and low backgrounds, and an easy availability of high beam intensity of negative ions. To date, negative ions of C, B, N, Cl, Br, I, P, S, Se, Te, Mo, W, Tc, Re, Os and Ir have been measured[155,156].

In 1991, a group at the California Institute of Technology[157] reported on a technique applied to trace Os, Re and Ir determinations. NTIMS has been shown to be a very sensitive and accurate method. The detection limit for Os is estimated to be 10^{-14} g/g, which is lower than that of AMS by three orders of magnitude.

LRIS possesses extremely high sensitivity that its present detection limits are comparable to or even lower than AMS[158,159]. In theory, LRIS can detect an individual atom without any detection limit, but it has not been well realized owing to some technical problems.

B. OTHER FUNCTIONS

Table IV shows a comparison of fundamental performances and advantages/disadvantages of AMS with some analytical techniques.

In regard to the function of multi-element analysis, since only very few cases have been reported on AMS, no critical comparison can be made yet.

The preliminary study of *in situ* surface microanalysis made with AMS measurements[90] has not proven attractive, because the primary Cs sputter ion beam could not be well focused. It yields a larger *in situ* area of detection, currently 300-400 μm in diameter[123,90], which is much larger than 1-2 μm achieved in SIMS[166] measurements and of 0.1-0.3 μm in PIXE[163,164].

Freeman et al.[168] at Oxford University are developing a negative ion source for AMS, incorporating a scanning micro-focused (0.2 μm) Ga^+ ion beam currently used in SIMS. The Ga^+ ion beam is associated with a crossed-beam of Cs vapor for enhancing the negative ion production at the target spot. For example, the secondary $^{14}\text{C}^-$ ions labels can be sputtered from biological tissue at a submicron area to obtain a subcellular resolution. This design is characterized by a creative combination of high sensitivity of AMS and micron resolution of SIMS.

Gove et al.[123] have reported a depth resolution of ca. 10 nm in an AMS profiling study with ^{36}Cl in silicon wafers. Presumably, this technique should be extended to other materials.

Table IV. Comparison of AMS with five analytical techniques for inorganic trace analysis¹⁾

| | AMS | AAS[160] | NAA[144] | PIXE[162] | ICP-MS[149] | SIMS[150,165] |
|---------------------------------------|---|--|---|---|--|--------------------------------|
| Elements analyzed | not many yet, to be extended | wide spectrum (except H, C, N, O, F, Cl, Br, inert gases and most radio-elements) | wide spectrum (except H, He, B, C, N, O, Ne, and Pb) by (n, γ) reaction | light and heavy elements are less sensitive | wide spectrum (except H, O, inert gases) | wide spectrum (all elements) |
| Multi-element analysis | yes[159] | A Hitachi new product Z-9000 AAS enables simultaneous determination of 4 elements. | yes | yes | yes | yes |
| In situ surface micro-analysis | yes[90] (not well focused, ~400 μm) | no | no | yes[163,164] (0.1-0.3 μm) | no | yes[166] (1-2 μm) |
| Depth profiling | yes[123] (preliminarily tested) | no | yes*[161] | yes | no | yes[167] (resolution 5-100 nm) |
| Isotopic ratio measurement | yes | no | seldom used | no | yes | yes |
| Destructive of sample | yes | yes | no | no | yes | yes |
| Massive equipment needed | yes | no | no(except reactor or accelerator) | yes | no | no |
| Expense | large | little | little (except reactor or accelerator operation) | medium | little | little |
| Operator | large crews | one | one or two (except irradiation service) | small crews | one or two | one or two |

* Suitable for He, Li, B, N, O, S, Cl, K by detecting α , p particles or recoil nuclei.

1) See Table III for an explanation of acronyms.

IV. CONCLUSION

After about 15 years of continuous development, AMS has become an important tool in the measurement of isotopic abundances of a number of cosmogenic long-lived radioisotopes, e.g., ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{41}Ca and ^{129}I , and stable isotopes of elements, e.g., B, P, Cr, As, Nb, Rh, Ag, Sb, Te, Os, Ir, Pt, Au, etc.. At present, more than 30 AMS facilities, mainly composed of a tandem accelerator, have been in operation or are being built in many countries. The AMS technique has increasingly gained popularity among the scientific community.

Since the AMS measurement is based on single atom counting, it has much higher sensitivity than both conventional MS and low level β -counting of long-lived radioisotopes. A very small isotopic abundance of a long-lived radioisotope can be measured with high discrimination of the accelerator system against interferences from other species, such as molecules, isotopes, and isobars. The sensitivity limit reaches 10^{-15} for $^{14}\text{C}/^{12}\text{C}$, $^{10}\text{Be}/^{9}\text{Be}$, $^{26}\text{Al}/^{27}\text{Al}$ and $^{36}\text{Cl}/\text{Cl}_{\text{tot}}$.

In comparison with conventional beta-counting of long-lived radioisotope AMS greatly reduces the size of samples for measurement. Usually a few milligrams of samples are used and in some instances only ten micrograms or even less are needed. Also, AMS makes the measurement time shorter. Particularly when a series of samples are measured, it offers shorter turnaround times in operation.

On the other hand, the difficulty of the AMS technique are the small choice of elements that can be practically used to study so far, its unsatisfactory overall efficiency and precision, and background troubles from contamination and other sources. Moreover, the operation of AMS technique needs large installations and large crews. Hence, it is relatively expensive than other analytical techniques.

To date, AMS has widely been employed to geoscience problems with remarkable success. Most of the problems were studied in relation to historical changes and solved with the dating method. We believe that this kind of studies will continue to play a dominant role in AMS applications. However, one should pay attention to the recent rapid rise of AMS applications on non-dating problems.

In the foregoing review, we have illustrated the current and potential applications of AMS in analysis of trace isotopes and trace elements in various studies. The achievements of these studies are quite encouraging. We may take the following examples as illustrations:

AMS with ^{36}Cl and ^{129}I has played a unique role in siting nuclear waste repositories by assessment of nuclide migration through groundwater from the repository site. The study of the exchange of bomb ^{14}C labeled CO_2 between the atmosphere and the ocean or the biosphere by AMS has given very important information about the transfer of CO_2 into these carbon reservoirs. This transfer is an inhibitory factor against the green-house effect caused by the excess of anthropogenic CO_2 in the atmosphere. For the development of advanced semiconductor devices, there is a pressing need to analyse accurately the quantitative contents of trace dopants and impurities in semiconductor matrices. Using the AMS techniques, the trace contents of Be, B, N, Cl, Cr, As, Nb and Sb etc. in semiconductor materials have been measured.

So far, only about fifteen papers have been published reporting on AMS applications in the life sciences. Very commendable work done by researchers at the Lawrence Livermore National Laboratory in 1990 shows the great promise of AMS to verify the identity of extremely small amounts of biological molecules. For instance, these researchers successfully detected one carcinogen adduct molecule (MeIQX) bound in 10^{11} to 10^{12} DNA bases.

We may confidently predict that AMS applications for non-dating problems, particularly in environmental, material, and biomedical sciences, will immensely increase in the near future. These studies will certainly benefit from the advantages of high sensitivity, shorter measurement time, and small sample sizes of the AMS technique.

The comparison of AMS with several sophisticated analytical techniques demonstrates that AMS is a technique of ultra-sensitive prominence, particularly, for analysis of long-lived radionuclides including ^{10}Be , ^{14}C , ^{26}Al , ^{32}Si , ^{36}Cl , ^{41}Ca , and ^{129}I based on the isotopic ratio measurement. However, like any sophisticated analytical technique AMS is not perfect under all circumstances.

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