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ULTRA-HIGH SENSITIVITY MASS SPECTROMETRY: APPLICATIONS TO RARE NUCLEAR AND COSMOLOGICAL PROCESSES ARCHIVED IN GEOLOGICAL SAMPLES

ULTRA-HIGH SENSITIVITY MASS SPECTROMETRY: APPLICATIONS TO RARE NUCLEAR AND COSMOLOGICAL PROCESSES ARCHIVED IN GEOLOGICAL SAMPLES *)

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1. INTRODUCTION

Interactions among particle physicists, cosmologists and astrophysicists are creating a new discipline, *astroparticle physics*[1], of great scientific importance. This new discipline is encouraging a variety of experiments, not involving high-energy accelerators, whose main aim is to test the predictions of Grand Unified Theories and neutrino physics .

A theoretical overview of the present situation in particle physics, with a particular emphasis on these new aspects, is given in Ref. 2. A recent review on experimental particle physics without accelerators is given in Ref. 3.

Two principal detection methods are employed in these experiments: Direct detection using characteristic radiation or kinematic reconstruction for identification, and passive accumulation of reaction products followed by chemical separation and identification by highly sensitive analytic techniques. Significant progress in the direct measurement of neutrinos and rare decays is forseen with the development of cryogenic detectors like superheated superconducting granules, bolometers, and tunneling junctions. Ultra-sensitive analytic techniques, developed in the last few years, such as laser-based Resonance Ionization Spectrometry (RIS) and Accelerator-based Mass Spectrometry (AMS) allow new and as yet unforseen opportunities for the detection of rare processes using *radiochemical* and *geochemical* methods. We will give an overview of the use of ultra-sensitive mass spectrometry in detecting the products of very rare processes like proton decay and double-beta decay, and the detection of particles of special interest such as relics from the Big Bang (particles whose inclusion results in isotopes with anomalously high masses or with fractional charges) and superheavy elements. In particular, we will describe and examine the potential of several new possibilities afforded by AMS to search for the products of double-beta decay accumulated in natural samples over geological times.

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2. EXPERIMENTAL CONSIDERATIONS

2.1. Integration of rare physical processes with geologic samples

Although several experiments have been proposed for the direct detection of the decay of the proton, inverse beta decay by solar neutrinos and double-beta decay, the small cross sections and decay constants hamper their measurement by direct counting techniques. An alternative exists, however, if the product nucleus or its presence is somehow distinctive (e.g. if the product nucleus is a rare isotope, or if its accumulation results in an isotopic anomaly). Radiochemical methods can be used to overcome the small cross sections by integrating the process over the lifetime of the daughter isotope (thereby capitalizing on the small decay constant); if the lifetime is long enough (or if the daughter is stable), integration can be extended to geologic time scales (the so called geochemical method). The geochemical method is most promising when suitable targets can be identified that are deeply buried (to reduce interference from competing reactions induced by cosmic rays) and chemically relatively pure (ores).

In evaluating possible geochemical experiments, the following parameters have to be considered:

a) Amount of sample necessary to perform the experiment,

b) Difficulty in the chemical separation of the product,

c) Purity of sample and ore location,

d) Difficulty of detection of product isotopes.

The amount of sample needed depends on the cross section (or decay constant in the case of p-decay), on the geologic age of the ore or the lifetime of the product (daughter), and on the abundance of the target (parent). An idea of the relative rarity of these processes is seen in these examples from Ref. 4:

i) proton decay: one decay per 1000 tons of target per year,

ii) solar neutrino detection: one reaction per ton of target per year,

iii) double-beta decay: one decay per 100 g of target per year.

Ultimately, the amount of material needed depends on the detection efficiency of the final analytic technique and the precision of the answer required. For example, if the detection efficiency is 10^{-3} and a precision of 10% is desired, then about 10^5 atoms must be made by the process in the target material. If the target is exposed for only a year, a sample of 10^8 tons would be necessary to measure proton decay, 10^5 tons for solar neutrinos, and 10 tons for double-beta decay. However, if a suitable geologic sample could be identified, the "duration" of the experiment might be extended to a time comparable to the age of the Earth (4.5×10^9 a). This would reduce the sample-size requirement by 10^9 and increase the concentration of the product nucleus proportionately. In the case of proton decay, instead of requiring the identification of 1 particle in 10^{31}, the problem would be reduced to 1 in 10^{22}. Although even this lower figure sounds formidable, it is within the range of the analytic techniques described in the next section.

The major limitations restricting the usefulness of geologic experiments come from blanks and backgrounds. If the resulting nucleus is stable, then at some level it already existed in the target material before the "experiment" began, and the requirement of sufficient detection sensitivity is

replaced by the requirement to measure isotopic ratios with sufficient precision that the isotopic anomaly produced by the process is detectable. Using precision isotope-ratio mass spectrometers, isotopic anomalies of 10^{-5} can be measured. Typically, every element is present in every geologic sample at least at the ppb level. If the nucleus produced by the process is a rare isotope, another factor of 10^4 in sensitivity may be possible. Hence, in a pure ore consisting of the appropriate target, it might be possible (under the best of circumstances) to detect a process with a daughter /parent ratio of 10^{-18} (10^{-9} x 10^{-4} x 10^{-5}). If the integration period is the age of the Earth, then conceivably, the lowest detectable rate for a process that produces a stable (or very long-lived) nuclide is 10^{-18} x 10^{-9} or 10^{-27} a^{-1}. An additional factor of 10^2 to 10^5 in sensitivity is possible when the final analysis is preceded by a stage of isotope enrichment, either as part of the experiment or the result of the sample collection process. Several examples of these will be given in the following.

For radioactive daughters, blanks pose less of a problem because primordial contamination is absent. However, backgrounds from alternative reactions induced by cosmic rays (mainly muons if the target is deeply buried) and natural radioactivity (uranium and thorium) interfere with experiments irrespective of whether the daughter is stable or unstable. Ores at a depth of more than 1500 m with uranium and thorium contents at the ppb level will reduce these backgrounds to an acceptable level for some of the particle-physics problems with geologic solutions discussed in the following sections.

