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ACCELERATOR MASS SPECTROMETRY: FROM NUCLEAR PHYSICS TO DATING*

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INTRODUCTION

One of my former teachers in nuclear physics, H Morinaga, once pointed out to me that essentially all the fundamental discoveries in nuclear physics were done without the use of accelerators. At first, this statement seemed extremely exaggerated, but now I think there is much truth in it. Once we build an instrument to investigate a specific problem we have already realized the existence of the problem and do not need the instrument to discover it. It is almost inevitable that the most interesting discoveries with a new technique will be made in fields for which the technique was not invented. Accelerators, built for nuclear physics, produced a great amount of data on nuclear structure and forces but the most fundamental discoveries were made in elementary particle physics. In any case, for almost 40 years the sole purpose of accelerators was to deliver beams of ever increasing energy and versatility to perform experiments after the accelerator. The analytic properties of accelerators were almost completely ignored even after the very early use of the Lawrence cyclotron at Berkeley as a mass spectrometer to discover ^{3}He in nature (Alvarez and Cornog, 1939 a; b). The enormous analytic power of accelerators is now fully recognized, but the joy of the revival is mixed with some disappointment that the great prospects for studying ^{14}C problems (Muller, 1977; Bennet et al, 1977; Nelson, Korteling, and Stott, 1977) have not yet been fulfilled. Fortunately, some of the papers of this conference show that a big step forward has been taken. But in view of what I said before it is not surprising that most of the studies were actually done with other radioisotopes.

Part of the reason for this situation is that Accelerator Mass Spectrometry (AMS) is independent of half-life and decay characteristics which allows for the selection of almost any radioisotope. Since the precision of conventional ^{14}C measurements cannot easily be achieved with such a highly

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complex instrument as an entire accelerator system, it is no wonder that interest first focused on other radioisotopes. Figure 1 gives an overview of the ca 140 radioisotopes with half-lives greater than one year. For shorter half-lives, decay counting will almost always be more practicable. It is interesting to note that ^{14}C is the only convenient radioisotope in a half-life range of almost two decades (5 x 10^2 to 5 x 10^4 yr). A similar outstanding position is occupied by ^{40}K. Only a very small fraction of the radioisotopes shown in the figure has been touched by AMS. A fairly complete account of ongoing and future AMS projects can be found in Henning et al (1981a). It seems quite possible that the whole range of radioisotopes (fig 1) will eventually be accessible to AMS. This will largely be determined by the energy of heavy ion beams available at the future generation of accelerators.

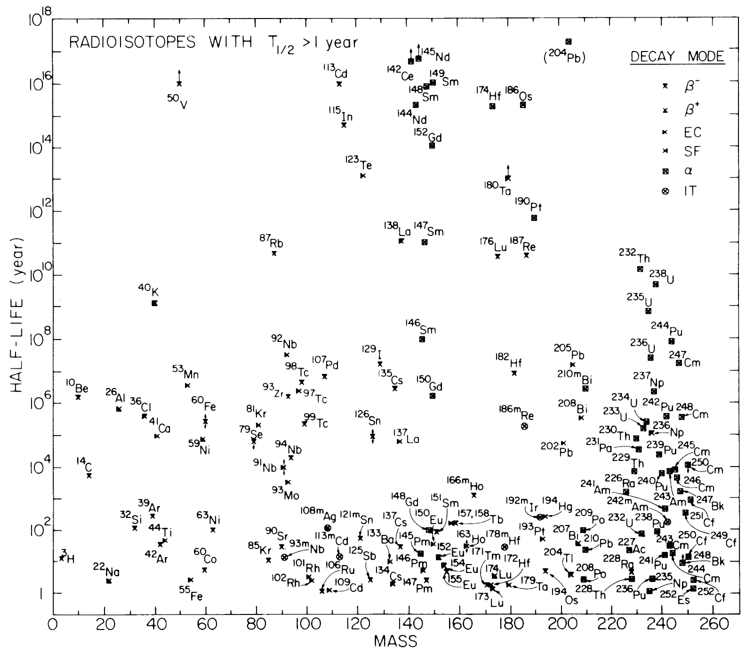

Fig 1. Overview of long-lived radioisotopes. Data are taken from Lederer and Shirley (1978) except ^{32}Si (Kutschera et al, 1980; Elmore et al, 1980) and ^{202}Pb (Nagai, Nitoh, and Honda, 1981). Arrows on data points indicate that the half-life is only approximately known in the respective direction. Isotopes in brackets are of uncertain assignment.

