ACCELERATOR MASS SPECTROMETRY IN NUCLEAR PHYSICS

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Abstract

The use of AMS to measure nuclear quantities is reviewed. Particular emphasis is put on the discussion of half-life measurements and searches for hypothetical particles.

1. Introduction

Accelerator Mass Spectrometry (AMS) is a method of counting atoms at low concentrations. The accelerator is an essential part of the method because it can remove interfering molecules and provides sufficient energy to make an unambiguous mass and nuclear charge identification possible. Since backgrounds can be reduced to zero in favorable cases, the ultimate sensitivity of detecting a small number of atoms from a sample is determined by the overall efficiency of ionizing, transporting, and counting the ions in a detector. For most AMS measurements, concentrations of a rare species (e.g. a long-lived radioisotope) in bulk material are measured through isotopic ratios of the rare to an abundant (stable) isotope of the same element. The stable isotope can be part of the original material or is added as carrier material. The lower limit of isotopic ratio measurements is chiefly determined by the ion source output and the transmission through the accelerator and can be as low as 10^{-16} . Depending on the concentration of the abundant isotope in the bulk material and/or on the amount of carrier added, the concentration of the rare species can be considerably lower in the bulk material.

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AMS is most widely used for the measurement of the concentration of long-lived radioisotopes [1, 2]. Since for half-lives much longer than a typical counting time many more atoms survive than decay, atom counting becomes more efficient than decay counting. AMS is being applied to a large variety of fields [2] both utilizing natural and artificial radioisotopes. The present paper will only discuss AMS application to nuclear physics, the field it has originally emerged from. This subject has recently been reviewed [3] and the reader is referred to it for additional information. Here, we will concentrate on a few selected topics.

2. Measurements of long half-lives

The half-life is the most important quantity if one wants to use a radioisotope for chronological studies. Half-lives of interest to AMS are in general too long to be measured through a change in decay rate within a reasonable counting period. Instead, their half-life can be measured in two other ways: i) the decay constant ($\lambda = (\ln 2)/t_{1/2}$) can be determined from the specific activity $(dN/dt)/N = -\lambda$, where dN/dt is the decay rate and N the number of atoms of the radioisotope; or ii) the half-life can be determined from the activity or radioisotope concentration in natural samples whose age is well known. Whereas in the first method all parameters can be studied in the laboratory, the second requires the understanding of the conditions in a natural environment over geological time scales. Although the first method seems to be preferable, it involves absolute measurements of decay rates and radioisotope abundances, which are difficult to accomplish with high accuracy. If the radioisotope abundance is too low for a measurement with low-energy mass spectrometry, AMS can be used. On the other hand, AMS is also useful for the second method if the radioisotope concentration is too low to be measured via decay counting. Examples for both applications of AMS are described below.

The primary challenge in AMS is the separation of the radioisotope from interferences of a stable isobar. Figure 1 shows how this problem was solved for four different radioisotopes of interest for half-life measurements. High energy is particularly helpful for isobar separation of the heavier isotopes. Therefore the superconducting heavy-ion linac ATLAS was used for the ^{60}Fe and ^{126}Sn measurements.

2.1. ^{32}Si

The half-life of ^{32}Si is a notoriously difficult case, even though it is in a range where a decay measurement can be performed. Figure 2 summarizes the present situation. Three geophysical measurements have been performed involving different natural reservoirs. Clausen [9] measured the depth distribution of ^{32}Si in ice cores from Greenland. DeMaster [10] did a similar measurement in a sediment core from the Gulf of California. Somalyajulu et al. [11] determined the half-life by a comparison of the global ^{32}Si inventory of the oceans and the average cosmic-ray production rate. Whereas the first two results depend on somewhat uncertain factors (e.g. constant ^{32}Si deposition rate as function of time, chronology of the investigated layers) the last method compares two well established geophysical quantities. The latter result compares fairly well with the three AMS measurements within the experimental uncertainties. The early AMS measurements were performed at Argonne [4] and Rochester [12], and the recent one at Zurich [13]. The errors are relatively large because, as mentioned above, absolute quantities have to be measured. Disregarding the ice and sediment result because of hard-to-assess uncertainties, the main puzzle remains the discrepancy with the high-precision decay measurement by Alburger et al. [14]. In this measurement, the activity of a ^{32}Si material was measured over a period of four years in approximately 4-weeks intervals relative to a ^{37}Cl ($t_{1/2} = 301,000$ yr) source. In principle, such a decay experiment should give the