2.2. Analytic capabilities of accelerator mass spectrometry

One of the most promising new analytic techniques is accelerator mass spectrometry. AMS is an analytic technique that uses an ion accelerator and its beam transport system as an ultra-sensitive mass spectrometer [Fig. 1] to provide several stages of mass analysis and, in most cases, element (Z) identification in the final detector. Detailed reviews of AMS and its applications can be found in Refs. 5 and 6. AMS has been successfully employed in the analysis of long-lived cosmogenic radioisotopes present in natural samples with isotopic ratios 10^{-9} - 10^{-15} (Refs. 7, 8 and 9). Usually
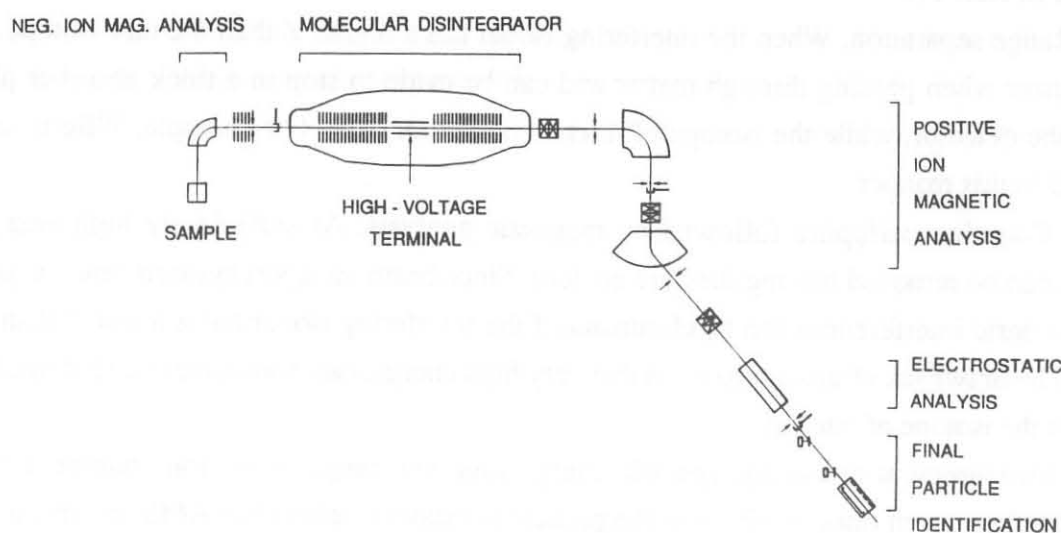

FIGURE 1

Schematic diagram of the XTU tandem accelerator mass spectrometry system (from Ref. 10)

a tandem accelerator, so called because negative ions are first accelerated to the center of a machine whose terminal is at a potential of several million electron volts (MV), converted there to positive-ions by removal of several electrons and subsequently accelerated back to ground potential (two sequential stages of acceleration), is used. The sensitivity of AMS is unaffected by the half-life of the isotope being measured since the atom itself is detected, and not the radiation or particles that result from its decay. And since definitive isotopic identification is normally possible, it is almost unaffected by the backgrounds that limit conventional mass spectrometry (elemental and molecular isobaric interferences, and "tails" of neighboring masses). AMS has higher sensitivity than decay counting for isotopes with lifetimes of > 600 years counted for periods of $<$ a month, and is 5 to 9 orders of magnitude more sensitive than conventional mass spectrometry.

The technique does of course have limitations that affect its selectivity, precision and sensitivity.

2.2.1 Selectivity

The backgrounds that plague conventional mass spectrometers are reduced in several ways. Molecular interferences are eliminated by analyzing ions in the 3^+ or higher charge state: without outer binding electrons, molecular bonds are so weakened that molecular lifetimes are less than 1 microsecond and molecules don't survive long enough to make it to the detector. Elemental isobars are more difficult to eliminate and several different approaches have been adopted:

i) At the ion source. Several elements do not form negative ions (or specific molecular negative ions may be stable for one element but not for neighboring elements) so that the competing isotopic isobar is eliminated at the ion source. ^{14}C dating of organic samples is a well known case, since the ubiquitous ^{14}N does not form negative ions but copious quantities of C^- can be formed . We'll discuss a similar case for the study of $^{70}Zn(\beta\beta)^{70}Ge$, where a rejection of about 10^{12} was obtained at the ion source for the interefering isobar. More general elemental specificity in negative-ion formation can be obtained by combining sputtering with RIS. A laser produces selective multi-step ionization of the atomic species of interest. The use of sputter-initiated -RIS AMS was recently discussed in Ref. 11.

ii) Range separation. When the interfering isobar has a higher Z than the rare isotope, it loses energy faster when passing through matter and can be made to stop in a thick absorber placed in front of the detector, while the isotope of interest passes through. For example, ^{10}Be is separated from ^{10}B in this manner.

iii) Complete stripping followed by magnetic analysis. At sufficiently high energies, all electrons can be removed leaving the bare nucleus. Since beam transport systems select a given m/q ratio -- isobaric interferences can be eliminated if the interfering isobar has a lower Z than the rare isotope. The drawback of this approach is that very high energies are sometimes needed to efficiently fully strip the isotope of interest.

iv) Measurement of energy, specific energy loss, and range in the ion counter (Fig. 2). At sufficiently low count rates ($< 10^4$ s^{-1}) the particle detectors employed in AMS are able to identify the Z of the the incoming ion if it enters with an energy above the Bragg peak. This and the break up of molecules are the aspects of AMS that largely account for its high sensitivity (selectivity).

v) Gas filled magnet. Charge changing collisions of an ion travelling through a gas result in an

average charge state that depends on the Z of the ion. Since the trajectory of an ion in a magnetic field depends on its charge, ions with different average charges (hence different Zs) will follow different trajectories resulting in isotope separation. Application of this technique will be discussed for a solar neutrino detector.

2.2.2 Precision

The precisions of the isotopic ratios measured by AMS are much lower than those obtainable with conventional mass spectrometers. In general, precision depends on counting statistics, isotopic fractionation and blank and standard corrections. A precision of better than 1 % is obtained at dedicated AMS facilities in the measurement of ^{14}C (see Ref. 12). More typically, isotopic ratios are measured with an uncertainty of 3-10%. This limitation comes from the complicated beam optics of AMS systems and/or from the small number of ions counted. We have already discussed the importance of high precision in isotope ratio measurements when looking for anomalies (section 2.1). Because of its lower precision, AMS has a sensitivity of about 10^4 times lower than conventional mass spectrometry in detecting isotopic anomalies -- but this is often compensated for by its higher selectivity. For isobaric processes that convert an isotope of one element into an isotope of another element (β decay, $\beta\beta$ decay ...) lack of sufficient isotopic selectivity may limit the sensitivity of conventional mass spectrometers to an extent that the sensitivity of AMS is actually greater.

2.2.3 Efficiency

The efficiency of an AMS system is the ratio of the number of the atoms (of interest) counted in the final detector to the number loaded in the ion source. This ratio is the product of (i) the negative-ion formation efficiency, (ii) the probability of forming the analyzed charge state, (iii) the efficiency of transporting the beam from the source to the ion counter, and (iv) the detection efficiency. The overall efficiencies of AMS systems range from 1% for ^{14}C detection to 0.02% for ^{129}I detection[9]. This is one of the major limitations in the application of AMS to solar neutrino detectors. AMS efficiency is much lower than that obtainable with other laser- or ion-based analytical techniques or with Neutron Activation Analysis.
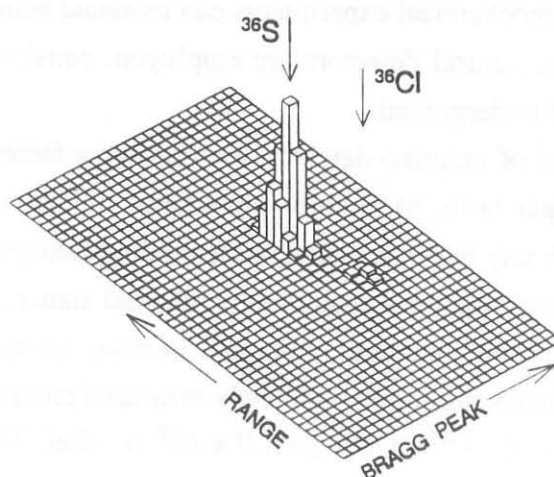

FIGURE 2
Isobaric separation with the Bragg detector (XTU term. volt. 7.75 MV, Q= 7+, $^{36}Cl/Cl = 10^{-11}$)

2.2.4 Background

Backgrounds include all the signals in the final detector that cannot be separated from those corresponding to the nuclides of interest contained in the samples. They include: (i) backgrounds deriving from the limited resolution of the ion detector in the measurement of energy, velocity and specific energy loss, (ii) ion cross contamination with other samples in the ion source, (iii) ion-source background and (iv) contamination before or during sample preparation. Factors (iii) and (iv) are the main limitations in the detection of stable isotopes (see section 8). Special ion sources made with ultra-pure materials have to be designed in order to detect traces of stable elements below the level of ng/g. For most long-lived cosmogenic isotopes, backgrounds corresponding to isotopic ratios below 10^{-15} are possible without any special ion source considerations.