SOME TECHNICAL ASPECTS OF AMS

Figure 2 shows the key for high-energy mass spectrometry, namely, the energy-loss separation of isobaric ions in matter. Since for every radioisotope below Bi there is a stable isobar in nature (except for mass 5 and 8), the main problem in AMS is to separate these usually interfering ions from the radioisotope ions. For an efficient separation, ion energies above the maximum in the energy loss curves are required. In this region, most of the electrons (> 70%) are removed from the ions and the Z^2/v^2 dependence of the energy loss (Bethe, 1930) results in a good separation of isobars especially for the lighter ions. Qualitatively, heavy ions passing swiftly through matter lose all the electrons for which orbital velocity is smaller than ion velocity (Bohr, 1940). Since the velocity of the innermost electrons increases proportionally to the atomic number Z, very high energies are required to strip off these electrons for the heavier ions. The dramatic increase in energy required to match the velocity of the last electron (hydrogen-like atom) is shown in figure 3. At these high energies, a relatively large fraction (> 10%) of the ions are fully stripped and a

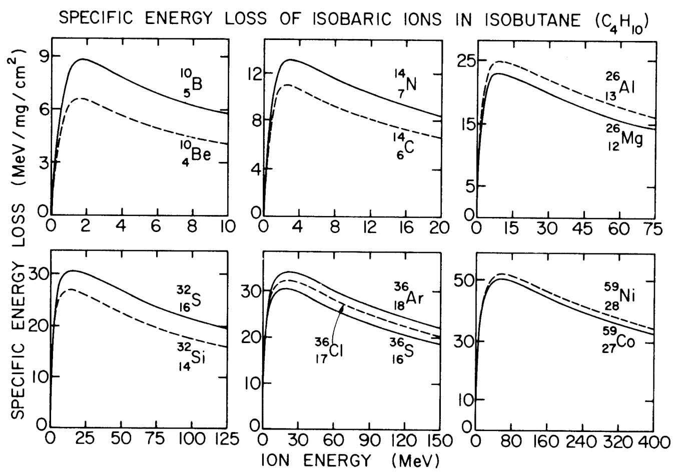

Fig 2. Energy loss in a typical counter gas of a heavy ion detector. The compilation of Ziegler (1980) was used to calculate the solid and dashed curves for stable and radioactive isotopes, respectively. N, Mg, and Ar do not form negative elemental ions (Mg and N, however, form negative hydride ions) and are, therefore, greatly suppressed in AMS with tandem accelerators.

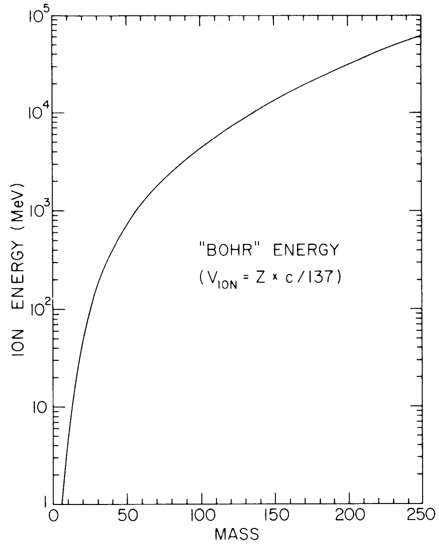

Fig 3. Ion energy required to strip off all but the last electron. According to Bohr (1940), all electrons with an orbital velocity smaller than the ion velocity are lost. At the "Bohr" energy, ca 10-20% of the ions are fully stripped.