most reliable result because only the relative change of activity need to be known. However, four years cover only a small portion of the half-life, leading to uncertainties which are perhaps difficult to assess.

Given this situation, it is encouraging that new measurements are under way. A group at Aarhus [15] performs yet another AMS measurement measuring the isotope and activity ratios of $^{31}\text{Si}/^{32}\text{Si}$ produced simultaneously by the reactions $^{18}\text{O}(^{16}\text{O}, 2\text{pn})^{31}\text{Si}$ and $^{18}\text{O}(^{16}\text{O}, 2\text{p})^{32}\text{Si}$. This experiment has the advantage of requiring only the measurements of relative quantities, except that the short half-life of ^{32}Si (2.6 hr) requires separate bombardments for the isotopic ratio and the activity ratio measurements. An interesting new approach is possible at heavy ion facilities equipped with mass separators which allow one to separate ^{32}Si after production in a fragmentation reaction. Collecting the fragments in a suitable detection system will lead to a known number of ^{32}Si nuclei. The decay constant can then be precisely determined from a beta-decay activity measurement.

2.2. ^{41}Ca

An accurate value for the half-life of ^{41}Ca became of renewed interest after AMS made it possible to measure the concentration of ^{41}Ca in both extraterrestrial and terrestrial materials. Because of the long half-life ($\sim 100,000$ yr) and an unfavorable electron-capture decay with only soft X-rays and Auger electrons emitted, decay counting at natural levels was never achieved. Fink et al. [16] have recently reviewed the status of ^{41}Ca measurements with AMS.

Although a specific activity measurement of ^{41}Ca with a highly enriched material of well-known ^{41}Ca abundance is under way [17], an interesting new method of half-life determination with AMS from the measurement of ^{41}Ca and ^{36}Cl concentrations in antarctic meteorites has recently been reported [16,18].

Assuming saturation of activity during cosmic-ray bombardment in outer space for both radioisotopes, a plot of ^{41}Ca versus ^{36}Cl concentration for meteorites of different terrestrial ages results in a slope determined by the ratio of their half-lives (see Figure 3). With the known half-life of ^{36}Cl one determines a half-life for ^{41}Ca of $t_{1/2} = (1.03 \pm 0.07) \times 10^5$ yr. This seems to confirm the corrected half-life value of Mabuchi et al. [19], which is $(1.14 \pm 0.15) \times 10^5$ yr.

2.3. ^{44}Ti

The half-life of ^{44}Ti is of particular interest in connection with supernova SN1987A, since ^{44}Ti has been predicted to be present in young supernova remnants [23]. Gamma rays from 77-day ^{56}Co decay have already been observed from SN1987A [24], and the 1.157 MeV gamma line from the ^{44}Ti decay may be observable after the activity from the shorter lived radioisotopes has sufficiently decreased.

Figure 4 summarizes results of half-life measurements of ^{44}Ti . The specific activity measurement combined with AMS of Frekers et al. [5] gave a half-life which is in fair agreement with two specific activity measurements combined with low-energy mass spectrometry [20,21]. However, a clear discrepancy exists with the decay measurement of Alburger and Harbottle [22] similar to the situation in ^{32}Si . Since ^{44}Ti has a favorable decay through ^{44}Sc to ^{44}Ca with a gamma ray of 1.157 MeV, the half-life could most probably be measured quite accurately relative to ^{60}Co over a time span of several years. It may also be possible to use the fragmentation technique mentioned above for ^{32}Si .