3. SOLAR NEUTRINO EXPERIMENTS

Two reviews of the experimental and theoretical status of the solar neutrino puzzle have recently appeared[13, 14]. The experimentally measured average flux of ν_e from the sun is still ~ 3.8 times lower than the prediction of the standard solar model and this has been variously attributed to problems with the physics of the Sun and problems with the physics of neutrinos (most notably, neutrinos might have mass which would allow them to "oscillate": the missing ν_es would arrive as ν_μ or ν_τ). The so called "non-standard" models are constructed by changing something, physics or parameters, primarily to reduce the central temperature of the Sun to severly reduce the flux of ^{8}B neutrinos. Some of these models suggest that the Sun is not in steady-state. It clearly would be advantageous to have more experimental data, especially on the flux of neutrinos from the p-p reaction (all models give about the same flux for these neutrinos) or on the flux integrated over a few million years (to check for existence of variations in solar output).

3.1 Geochemical experiments

AMS allows the direct detection of rare radioisotopes, present in natural samples with abundances 10^{-15} of the matrix. New solar neutrino detectors have been proposed based on this new technique that do not require the prompt decay of the reaction product as in radiochemical experiments. Furthermore, geochemical experiments can integrate neutrino reactions over geologic times. In these experiments, natural detectors are employed, consisting of appropriate elements contained in minerals buried underground.

In a search for this kind of neutrino detector, the following factors have to be considered: a) equilibrium product-to-target ratio, b) isotopic abundance of the target, c) background due to competing reactions, d) difficulty in the detection of the product isotope.

In the following we summarize the present experimental status of several proposed neutrino detectors using long-lived product isotopes, updating, in many cases, the reaction rates based on recent revisions of the standard solar model and newly measured cross sections.

3.1.1 $^{41}K + \nu \rightarrow e^- + {}^{41}Ca$ $(T_{1/2} \cong 1.0 \times 10^5 \text{ a})$ (Ref. 15)

The effective threshold for this reaction is 2.36 MeV. Therefore, the process is sensitive only to ^{8}B neutrinos. A cross section of $(1.45 \pm 0.05) \times 10^{-42}$ cm^2 has been estimated. Assuming a ^{8}B neutrino flux of 5.8×10^{-6} cm^{-2}s^{-1}, a capture rate for ^{41}K of 8.4 ± 0.3 SNU (1 Solar Neutrino Unit

$= 10^{-36}$ captures/ s . target atom) is obtained.

If the potassium ore is buried at a depth of 1500 m, cosmic-ray muons will contribute only 0.77 SNU to the ^{41}Ca. The contribution of ^{40}Ca(n,γ) and (α,p) followed by ^{41}K(p,n) will be below 10% if uranium and thorium contents of the potassium ore are less than 2.2 and 3.5 ppb, respectively.
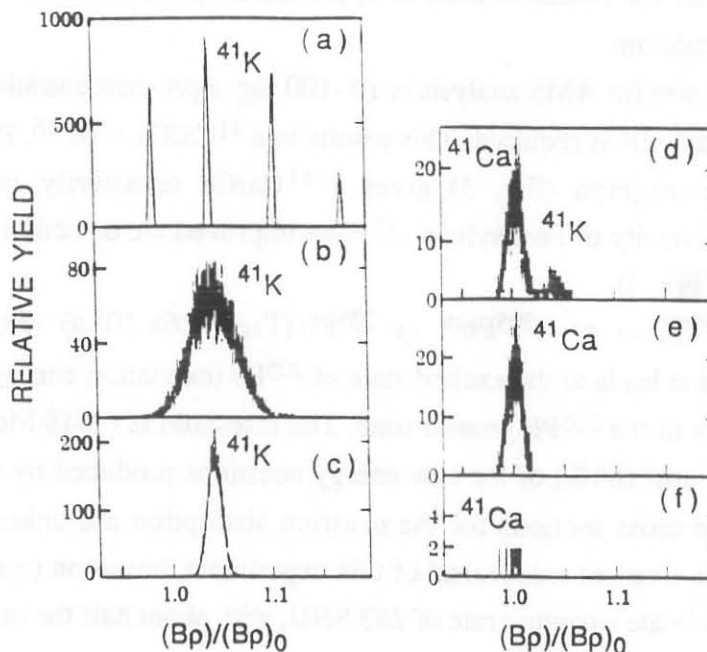

FIGURE 3

Spectra from a magnetic spectrograph focal-plane detector. Figures (a)-(c) show the collapse of the magnetically dispersed charge-state spectrum of ^{41}K into a single line as nitrogen gas is introduced into the magnetic field region. Spectra from standard (d) and (e) and natural (f) samples illustrate the identification of ^{41}Ca at a concentration level corresponding to ^{41}Ca/Ca = 10^{-12} (from Ref. 16). ^{41}Ca^{10+} ions accelerated to 200 MeV
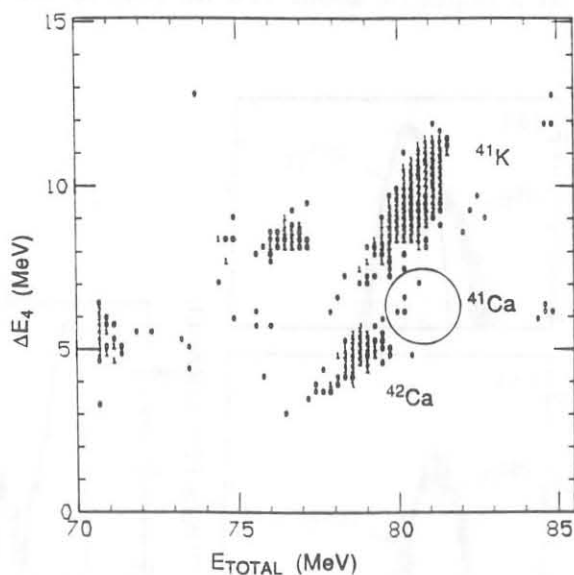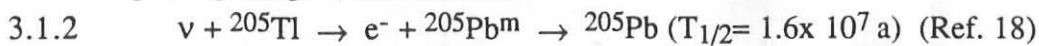

FIGURE 4

E-ΔE spectrum from a commercial CaH$_2$ (blank) in which 4 counts were detected in 90 minutes, corresponding to ^{41}Ca/^{40}Ca=6 x 10^{-16}. ^{41}Ca^{9+} ions accelerated to 84.42 MeV

There are many deposits of potassium satisfying the above criteria. The KCl deposit at Regina, Sasketchewan, Canada, was suggested in Ref. 15. In order to measure the neutrino flux with 10% accuracy we need to observe 100 ^{41}Ca events. With an overall efficiency of 1%, 10^4 atoms are needed for this analysis and they can be extracted from 0.5 tons of KCl (the natural abundance of ^{41}K is 6.77%). Since the Ca contamination is approximately 0.1%, the above extraction will produce 1 kg of natural calcium.

