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ACCELERATOR MASS SPECTROMETRY: FROM NUCLEAR PHYSICS TO DATING*

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1. 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 to me extremely exaggerated, but now I think there is a deep truth behind it. Once we build an instrument to investigate a specific problem we have realized the existence of the problem already and don't need the instrument to discover it. Therefore it is almost inevitable that the most interesting discoveries with a new technique will be done in fields the technique was not invented for. For example, accelerators which were built for nuclear physics produced an enormous amount of detailed information on nuclear structure and nuclear forces but the most fundamental discoveries were done with it in elementary particle physics. In any case, for almost

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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. And this in spite of the fact that in the very early days of accelerators the Lawrence cyclotron at Berkeley had been used as a mass spectrometer to discover ^3He in nature (Alvarez and Cornog, 1939 a, b). We are now in a situation where the enormous analytic power of accelerators is fully recognized, but the joy about this revival is mixed with some disappointment that the great prospects for studying radiocarbon problems (Muller, 1977 b; Bennet et al., 1977; Nelson et al., 1977) have not yet been fulfilled. Fortunately, some of the contributions to this meeting 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. This allows one to pick out almost any radioisotope of interest. Since the precision of conventional radiocarbon measurements cannot easily be achieved with such a highly complex instrument as an entire accelerator system, it is no wonder that one first concentrated on other radioisotopes. Fig. 1 gives an overview of the approximately 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 (5×10^2 – 5×10^4 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 the

proceedings of the Symposium on Accelerator Mass Spectrometry at Argonne (Henning et al., 1981 a). It seems quite possible that the whole range of radioisotopes shown in 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.

2. Some technical aspects of AMS

Fig. 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 whose orbital velocity is smaller than the ion velocity (Bohr, 1940). Since the velocity of the innermost electrons increases proportional 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 Fig. 3. At these high energies a relatively large fraction (> 10%) of the ions are fully stripped and a separation of the radioisotope from the interfering stable isobar by magnetic or electrostatic devices is possible if the radioisotope has the higher Z (^{26}Al , ^{36}Cl and ^{59}Ni in Fig. 2 are such cases). The feasibility of this technique has been demonstrated for the isobar pairs $^{26}\text{Al} - ^{26}\text{Mg}$ (Raisbeck et al; 1979 c), $^{41}\text{Ca} - ^{41}\text{K}$ (Raisbeck

and Yiou, 1980), ^{59}Ni - ^{59}Co (Henning et al., 1981 b) and ^{36}Cl - ^{36}S (Gustavsson et al., 1981). Fig. 4 gives an example of the ^{59}Ni - ^{59}Co separation where the tandem energy of 93.5 MeV was boosted to 323 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 for an efficient stripping of Ni and Co much higher energies (compare fig. 3) are required. These will be available within a few years at Argonne and other new-generation heavy ion facilities. There is, however, a drawback of using these more complex accelerator systems. They often require a beam feed-back signal for a stable operation (e.g. to synchronize the pulsing system of a d.c. beam from a tandem with the rf of a linac post-accelerator). Therefore, 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 raised to the level required for the generation of the feed-back signal. Although such an operation has the advantage of guaranteeing a 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.

3. Search for the Unknown

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

searching in the dark and great care must be taken to reliably scan a certain parameter space. Fig. 5 gives a summary of searches for anomalously heavy isotopes (Klein et al., 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. The predictions are based on considerations of the early universe, where under the extreme conditions of the big bang a certain fraction of anomalous particles may have been formed and survived until today. Although none of the searches have yielded any positive result so far, they will certainly be continued as long as we are curious enough.

4. On the Use of Cosmogenic Radioisotopes

As mentioned earlier 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 about $1 \mu A$ (6×10^{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 about 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 this latter category.

Fig. 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 the large reservoir of stable isotopes on earth. As can be seen ^{10}Be has an exceptionally high abundance even in terrestrial material. This is caused by the low abundance of stable Be in nature (Suess and Urey, 1956). The high abundance makes it relatively easy to measure $^{10}\text{Be}/^9\text{Be}$ ratios with AMS in spite of 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). Fig. 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. A high production rate in the atmosphere by spallation of nitrogen and oxygen (Lal and Peters, 1967) 2. A short residence time in the atmosphere of about one year. This makes it useful to study secular variations in the production rate with possible implications on cosmic ray variations (Raisbeck, 1981 a; Wahlen, 1982). 3. A long half-life (1.6×10^6 yr) which allows one 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., 1978 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 (e.g. ^{26}Al , ^{32}Si , ^{36}Cl) it will be most important to measure ratios of different radioisotopes such as $^{10}\text{Be}/^{26}\text{Al}$ or $^{10}\text{Be}/^{36}\text{Cl}$ (Raisbeck, 1981 a). In contrast to ^{14}C there exists 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.