2.4. ^{60}Fe

The half-life of ^{60}Fe was only very poorly known [25] prior to the AMS measurement with the superconducting linac at Argonne [6]. The AMS value of $t_{1/2} = (1.5 \pm 0.3) \times 10^6$ yr is considerably longer than the previous estimate [25]

of $\sim 3 \times 10^5$ yr. The most interesting application of its half-life would be the observation of traces from extinct primordial ^{60}Fe in meteorites (through ^{60}Ni isotopic anomalies), and a possible observation of gamma rays from its daughter ^{60}Co in interstellar space.

2.5. ^{126}Sn

There are still a few long half-lives that are poorly known. The long-lived fission product ^{126}Sn ($\sim 10^5$ yr) has been investigated at Argonne [7] with ATLAS and the gas-filled spectrograph [8]. Figure 1d shows a spectrum at 400 MeV, where a clear signal of ^{126}Sn in the tin fraction extracted from a spent nuclear fuel rod is observed. Unfortunately, a reliable $^{126}\text{Sn}/\text{Sn}$ ratio measurement was not possible for this material because of interferences of various magnetically degenerate charge states originating from double-stripping after tandem acceleration, where the normalization to stable Sn isotope currents was performed. However, the new ECR source positive ion injector [26] for ATLAS should allow such a ratio measurement. The particular interest of the half-life of ^{126}Sn is its role as an important contributor to the overall radiation dose of nuclear waste in the 100- to 10,000-year timescale [27].

Another poorly known half-life which could be measured by a similar AMS technique is the one of ^{79}Se ($t_{1/2} < 6.5 \times 10^4$ yr).

3. Cross sections

Most of the natural long-lived radioisotopes measured by AMS in extraterrestrial and terrestrial matter are produced by cosmic-ray primaries or secondaries. For a deeper understanding of the coupling between observed radioisotope concentrations and cosmic-ray properties (e.g. composition, temporal variations), the production mechanism has to be known requiring the knowledge of production cross sections over a wide range of energies.

Spallation cross sections for proton-induced reactions on a number of targets have been measured for ^{10}Be [28], ^{26}Al [29,30], and ^{41}Ca [30,31]. Recently, first cross-section measurements for the more difficult fast neutron-induced production of ^{14}C and ^{26}Al from ^{16}O and natSi have been reported [32].

Although a variety of semi-empirical models exist to calculate spallation cross sections, the agreement with experimental data over a large number of projectile-target combinations [33] and/or large energy range [28] is not satisfactory. For reliable estimates of radioisotope production it seems necessary to perform accelerator experiments, at least at selected energies to properly normalize model calculations. These cross section measurements are perhaps not very challenging, but they provide indispensable information if a quantitatively reliable relationship between cosmic-ray properties and cosmic-ray reaction products is to be established.

Other selected cross section measurements have been performed with AMS, such as the thermal-neutron induced reactions $^{10}\text{B}(\text{n},\text{p})^{10}\text{Be}$ [34] and $^{36}\text{Ar}(\text{n},\text{p})^{36}\text{Cl}$ [35]. For additional discussions on cross section measurements with AMS the reader is referred to the review article [3] mentioned above.

4. Search for hypothetical particles

An AMS system is usually tuned to detect particles with known mass (A), nuclear charge (Z), and ionic charge state (Q). However, AMS can cover a much larger parameter space than that occupied by known particles. Moreover, since an AMS system can be tuned to any arbitrary set of values of A , Z and Q , it is ideally suited to search for hypothetical particles. By definition, a particle with a hitherto unknown set of A , Z , and Q should be free of known background particles allowing one to search for particularly low concentrations.