separation of the radioisotope from the interfering stable isobar by magnetic or electrostatic devices is possible if the radioisotope has the higher Z (eg, ^{26}Al, ^{36}Cl, and ^{59}Ni in fig 2). The feasibility of this technique has been demonstrated for the isobar pairs ^{26}Al - ^{26}Mg (Raisbeck and Yiou, 1979), ^{41}Ca - ^{41}K (Raisbeck and Yiou, 1980), ^{59}Ni - ^{59}Co (Henning et al, 1981), and ^{36}Cl - ^{36}S (Gustavsson et al, 1981). Figure 4 gives an example of the ^{59}Ni - ^{59}Co separation when the tandem energy of 93.5 MeV was boosted to 328 MeV with the new superconducting linac at Argonne and the fully-stripped ions were separated magnetically. Table 1 gives the fraction of fully-stripped ions at energies presently available at Argonne. It is evident that much higher energies (cf fig 3) are required for an efficient stripping of Ni and Co . These will be available within a few years at Argonne and other new-generation heavy ion facilities. A drawback of these more complex accelerator systems is that they often require a beam feedback signal for a stable operation (eg, to synchronize the pulsing system of a dc beam from a tandem with the rf of a linac post-accelerator). Thus, the "zero-beam" radioisotope acceleration mode is not possible. The background beam of an ion-optically identical particle (^{59}Co in our example) must be

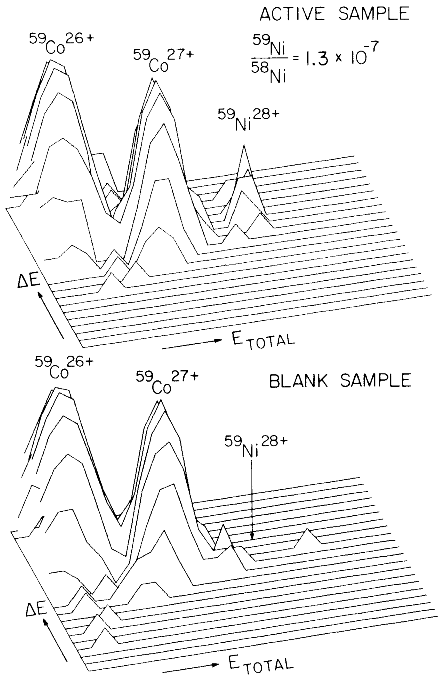

Fig 4. E_{total} vs ΔE spectra measured in a Si ΔE-E telescope detector system. Ions of 328 MeV were stripped in a 100 $\mu g/cm^2$ carbon foil and separated with a magnet set to accept mass-59, charge-28+ ions of full energy. The ^{59}Co background beam was 10^7 times stronger than the ^{59}Ni component before the charge state separation.

raised to the level required for the generation of the feedback signal. Although such an operation can guarantee long-term stability of the system, it has severe effects on the radioisotope detection sensitivity. The development of sophisticated beam pick-up devices operating at extremely low current levels are required if the sensitivity of free-running accelerators is to be matched.

SEARCH FOR THE UNKNOWN

One of the most exciting applications of AMS is the search for hitherto unobserved particle species (eg, quarks, anomalously heavy isotopes). Since particles with properties very different from normal matter are generally sought, the background can be very low and, consequently, the sensitivity very high. The main problem in these studies is that we are

TABLE 1. Some highly stripped heavy ion beams from the Argonne tandem-superconducting linac system

Ion	Energy (MeV)	Charge state	Fraction* (%)	"Bohr" energy** (MeV)
$^{17}_{8}O$	130	7^+	16	25
		8^+	84	
$^{35}_{17}Cl$	165	16^+	39.5	251
		17^+	11.5	
$^{40}_{20}Ca$	170	19^+	6.3	397
		20^+	0.4	
	260	19^+	33.7	
		20^+	8.7	
$^{59}_{27}Co$	328	25^+	0.97	1067
		27^+	0.023	
$^{58}_{28}Ni$	314	27^+	0.45	1128
		28^+	0.014	

*Measured after stripping in a 100μg/cm^2 carbon foil except ^{17}O where a 150μg/cm^2 gold foil was used.
**See figure 3

searching in the dark and must take great care to reliably scan a certain parameter space. Figure 5 summarizes searches for anomalously heavy isotopes (Klein, Middleton, and Stephens, 1981). Not included in this figure is the search for anomalous hydrogen in extremely enriched D_2O with a low-energy time-of-flight spectrometer (Smith _et al_, 1981). Due to the large enrichment factor ($\sim 10^{11}$), extremely low limits on the concentration of heavy hydrogen in normal hydrogen ($\sim 10^{-30}$) were reported.