5. 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. With AMS much lower radioisotope concentrations can be measured and the method can therefore be extended to more difficult cases. One such case is ^{32}Si whose half-life was assumed to be around 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. Fig. 8 shows the history of ^{32}Si half-life measurements. The first thing to note

is 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 one is 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 et al., 1973) is currently underway at Brookhaven (Alburger, 1982). It will hopefully give a decisive answer.

6. Conclusion

Although this is a radiocarbon conference I have not discussed ^{14}C measurements with AMS. The encouraging results on high-precision measurements of $^{14}\text{C}/^{12}\text{C}$ or $^{14}\text{C}/^{13}\text{C}$ ratios described in various contributions to this conference 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 had probably had a strong impact on this field as well.

Inspired by a comment in Dr. Korff's talk that we should look for grand correlations of ^{14}C variations with ancient cultural events I was reminded of an amazing coincidence pointed out to me during a recent visit to Jerusalem (Paul, 1982)

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

Genesis (bible) = 5743 yr B.P.

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

Ion	Energy (MeV)	Charge State	Fraction* (%)	"Bohr" energy** (MeV)
$^{17}_8\text{O}$	130	7^+	16	25
		8^+	84	
$^{35}_{17}\text{Cl}$	165	16^+	39.5	251
		17^+	11.5	
$^{40}_{20}\text{Cl}$	170	19^+	6.3	397
		20^+	0.4	
	260	19^+	33.7	
		20^+	8.7	
$^{59}_{27}\text{Co}$	328	26^+	0.97	1067
		27^+	0.023	
$^{58}_{28}\text{Ni}$	314	27^+	0.45	1128
		28^+	0.014	

*Measured after stripping in a $100 \mu\text{g}/\text{cm}^2$ carbon foil except ^{17}O where a $150 \mu\text{g}/\text{cm}^2$ gold foil was used.

**See figure 3.

Figure Captions

- Fig. 1** Overview of long-lived radioisotopes. Data are taken from Lederer and Shirley (1978) except ^{32}Si (W. Kutschera et al., 1980; D. Elmore et al., 1980) and ^{202}Pb (Nagai et al., 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.
- 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 are lost which have an orbital velocity smaller than the ion velocity. At the "Bohr" energy about 10-20% of the ions are fully stripped.
- Fig. 4** E_{total} versus ΔE spectra measured in a Si ΔE -E telescope detector system. Ions of 328 MeV were stripped in a $100 \mu\text{g}/\text{cm}^2$ 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.
- Fig. 5** Summary of searches for anomalously heavy isotopes of various elements (Klein et al., 1981). The He, Li, Be and ^{18}O measurements were done with a tandem (Middleton et al., 1979; Klein et al., 1981), one ^2H search with a mass separator (Alvåger and Naumann, 1967), and the other H and ^2H searches with a cyclotron (Muller et al., 1977 a).

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.

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. ^aRaisbeck et al. (1981 b), ^bRaisbeck et al. (1979 b), ^cNelson et al. (1982), ^dRaisbeck et al. (1980 a), ^eRaisbeck et al. (1978 b), ^fBeer et al. (1982), ^gFarwell et al. (1982), ^hRaisbeck et al. (1979 a), ⁱThomas (1982), ^jBrown et al. (1981), ^kBrown et al. (1982), ^lWahlen et al. (1982), ^mPal et al. (1982), ⁿTurekian et al. (1979).

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).

Fig. 1

SPECIFIC ENERGY LOSS OF ISOBARIC IONS IN ISOBUTANE (C_4H_{10})

Fig. 2

Fig. 3

ACTIVE SAMPLE

$$\frac{^{59}\text{Ni}}{^{58}\text{Ni}} = 1.3 \times 10^{-7}$$

BLANK SAMPLE

Fig. 4

Fig. 5

Fig. 6

AMS MEASUREMENTS OF COSMOGENIC ^{10}Be

Fig. 7

Fig. 8

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