In any quest for unknown species in bulk material, there are three crucial questions to be addressed: i) what are the physical properties? ii) what are the chemical properties? iii) where should one look for? Theoretical speculations

about hitherto unknown species usually give some guidance to the first question. However, there usually remain large uncertainties in trying to find answers to the second and third question. Perhaps the best justification for searches of this kind was given by Cahn and Glashow in their paper on Chemical Signatures for Superheavy Elementary Particles [36] where they said: " It is an important truism that if we do not search for these particles we will not find them." Bearing this in mind, one might just as well perform a search for the unknown without much relation to theoretical prediction, simply using the most sensitive method on hand. In this sense, the AMS search for non-Paulian particles reported at this meeting [37] is a good example, since it was probably more driven by experimental feasibility than by the expectation to see an effect.

4.1. Fractionally charged particles

As mentioned above, AMS can be used to search into an unknown realm of mass and charge. In general there are however two contradicting requirements. While one wants a selective procedure to efficiently remove background which is best done by setting tight constraints on mass and charge, one also would like to have a large acceptance for unknown parameters. AMS can well adjust to these requirements. In searches for fractionally charged particles (FCPs) with unknown mass a purely electrostatic system can be used which accepts particles irrespective of their mass because electric forces depend only on E/Q . In practice, ion beam production, stripping, and pulse-height defects in ionization detectors may introduce some mass dependence. Searches of this kind have been performed at Argonne [38] and other places [39-41]. Depending on the particular conditions, concentration limits as low as 10^{-19} FCP/nucleon have been reported [41]. Of course, all of this was triggered by the unresolved puzzle of the observation of fractionally charges on superconducting Nb spheres in the Stanford levitation experiments [42].

4.2. Superheavy elements

AMS searches for other hypothetical particles have been performed, including superheavy elements and anomalously heavy isotopes of known elements. The search by W. Stephens et al. [43] for naturally occurring superheavy element with $Z = 110$ and $A = 294$ demonstrates well what kind of conclusions one can draw from such measurements. They investigated a nugget of platinum, the chemical homolog of element 110, which was found in a placer (sand deposit) in Alaska. Using a time-of-flight technique in connection with acceleration through an FN tandem to identify mass 294, a limit of 10^{-11} was set for the concentration of $^{294}_{110}$ in Pt (this limit was apparently set by assuming that $^{294}_{110}$ has the same negative-ion formation probability and transmission through the accelerator than Pt, which may not necessarily be the case). Figure 5 summarizes the interpretation of their result. Starting point is a primordial 110/Pt ratio of 0.02 to 0.06 estimated by Fowler and Schramm [44]. With a half-life of 2×10^8 yr and no chemical segregation from Pt, one could have expected a present-day concentration of $\sim 10^{-9}$ (allowing for a decay time of 4.7×10^9 yr). The much lower limit found in the present experiment can be interpreted as evidence that either $^{294}_{110}$ was not produced to the extent predicted [44] or the half-life is shorter than 1.5×10^8 yr or geochemical effects have separated it from Pt.

4.3. Anomalously heavy isotopes

Rather intensive searches have been performed with AMS and other methods to find anomalously heavy isotopes of known elements (references pertinent to this subject can be found in [3]). The anomalous mass may result from binding a hitherto unknown heavy particle to the nucleus. The heavy particle is usually called an X particle and can either be neutral or carry an integral charge of either sign. In general, the binding to the host nucleus is assumed to be provided by strong interaction for a neutral X and purely electromagnetic for

X^- . The X^+ particles remain unbound and may appear as heavy hydrogen isotopes. These particles may have been produced during the Big Bang, or later by cosmic rays. The detection of very heavy isotopes of some known element may also be interpreted as the existence of another variety of exotic particles called "strange matter". They may have evolved from phase transitions of the early universe or may have been formed in astrophysical catastrophes at a later stage. Characteristic features of this species are a low charge and a high mass, the low charge being the result of combining the charges of up(+2/3), down(-1/3), and strange(-1/3) quarks in about equal proportions. It was argued that strange matter may be a stable form for almost any mass ranging from $A \sim 10^2$ (ordinary nuclei) to $A \sim 10^{57}$ (neutron stars). No AMS search has been performed so far which specifically looked for strange matter, but some of the X-particle searches do include certain small fractions of the vast mass range predicted by the various theoretical estimates. An interesting approach to search for strange matter was performed at GSI [45] by Rutherford backscattering of very heavy beams (Pb and U), which only scatter back from constituents much heavier than the beam. The result of this experiment is included in the summary of heavy-particle searches presented in figure 6.