The experimental limits for the concentration of the different species measured so far are all considerably lower than the theoretical predictions. These are based on considerations of the early universe, where, under extreme conditions of the big bang, a certain fraction of anomalous particles may have formed and survived until today. Although none of the searches have yet yielded positive result, they will be continued as long as we are curious.

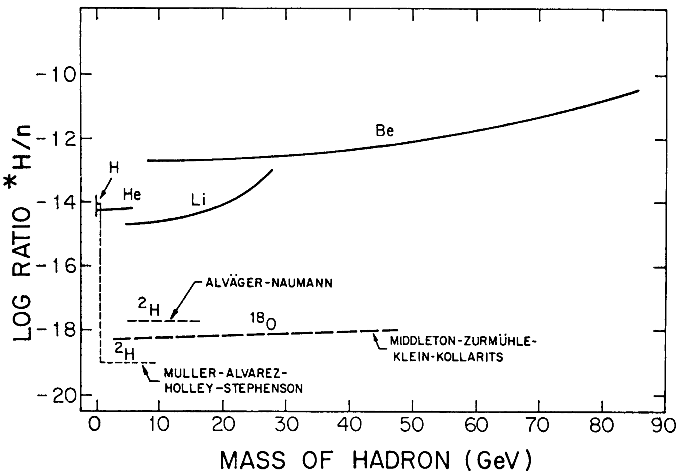

Fig 5. Summary of searches for anomalously heavy isotopes of various elements (Klein et al, 1981). He, Li, Be, and ^{18}O measurements were done with a tandem (Middleton et al, 1979; Klein et al, 1981), one ^{2}H search with a mass separator (Alväger and Naumann, 1967), and the other H and ^{2}H searches with a cyclotron (Muller et al, 1977).

ON THE USE OF COSMOGENIC RADIOISOTOPES

As mentioned above, the half-life of a radioisotope is not a limiting factor for the AMS technique. What counts most is the beam current from the sample. Since most of the AMS experiments use tandem accelerators, the efficiency of negative ion formation largely determines the lower detection limit. For many elements, negative ion beams of ca 1μA (~ 6 x 10^{12} ions/sec) can be produced. If we assume a 10% transmission from the ion source to the detection system (including charge state selection) and one count per hour as the lower limit to detect a radioisotope ion in a background-free condition, then we reach a radioisotope abundance limit of ca 10^{-15}. In practice, this limit may well turn out to be several orders of magnitude higher. Low negative ion yield, low transmission, and high background level can be reducing factors. It is interesting to note that, by far, the most used radioisotope in AMS, ^{10}Be, falls into the latter category.

Figure 6 gives an overview of the abundance of cosmogenic radioisotopes in various samples (Nishiizumi and Arnold, 1981). In general, the abundance is larger for extra-terrestrial material since the radioisotope is not diluted by

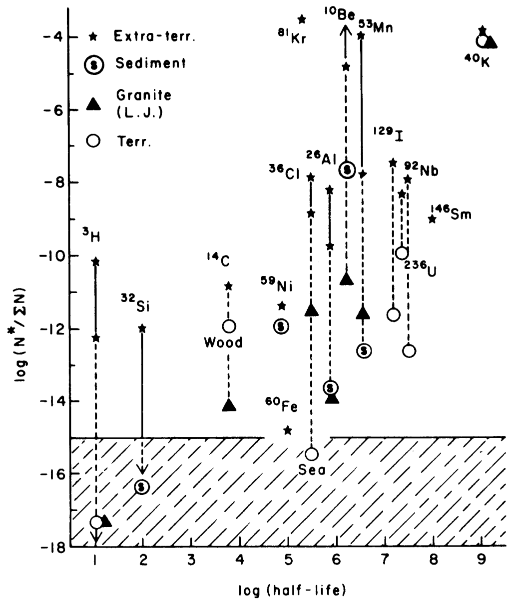

Fig 6. Ratio of cosmogenic radioisotopes, N*, to the sum of the respective stable isotopes, ΣN, as a function of the half-life in years in various natural materials (Nishiizumi and Arnold, 1981). The hatched area shows the detection limit of AMS for the most favorable conditions. Enrichment processes prior to the AMS measurement could lower the limit further.