Because the sample size for AMS analysis is 10 -100 mg, a pre-concentration of ^{41}Ca relative to natural calcium by at least 10^5 is required. This results in a ^{41}Ca/Ca $\sim 10^{-16}$. Particle identification using a gas-filled spectrograph (Fig. 3) gives a ^{41}Ca/Ca sensitivity of 6×10^{-14}. Recent measurements at the University of Pennsylvania[17] have improved the detection level to ^{41}Ca/^{40}Ca < 6×10^{-16} using CaH$_3^-$ (Fig. 4).

3.1.2 $\quad \nu + {}^{205}\text{Tl} \rightarrow e^- + {}^{205}\text{Pb}^m \rightarrow {}^{205}\text{Pb}$ ($T_{1/2} = 1.6 \times 10^7$ a) (Ref. 18)

This neutrino reaction leads to the excited state of ^{205}Pb (excitation energy 2.3 KeV), followed by a fast isomeric decay to the ^{205}Pb ground state. The threshold is 0.048 MeV and therefore this detector is sensitive to most (85%) of the low-energy neutrinos produced by the *p-p* fusion in the Sun. Unfortunately, the cross sections for the neutrino absorption are unknown, and can not be calculated accurately. In a recent reappraisal of this experiment, based on (p,n) data of Krofcheck, Bahcall and Ulrich[14] estimate a capture rate of 263 SNU, with about half the rate (173 SNU) coming from neutrinos produced by the *p-p* reaction.

The main background comes from the reaction ^{205}Tl(p,n)^{205}Pb where the protons are produced by high-energy cosmic-ray muons. According to Ref. 18, a depth of 300 m for the ore would keep this background at an acceptable level. Uranium and thorium induced reactions leading to ^{205}Pb should also be kept below the 10% level.

Thallium minerals are very rare. A Lorandite (TlAsS$_2$) deposit of 10 Ma is located in Southern Macedonia, Yugoslavia, at a depth of about 120 m. Due to the erosion of the rocks, this
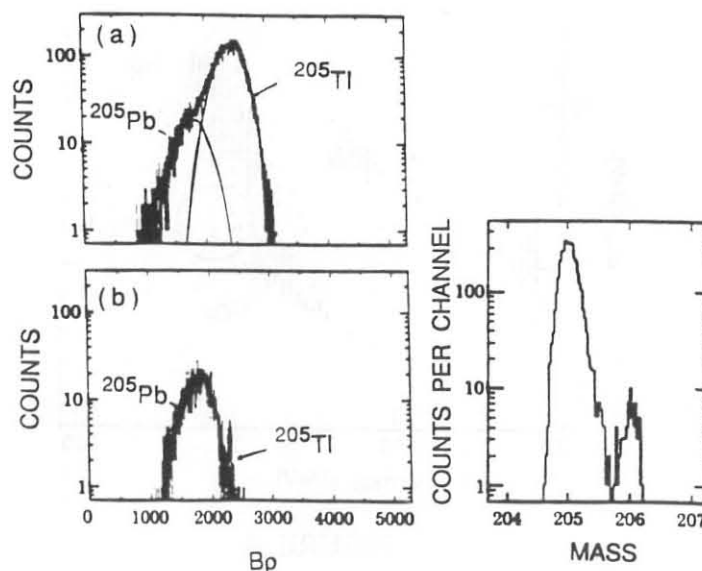

FIGURE 5
Charge (left) and mass (right) spectroscopy with an incident energy of 2.3 GeV (from Ref. 16)

deposit was much deeper during the last million years, providing sufficient shielding against the cosmic-ray muons. Estimates show that backgrounds from cosmic ray and natural radioactivity are acceptable.

The reaction $^{205}Tl(\nu,e^-)^{205}Pb$ gives the most favorable equilibrium product-to-target ratio: $^{205}Pb/^{205}Tl = 3\times10^{-19}$. From the measured Pb contamination in the ore, 3 ppm, a $^{205}Pb/Pb$ concentration of $10^{-13} - 10^{-14}$ is expected in the final sample material. To study the feasibility of this solar neutrino detector, a series of experiments have been performed at the GSI heavy ion facility UNILAC[16]. Pb and Tl ions were accelerated to 2.3 GeV and identified using a time-of-flight system and a magnetic spectrometer (Fig. 5). The present performance of the system is sufficient to suppress neighboring isotopes at the level of 10^{-17}. Isobaric separation between ^{205}Tl and ^{205}Pb is at best 10^{-3} using energy-loss measurements. Hence, chemical separation is required to reduce the Tl/Pb ratio below the level of 10^{-11}. A technique based on isobar separation with a gas-filled magnet will allow the reduction of requirements for the chemical purification. The major problem is the low efficiency of the source, $10^{-6} - 10^{-5}$, giving an overall efficiency of $10^{-9} - 10^{-8}$ for the AMS system. It has been estimated that 10^7 atoms of ^{205}Pb are contained in 100 kg of Lorandite which are obtained from 20 tons of mineral. A recent review on solar neutrino detection with ^{205}Tl is given in Ref. 19.

3.1.3 $\nu + {}^{98}Mo \rightarrow e^- + {}^{98}Tc$ ($T_{1/2} = 4.2 \times 10^6$ a) (Ref. 20)
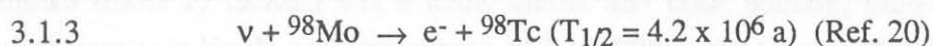
The effective threshold for this reaction is > 1.7 MeV and can be induced only by high-energy ^8B neutrinos. A capture rate of $17.4 \, {}^{+18.5}_{-11}$ SNU is obtained using the standard model ^8B neutrino flux of Ref. 14 and the cross sections in Ref. 21. A mineral containing molybdenum is molybdenite (MoS_2). (α,p) reactions on Mo can produce ^{98}Tc but they are suppressed by high-energy thresholds. ^{98}Tc can be produced with higher rates by reactions $S(\alpha,p)Cl$ and $S(n,p)P$ followed by $^{98}Mo(p,n)^{98}Tc$ and by the (n,p) reaction induced by fission neutrons on ^{98}Ru. Another source of ^{98}Mo background is the spontaneous fission of uranium and reactions induced by cosmic-ray muons. According to Ref. 20, the latter backgrounds can be kept at the 10% level if the mineral is at a depth of 1400 m.

The Henderson ore body, Red Mountain, Colorado, was suggested as a source of Molybdenum for this experiment. 2600 tons of ore would produce 13 tons of molybdenite, yielding 10^7 atoms of ^{98}Tc. Because there are no stable Tc isotopes the ^{98}Tc atoms could be extracted using rhenium as a carrier since it is a chemical homologue of technetium. A sample of about 10^7 atoms (obtained from 20 boxcars of ore) is now essentially ready for mass spectrometric analysis. Results from this experiment can be expected in 1989.