5. Conclusion

This paper presents a selection of topics where AMS is used to measure nuclear quantities. A more complete description can be found in ref. [3]. A particularly intriguing possibility for AMS measurements is the detection of solar neutrinos [3]. This has been investigated to some extent for a geochemical experiment with the $^{205}\text{Tl}(\nu, e^-)^{205}\text{Pb}$ reaction. The AMS detection part does not seem to be insurmountable. The major challenge is the level of understanding of the geochemical and geophysical conditions in the thallium ore deposit, required to reliably track the observed ^{205}Pb concentration to solar

neutrino interactions. A similar condition, perhaps not as stringent, exists for another geochemical experiment with the $^{98}\text{Mo}(\nu, e^-)^{98}\text{Tc}$ reaction, pursued for several years in Los Alamos. Another possibility for an AMS solar neutrino experiment may develop with the advent of ECR sources and noble-gas radioisotopes [7]. The long-lived radioisotope ^{81}Kr ($t_{1/2} = 2.1 \times 10^5$ yr) offers the possibility to consider a Davis-type experiment with the reaction $^{81}\text{Br}(\nu, e^-)^{81}\text{Kr}$. In a year exposure time of the Davis detector filled with CHBr_3 one might expect about 3000 atoms of ^{81}Kr [3]. It will be interesting to see whether positive-ion AMS will reach the required overall efficiency of 10% to count such a small number of heavy atoms.

The role of AMS in searches for hypothetical particles will most likely stay as one of the most interesting applications in nuclear physics. Perhaps a really concentrated effort on a particular AMS facility should be mounted to search for strange matter.

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Figure captions

- Fig. 1. Radioisotope separation in the focal-plane detector of an Enge splitpole magnetic spectrograph using various methods to disperse the isobars a) ^{32}Si - ^{32}S , 80 MeV, 1.5 mg/cm² Al [4]; b) ^{44}Ti - ^{44}Ca , 80 MeV, 50 $\mu\text{g}/\text{cm}^2$ Au on 10 $\mu\text{g}/\text{cm}^2$ C [5]; c) ^{60}Fe - ^{60}Ni , 360 MeV, 2.8 mg/cm² Al [6]. d) ^{126}Sn - ^{126}Te , 400 MeV, 6 torr N₂ (gas-filled spectrograph) [7,8].
- Fig. 2. Summary of half-life measurements for ^{32}Si . The results are labeled by their method and the measured half-life values in years. References to the measurements are given in the text.
- Fig. 3. Plot of the ^{41}Ca concentration of Antarctic meteorites with different terrestrial ages versus the concentration of ^{36}Cl in the same meteorites [16,18]. The half-life of ^{41}Ca is determined from the slope and the known half-life of ^{36}Cl .
- Fig. 4. Summary of half-life measurements for ^{44}Ti . The results are labeled by their method and the measured half-life in years. LEMS stands for low energy mass spectrometry. References to the measurements are given in the text.
- Fig. 5. Concentration of $^{294}\text{110}$ in Pt as a function of half-life [43]. A detailed description of this plot can be found in ref. [43]. The lowest curve labeled "ABUNDANCE" is pertinent to the AMS measurement discussed in the text.
- Fig. 6. Summary of concentration limits for anomalously heavy isotopes (X particles) in matter. Details of this plot are described in ref. [3]. Solid lines are results obtained with AMS measurements, dashed lines are LEMS results, and dotted lines are from various other techniques including neutron activation and laser hyperfine spectroscopy. Labels

with isotopic mass indicate the use of pre-enriched material, assuming the same enrichment for X particles. The heavy dashed lines are the result of a strange matter search in an iron meteorite and a lanthanide sample measured via Rutherford backscattering [45].

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