the large reservoir of stable isotopes on earth. ^{10}Be has an exceptionally high abundance even in terrestrial material because of the low abundance of stable Be in nature (Suess and Urey, 1956). The high abundance makes it relatively easy to measure $^{10}Be/^{9}Be$ ratios with AMS despite the difficulties mentioned above. In addition, the specific energy loss separation of ^{10}Be from the interfering ^{10}B background is large (cf fig 2). Figure 7 summarizes AMS measurements with ^{10}Be. It shows that ^{10}Be has been measured in many different reservoirs covering a large range of concentrations. This great use is due to some outstanding properties of ^{10}Be : 1) high production rate in the atmosphere by spallation of nitrogen and oxygen (Lal and Peters, 1967), 2) short residence time in the atmosphere of about one year which makes it useful to study secular variations in the production rate with possible implications on cosmic ray variations (Raisbeck and Yiou, 1981; Wahlen et al, 1982), 3) long half-life (1.6×10^{6} yr) which allows us to study slow geophysical processes such as the growth rate of manganese nodules (Turekian et al, 1979) and the mixture of deep-sea sediments into lava (Brown et al, 1982).

As demonstrated in the pioneering work at Orsay (Raisbeck et al, 1978b, a) and in many measurements of this group thereafter, ^{10}Be can well be measured with a cyclotron. Although, at present, all other work in AMS is done with tandems, the cyclotron or any other accelerator starting with positive ions may well turn out to be an important alternative whenever elements are involved which do not easily form negative ions (or not at all as do the noble gases except He).

With respect to dating applications of ^{10}Be and other radioisotopes now available through AMS (eg, ^{26}Al, ^{32}Si, ^{36}Cl), it will be most important to measure ratios of different radioisotopes such as $^{10}Be/^{26}Al$ or $^{10}Be/^{36}Cl$ (Raisbeck and Yiou, 1981). In contrast to ^{14}C, there exists

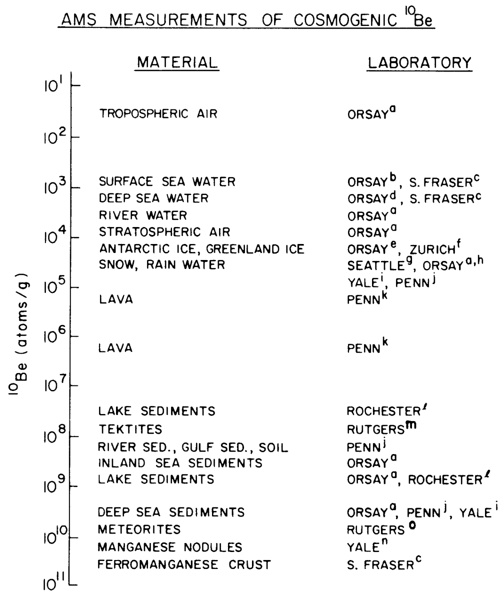

Fig 7. Summary of ^{10}Be measurements in various materials. The ordering is done according to the ^{10}Be concentration measured in the respective materials. [a]Raisbeck et al (1981), [b]Raisbeck et al (1979a), [c]Nelson et al (1983), [d]Raisbeck et al (1980), [e]Raisbeck et al (1978a), [f]Beer et al (1983), [g]Farwell et al (1983), [h]Raisbeck et al (1979b), [i]Thomas (1982), [j]Brown et al (1981), [k]Brown et al (1982), [l]Wahlen et al (1982), [m]Pal et al (1982), [n]Turekian et al (1979), [o]Kruse et al (1983).

no well-equilibrated "time-zero" abundance of any of these radioisotopes with respect to its stable isotope(s). The reliability of a date based on the time variation of the ratio of two or more radioisotopes will critically depend on the understanding of the respective chemical and physical fractionation effects.