The main problem in the AMS measurement is the separation of ^{98}Tc from the ^{98}Mo remaining after the chemical separation. A feasibility study was performed by the Rochester group[22]. It was shown that ^{98}Mo contamination must be at the 10 ppt level in the Re matrix. This means that the 10^7 atoms of ^{98}Tc should be contained in a mg of Re with no more than 10^8 atoms of ^{98}Mo. Less restrictive chemical purity would be possible using a gas-filled magnet for isobar separation.

An interesting variation of the ^{98}Mo experiment is given in Ref. 23. Haxton and Johnson suggest using the reaction

$$^{98}Mo(\nu_e,e^-)^{97}Tc + n$$
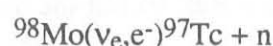
$$(T_{1/2} = 2.6 \times 10^6 \text{ a, E(threshold)} = 8.96 \text{ MeV})$$

to measure the neutrino flux from stellar collapse. This proposal is very attractive. They predict, at a stellar collapse rate of 0.09 a^{-1} with an average of 0.8×10^{53} erg in the electron-neutrino channel, that the average ν_e flux at the Earth due to supernovae will be 3.6×10^3 v/s. Although this is ~ 300 times smaller than the flux of ν_e from the ^8B reaction (using the low-metallicity non-standard solar model adjusted to fit the ^{37}Cl counting rate), the galactic neutrino contribution to ^{97}Tc is predicted to be 40% of the solar contribution. As just described, the ^{98}Mo to ^{98}Tc experiment is already underway. It costs nothing to look for ^{97}Tc at the same time, and integrated over the lifetime of ^{97}Tc this may provide a good estimate of the rate of stellar collapse in the gallaxy.

3.2. A radiochemical experiment (using AMS)

A new application of AMS and radiochemical methods to solve the "solar neutrino puzzle" has been recently suggested[24].

The following reaction is considered:

$$\nu + {}^7Li \rightarrow e^- + {}^7Be \quad (T_{1/2} = 54 \text{ d}).$$
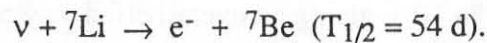
According to the standard solar model, the capture rate in ^7Li corresponds to 51.8 ± 0.31 SNU. 100 tons of lithium would produce 3000 ^7Be atoms. Such a low number of atoms cannot be measured by counting techniques. In fact, 90% of ^7Be decays emitting 50 eV electrons and only 10% decays with 0.477 MeV γ-rays. Direct measurement of the atoms is possible with AMS. The use of 10 µg of ^9Be carrier would produce a $^7Be/^9Be$ of 2.2×10^{-15}. A major problem is the efficiency of the AMS system, as we mentioned in section 2. Fireman et al. suggest a multiple sputtering procedure to concentrate BeO on the source electrode and obtain at least 100 events in the final detector.

4. PROTON DECAY

According to the Grand Unified Theories, the proton is expected to decay with a lifetime of 10^{31} years (Refs. 25, 26 and 27). The geochemical method can be used in this case to search for the disappearance of the proton and is inclusive of all the decay channels.

One proposal is the decay chain

$$^{130}Te \rightarrow {}^{129}I \rightarrow {}^{129}Xe \quad (\text{Ref. 28})$$
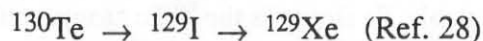
where the proton decays in a tellurium mineral of geological age (10^9 a), producing an isotopic anomaly in the ^{129}Xe abundance. A limit for the proton decay half-life of 1.6×10^{25} a was obtained by Evans and Steinberg, analyzing the data obtained from geochemical $\beta\beta$ experiments[29].

It was later pointed out that there exists the possibility of detecting the ^{129}I, $T_{1/2} = 1.6 \times 10^7$ a, directly, using accelerator mass spectrometry[30]. ^{129}I does not have a stable isobar and the isotopic background can be separated by time-of-flight measurements

Assuming a half-life of 10^{30} a for the proton decay, there are 1325 atoms of ^{129}I per kg of tellurium at equilibrium. The amount of stable iodine per kg of Te must be kept below 2 mg in order that the isotopic ratio of $^{129}I/^{127}I \geq 10^{-16}$. The present AMS background for ^{129}I detection corresponds to $^{129}I/^{127}I = 2 \times 10^{-14}$ (see Ref. 9) but the above isotopic ratio should ultimately be measurable. Overall efficiency for an AMS system is 2.3×10^{-4} and 300 kg of tellurium would be

necessary to obtain 100 counts of ^{129}I in the detector for a precision of 10% in the measurement of the isotopic ratio.

Several background sources in the tellurium mineral have to be considered: spontaneous fission, reactions induced by cosmic-ray muons and neutron capture in ^{128}Te. At depths greater than 4000 m, the most important background is due to the reaction.

$$^{128}Te(n,\gamma)^{129}Te \rightarrow {}^{129}I$$
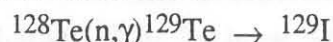
induced by neutrons produced by high-energy muons. It has been estimated that 3000 ^{129}I atom per kg of tellurium would be produced at a depth of 8000 m water equivalent, so that much greater depths would be necessary.

5. SEARCHES FOR STABLE HEAVY HADRONS

Many recent Big Bang cosmologies[31, 32] have predicted the formation of heavy (10 GeV to 100 TeV) stable or nearly stable (lifetimes comparable to the age of the universe) particles during the early moments of the universe at abundances with respect to ordinary hadronic matter of 10^{-10} to 10^{-12}. There are many candidates for this material: the lightest of the technicolor baryons[33], the fermion partners of bosonic particles (predicted by Supersymetric theorists)[34], the so called X$^-$ and X$^+$ particles of Cahn and Glashow that only have electromagnetic interactions with ordinary matter[33], or the lightest six-quark state[35], to name just a few. Regardless of their origin, if they exist and if they are charged, they should be observable in ordinary matter. If they are positively charged, they probably have similar chemical properties to hydrogen and would appear as an anomalously heavy isotope of hydrogen. If they are negatively charged, they would probably exist bound to an otherwise ordinary nucleus (with nuclear charge Z) and would behave as an anomalously heavy isotope of an element with Z-1. Neutral particles may or may not be bound to ordinary nuclei. If they are, then the searches that will be described shortly would detect them; but if they exist as heavy isolated neutral particles, they could have heretofore escaped notice. Recently[36], the possible
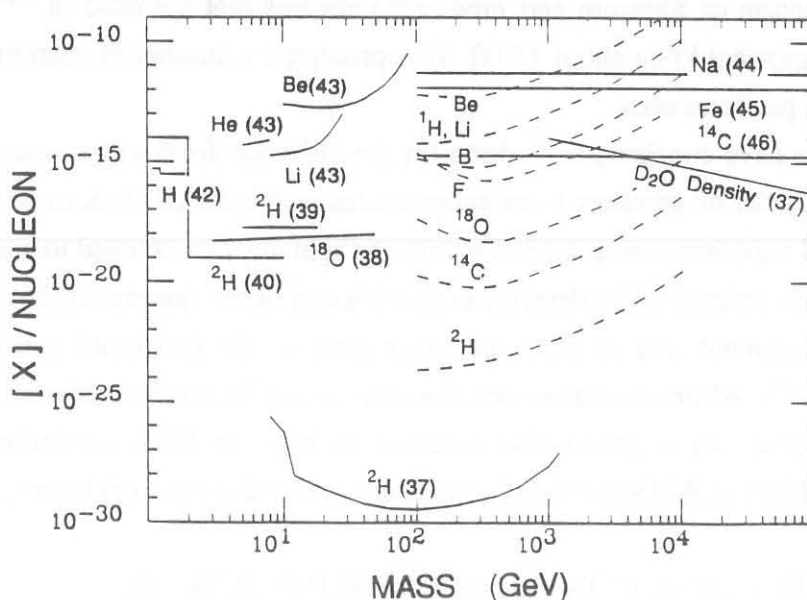
FIGURE 6
Concentration limits (90% confidence level) for the existance of heavy isotopes in matter[41]

existence of yet another class of particles has been proposed that would result in isotopes that are lighter than ordinary ones; so far there have not been searches to look for them. Before the advent of accelerator searches, the limits on anomalous nuclei of arbitrary mass were generally rather poor, with an upper limit for X/nucleon ~ 10^{-6} (where X is a hadron of anomalous mass).