THE MEASUREMENT OF HALF-LIVES WITH AMS

Perhaps one of the most important contributions AMS can make to dating is the measurement of uncertain or believed-to-be-certain half-lives. I became interested in this subject when I learned at the first conference on AMS at Rochester that the half-lives of some interesting long-lived radioisotopes produced by cosmic rays are still uncertain (Arnold, 1978).

Long half-lives can be determined uniquely from the relation $dN/dt = -\lambda N$, when both the decay rate, dN/dt, and the number of radioisotope atoms, N, is measured. The latter quantity is traditionally measured with low-energy mass spectrometry if sufficient material is available. Much lower radioisotope concentrations can be measured with AMS; thus, the method can be extended to more difficult cases such as ^{32}Si, the half-life of which was assumed to be ca 300 yr. AMS measurements at Argonne (Kutschera et al, 1980) and Rochester (Elmore et al, 1980) on different samples yielded half-lives of 101 ± 18 yr and 108 ± 18 yr, respectively. Figure 8 shows the history of ^{32}Si half-life measurements. First, we note an intriguing trend toward shorter half-life values with time. All the measurements marked with a cross relied on estimates of the respective production cross-sections, since the ^{32}Si concentration was not measured explicitly. Based on more recent cross-section systematics, arguments can be found (Kutschera et al, 1980) to bring these values down to the accelerator results. A serious disagreement exists with the results of two geophysical half-life measurements, where the depth profile of natural ^{32}Si activity was measured in ice-cores from Greenland (Clausen, 1973) and in a varved sediment core from the Gulf of California (Demaster, 1980). Assuming that the AMS results are correct, we are encouraged to measure the decay rate as a function of time for a half-life determination. A measurement over a period of a year or longer with a high-precision differential counting technique (Harbottle, Koehler, and Withnell, 1973) is currently underway at Brookhaven (Alburger, 1982 pers commun). It will hopefully give a decisive answer.

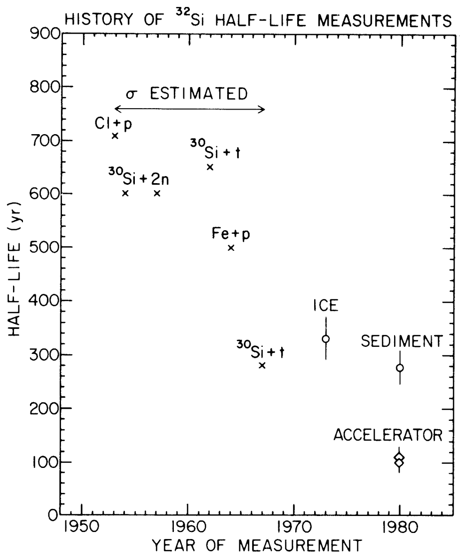

Fig 8. Summary of the attempts to measure a ^{32}Si half-life in the last 30 years (see text for references to the various measurements).

CONCLUSION

The encouraging results on high-precision measurements of $^{14}C/^{12}C$ or $^{14}C/^{13}C$ ratios described in various contributions to this issue justify optimism for this field. Details of the current status of the various facilities can be found in the respective contributions. Ironically, perhaps the strongest reaction to the original excitement about ^{14}C measurements with AMS was the development of miniature gas counters capable of measuring milligram quantities of carbon. The concurrent development of microprocessors allowing the automatic measurement of arrays of counters probably had a strong impact on this field as well.

It is interesting to speculate about a possible correlation between ^{14}C variations and ancient cultural events. Although it seems purely accidental, I should like to mention an amazing coincidence pointed out to me during a recent visit to Jerusalem (Paul, 1982, pers commun)

$$T_{1/2}(^{14}C) = 5730 \pm 40 \text{ yr}$$

$$\text{Genesis (Bible)} = 5743 \text{ yr B P}$$

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