Fig. 6 summarizes the recent searches for anomalous isotopes. More then half of these searches used AMS. The limits below the level of 10^{-15} include a "pre-enrichment" stage such as electrolysis[37] or isotope enrichment[38, 39, 40]. The measurement shown with dashed lines were made using an all electrostatic AMS analysis system[41] which meant that the system was largely mass independent, and anomalous isotopes over the entire mass range indicated could be looked for simultaneously. The other AMS searches required changing all the magnetic elements (bending magnets and magnetic quadrupole lenses) in the injector and high-energy analysis systems in overlapping steps to continuously scan the indicated mass regions. If one assumes that geologic fractionation has not dramatically reduced the concentration of heavy isotopes in ordinary materials, these searches appear to rule out the existence of charged (and possibly even neutral particles) with masses of 1 GeV to 10^5 GeV.

6   SEARCHES FOR SUPERHEAVY ELEMENTS

There is a certain fascination connected with the notion that there is an island of nuclear stability lying just beyond the known peninsula of nuclei forming our world. Several accelerator experiments have attempted to create these nuclei, but they have not been successful, perhaps because the island does not exist, but maybe because of the difficulties of creating such elements by the collision of two heavy nuclei. An alternative to the accelerator experiments is to search for superheavies in nature.

A few years ago, Nilsson, Thompson, and Tsang[47] predicted that the superheavy element with Z = 110 and A = 294 (^{294}110) might have a lifetime on the order of 10^8 years. Although there is some uncertainty whether superheavy elements can be produced by the r process during stellar collapse, the prediction of Shramm and Fowler[48] indicates that the ratio of ^{294}110/Pt (Pt is its chemical homologue) should be about 1/100. This prompted a number of searches for superheavy elements in various platinum ores.

These searches have consisted of looking for the radiation from a characteristic decay (alpha emission, fission tracks or gammas from spontaneous and induced fission) and one accelerator search[49]. The AMS experiment consisted of a time-of-flight detector coupled to a gas cell to measure energy and a silicon surface barrier detector to provide one of the fast signals for timing. The result of the AMS measurement was to place an upper limit on the fractional number abundance of ^{294}110/Pt < 1 x 10^{-11}. While consistent with the other searches, it is inconsistent with a lifetime on the order of 10^8 years or a production fraction as high as 0.01. Nonetheless, this search demonstrated the ability of AMS to detect (or set limits on the detection of) heavy, high-Z nuclei.

7.   SEARCHES FOR FRACTIONALLY CHARGED PARTICLES

AMS offers an attractive method of searching for fractionally charged particles (FCP) because of the variety of materials that can be studied, and, assuming a positive result, the additional

information about the FCP that can be determined, e.g. mass. In general, the energy requirements for these searches is low, most of them have been performed with 3 MV machines or smaller. Tandem accelerators offer an additional advantage of even more positive identification because of analysis as both negatively charged and positively charged ions. Often, very clever schemes can be employed to extract fractionally charged materials from the ion source.

The first AMS search for FCPs was done by Elbert[50] with a single-ended 1 MV accelerator, an RF ion source, an electrostatic and magnetic analysis system and a silicon detector. He was able to set a limit of about 10^{-13} per nitrogen molecule. Subsequent searches by Boyd[51, 52], Schiffer[53], Kutschera[54], Chang[55] and McKeown[56] were inspired by the claims of FCPs detected by the Fairbanks group. The sensitivities (10^{-15} to 10^{-20} per atom over a mass range of about 0.2 to 300 GeV/c^2) of these studies are comparable to the levels at which Fairbanks claims to have seen FCPs.

The Rochester group constructed a special all electrostatic analysis system to search for FCPs and anomalous-mass isotopes[57]. In an experiment on fractionally distilled xenon enriched an assumed 10^6 times, they set a limit of 10^{-18} on the existence of FCPs.

Despite the fact that no AMS search has detected a single FCP, searches will undoubtably continue, especially as new source materials are suggested, e.g. meteorites, or as new detection schemes are developed. Without pre-enrichment of some sort, however, the limits set by AMS are not likely to be much better than 10^{-16} to 10^{-18}, because of the ion source limitations.

8. NEW OPPORTUNITIES TO DETECT $\beta\beta$

The importance of Double Beta Decay (DBD) experiments as tests of lepton number conservation, mass and charge conjugation properties of the electron neutrino, and right-handed admixtures in the weak leptonic currents[58] is well known.

There are more than 30 DBD reactions that can be looked for among naturally occurring parent isotopes (Table 1). Direct detection is being attempted using solid state detectors, e.g. ^{100}Mo($\beta\beta$)^{100}Ru (Ref.59) and time projection chambers, e.g. ^{82}Se($\beta\beta$)^{82}Kr (Ref. 60) but the best detection efficiency is obtained when the detector is made from the parent material, e.g. ^{76}Ge($\beta\beta$)^{76}Se (Ref. 61) and ^{136}Xe($\beta\beta$)^{136}Ba (Ref. 62) .The latter method can be extended to ^{100}Mo $\beta\beta$-decay and other cases with cryogenic detectors[63].

An alternative to direct detection is the geochemical method, where the signal is an isotopic anomaly in the daughter isotope present in an ancient , natural occurring sample of geological age. Maximization, and indeed detection, of the anomaly require a sample with the parent as the major element and the daughter as an ultratrace element. At best DBD will yield about 10^9 daughter atoms/g. As an illustration of the magnitude this limitation represents, consider a daughter with an isotopic abundance of 15% and an atomic weight of 50. if the the endogenous or primordial concentration of the daughter element is 50 ppt in the geological sample, the corresponding isotopic anomaly will be 1%. Such extraordinarily low concentrations lie well below the detection limits of conventional chemical analytical methods thus restricting the preliminary selection of promising systems to inferences based on well-established geochemical trends.

Considering the possible DBD cases of Table I, four of them produce daughter isotopes that

TABLE 1 - $\beta^-\beta^-$ decay transitions from naturally occurring parent isotopes[64].

Transition			T_0 (KeV)	Abundance (%)
46Ca	$\rightarrow$	46Ti	985	0.0035
48Ca	$\rightarrow$	48Ti	4272	0.187
70Zn	$\rightarrow$	70Ge	1001	0.62
76Ge	$\rightarrow$	76Se	2045	7.8
80Se	$\rightarrow$	80Kr	136	49.8
82Se	$\rightarrow$	82Kr	3005	9.2
86Kr	$\rightarrow$	86Sr	1249	17.3
94Zr	$\rightarrow$	94Mo	1148	17.4
96Zr	$\rightarrow$	96Mo	3350	2.8
98Mr	$\rightarrow$	98Ru	111	24.1
100Mo	$\rightarrow$	100Ru	3033	2.8
104Ru	$\rightarrow$	104Pd	1301	18.7
110Pd	$\rightarrow$	110Cd	2014	11.8
114Cd	$\rightarrow$	114Sn	540	28.7
116Cd	$\rightarrow$	116Sn	2808	7.5
122Sn	$\rightarrow$	122Te	358	4.56
124Sn	$\rightarrow$	124Te	2278	5.64
128Te	$\rightarrow$	128Xe	869	31.7
130Te	$\rightarrow$	130Xe	2533	34.5
134Xe	$\rightarrow$	134Ba	843	10.4
136Xe	$\rightarrow$	136Ba	2481	8.9
142Ce	$\rightarrow$	142Nd	1414	11.1
146Nd	$\rightarrow$	146Sm	61	17.2
148Nd	$\rightarrow$	148Sm	1928	5.7
150Nd	$\rightarrow$	150Sm	3367	5.6
154Sm	$\rightarrow$	154Gd	1250	22.6
160Gd	$\rightarrow$	160Dy	1731	21.8
170Er	$\rightarrow$	170Yb	655	14.9
176Yb	$\rightarrow$	176Hf	1077	12.6
186W	$\rightarrow$	186Os	489	28.6
192Os	$\rightarrow$	192Pt	408	41.0
198Pt	$\rightarrow$	198Hg	1043	7.2
204Hg	$\rightarrow$	204Pb	414	6.9
232Th	$\rightarrow$	232U	850	100.0
238U	$\rightarrow$	238Pu	1146	99.275

are noble gases; three of these cases (^{82}Kr, ^{128}Xe and ^{130}Xe) have already been studied with geochemical methods by conventional mass spectrometry[65]. Among the remaining cases, 3 have as parents either Kr or Xe which do not form solids on Earth. Six other parent elements, Ce(Nd), Nd(Sm), Sm(Gd), Gd(Dy), Ru(Pd) so resemble their daughters chemically that finding an ore of the parent not containing a significant amount of the daughter is unlikely. We are left then with the following elemental pairs ; Ca(Ti), Zn(Ge), Ge(Se), Zr(Mo), Mo(Ru), Pd(Cd), Cd(Sn), Sn(Te) and U(Pu). With the exception of U(Pu), most of these elemental pairs share one discouraging feature, namely a tendency to concentrate in sulphur-bearing phases.

Of the three most promising cases (see also Ref 66), Ca(Ti), Zn(Ge) and Zr(Mo), in light of the AMS requirements, the decay ^{70}Zn($\beta\beta$)^{70}Ge is most attractive.

8.1 The ^{70}Zn($\beta\beta$)^{70}Ge case

The feasibility of this measurement must be evaluated with respect to the following parameters: backgrounds from the accelerator and the sample, AMS selectivity and precision in measuring the isotopic ratios and the overall efficiency of the AMS system.

The following formula relates germanium contamination (F, fraction by weight) to the anomaly (ΔR) in the natural isotopic ratio (R) induced by the DBD of ^{70}Ge (decay constant λ) in a mineral of geological age T:

$$\Delta R/R = 0.0302 \times \lambda \times T / F$$

As an example, if the germanium background is 10^{-12} (by weight), with a DBD half-life of 10^{21} a, an isotopic anomaly larger than 4% is produced in a sample with a geological age of 1.8×10^9 a. A background of 10^{-9} will produce the same effect with 10^{18} a half life.

8.1.1. Ion Source background

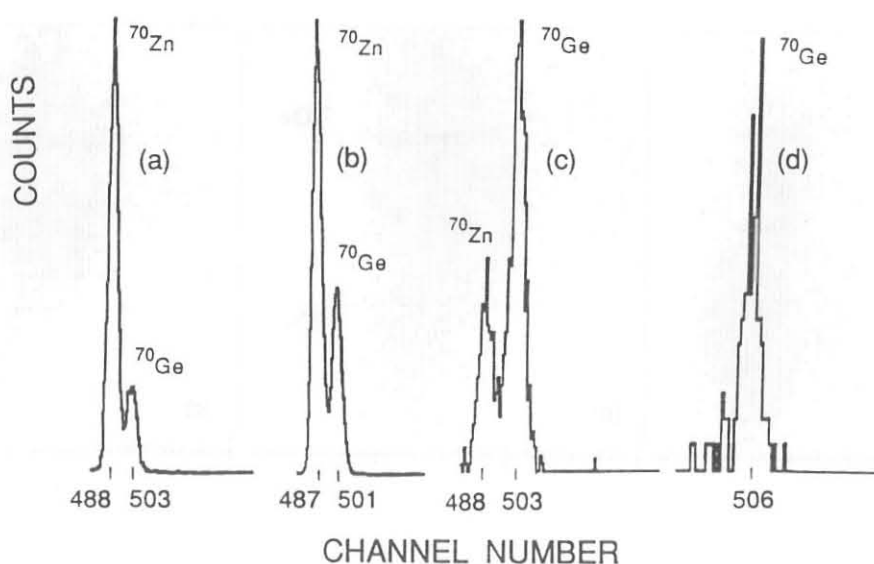Our measurements at the XTU Tandem of the Laboratori Nazionali di Legnaro show that

FIGURE 7

Bragg peack spectra, (a), (b) and (c), from a zinc sample and with the slits after the injection magnet progressively closed, (d) from a cathode of pure tantalum. XTU terminal vltage 8.3 MV, charge state 9$^+$.

with a cathode of very high purity tantalum, a ^{70}Ge counting rate of 5 cps is observed when injecting and transmitting mass 70 through the accelerator at 8.3 MV, equivalent to a germanium contamination of about 1 ppb (Fig. 7d). The Cu$^-$ current was 0.2 µA. The same background was observed at the FN tandem at the University of Pennsylvania, where a ^{70}Ge counting rate of 65 cps was observed with a Cu current of 2.75 µA (Fig. 8-c).

J.M. Anthony and D. J. Donahue suggest that modification of the Cs ion source and sample holder can lower the source background for germanium to the ppt level[67].

8.1.2. Sample background

The principal zinc ores are sphalerite and wurtzite, two forms of ZnS, and a variety of others evidently formed from it under oxidizing conditions: ZnO, ZnCO$_3$, Zn$_2$SiO$_4$, etc. Sphalerite often contains appreciable amounts (100 ppm) of germanium. We could not find Ge analysis for the minor minerals listed above but suspect that some Ge accompanies the Zn, perhaps to form germanates. Some sphalerite, namely that formed at high temperatures, contains "little" germanium. Thus it may be, depending on the detailed conditions, that some of the secondary zinc minerals that originate as low-Ge sphalerite could wind up with a few ppt of germanium. Louis Cabri, from Energy, Mines and Resources (Canada), provided some pure sphalerite from different ores. They were analyzed for germanium contamination by M. Rivers, S. Sutton, A. Hansen and K. Jones using synchrotron radiation - based X-ray fluorescence at the Brookhaven National Light Source with the following results: i) Wiarton ore (430x10^6 a), 730 ppm germanium, ii) Gaina ore (900 x 10^6 a), < 50 ppm germanium, iii) Nygroven ore (1850 x 10^6 a), < 50 ppm germanium. Ge was detected using the K$_\beta$ peak at 11 KeV because of the Zn interference at the K$_\alpha$ energy. Neutron Activation Analysis of samples ii) and iii) will be performed before attempting AMS measurements.

8.1.3. AMS selectivity

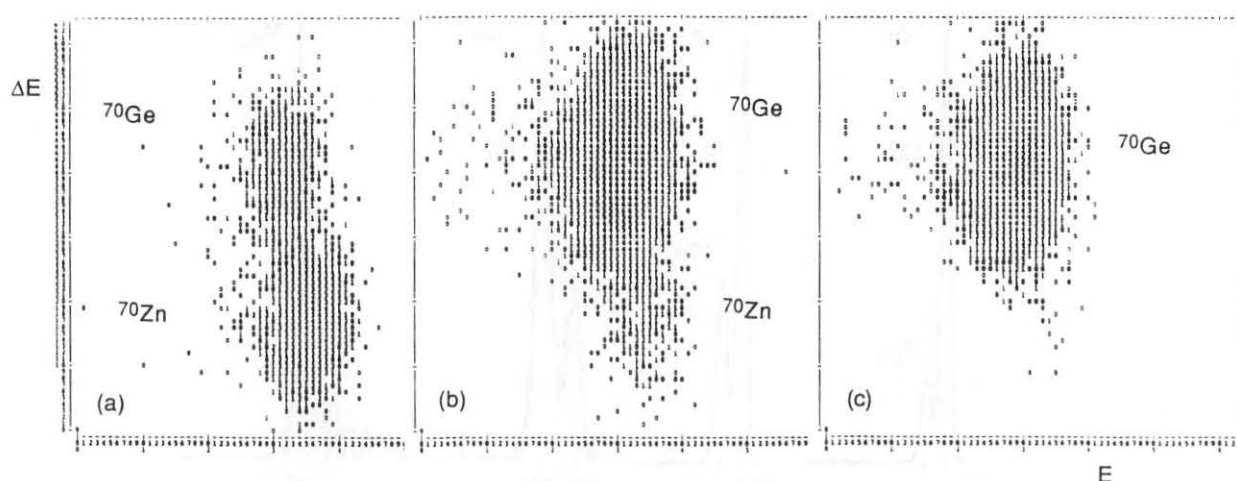The ^{70}Ge/^{70}Zn ratio in sphalerite samples will be 10^{-12} if DBD half-life is 10^{21} years.

FIGURE 8

E-ΔE spectrum from a cathode of commercial zinc (a) with source slits open, (b) with source slits closed to 3.55 mm, and (c) from a cathode of pure tantalum obtained with the FN tandem at the University of Pennsylvania at an incident energy of 80 MeV (for a description of this AMS system, see Ref. 68)

Measurements at the XTU tandem of Legnaro and at the FN tandem of the University of Pennsylvania show that the ratio of the Zn^- ionization probability to Cu^- ionization probability is 10^{-11} - 10^{-12}. In fact, a ^{70}Zn counting rate of few cps was observed in both cases with a cathode containing commercial zinc (Figs. 7-a-b-c and 8-a-b). There is a strong evidence that this intereference does not come from the injection of $^{70}Zn^-$ but from the $^{70}ZnH^-$ tail. The alignment of the beam optics was obtained by transmitting $^{65}Cu^{9+}$ at the appropriate terminal voltage.

In conclusion, the sputter source and the final particle identification system are the key elements of the AMS system which make it possible to obtain the required separation between ^{70}Ge and ^{70}Zn during the ^{70}Ge analysis of sphalerite minerals. AMS selectivity is sufficient but the background (mainly from the original sample) appears to be the major factor limiting sensitivity.

The remaining cases will be briefly discussed in the following.

8.2. The $^{46,48}Ca(\beta\beta)^{46,48}Ti$ case.

Extraterrestrial materials offer the advantage of old $(4.5 \times 10^9$ a) ages but the disadvantage of significant cosmic ray bombardment. The curve of cosmic abundances is such that of all the pairs in the list of Table 1, only the Ca and Zr form independent minerals in meteorites. In practice, only Ca compounds are abundant enough in meteorites to be worth further consideration. Most Ca-bearing minerals in meteorites contain significant quantities of Ti. The phosphates whitlockite and apatite may not. Unfortunately these minerals are enriched in U and Th and the decay products of extinct ^{244}Pu. To our knowledge, no Ti analyses are available for the phosphate minerals in meteorites.

Terrestrial materials are customarily classified as igneous, sedimentary or metamorphic. The lowest Ti content observed in feldspars is about 10 ppm. It is improbable that a sedimentary, Ca-bearing mineral would exclude Ti. It might be that some minor mineral such as fluorite (CaF_2) excludes Ti because of a mismatch in ionic radii.

8.3. The $^{92,94}Zr(\beta\beta)^{92,94}Mo$ case.

This case seems promising from the geochemical point of view. Unlike Ca and Ti, the two elements have decidedly different chemical affinities. Zr is strongly lithophile while Mo may be siderophile or chalcophile. In addition, Zr forms a well-defined igneous mineral, zircon, which in a recent report of trace element analyses of zircon, gave no results for Mo.

9. CONCLUSION

The use of AMS to measure the products of rare nuclear and cosmological processes accumulated or archived for geologic times in natural samples would appear to offer a fantastic potential for determining very low rates and very small cross sections. To some extent, this potential has been realized. The limits set by AMS on the existence of relic particles leftover from the Big Bang are comparable to, or far exceed the limits that can be set by any other analytic technique. The limits on rare isotopes of many types: anomalously heavy, fractionally charged, or of superheavy elements, have in general been good enough to challenge the predictions made by theory. These successes have occurred when the full sensitivity of AMS could be exploited: when the isotope being sought was in itself distinctive.

On the other hand, major difficulties arise when attempting to use AMS to exploit the

advantages of natural samples in accumulating products of rare processes over geologic time. The advantage of natural samples, compared with the detectors used in direct counting experiments whose duration is a year or so, are their comparatively small size and the 10^6 to 10^9 times larger concentrations of product. But because the nuclei that result from double-beta decay, inverse beta decay or proton decay are "ordinary" isotopes, high sensitivity is only possible when the resulting element has been geochemically excluded from the target material. This rarely occurs to a level of better than a part per billion and finding samples where levels are this low is difficult. In addition, backgrounds may occur from handling the sample, or from contamination in the ion source. Finally, as selective as AMS machines are, they do not have the precision of isotope-ratio machines, restricting the size of measurable isotopic anomalies to being larger than they might otherwise have to be.

Improvements in the efficiency, background and precision of AMS are constantly being made. Increased sensitivity is possible because of the development of high-intensity ion sources and techniques of universal usefulness in removing isobaric interferences. But in overcoming the geological and geochemical restrictions we must depend on the kindness of Nature (and on the collaborative attitude of geologists). The ultra-pure separations from enormous amounts of mineral material required in some of the geological experiments needs the contributions of large and well organized groups. This interdisciplinary effort is justified only as long as we think these methods offer the possibility of testing some of the fundamental predictions made by the new physical theories.

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