CONF-8410172--7

DE85 007994

ACCELERATOR MASS SPECTROMETRY WITH A COUPLED TANDEM-LINAC SYSTEM

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Abstract

Accelerator mass spectrometry is at present almost exclusively performed with tandem accelerators. The extension of this technique to higher energies by coupling a linear accelerator to the tandem is discussed. Particular emphasis is placed on describing the suppression of strong stable-isobar backgrounds in the detection of long-lived radioisotopes.

I. Introduction

In this paper I should like to discuss the use of a coupled tandem-linac system for experiments in accelerator mass spectrometry (AMS). After the paper by W. Wölfli[1] describing all the wonderful things one can do with AMS using a well-equipped tandem accelerator, one may wonder why one wants to complicate things further and use two instead of just one accelerator. Maybe one of the reasons is, that some of us like complicated things or are simply "addicted to tuning". However, there are also some other reasons, in particular that a coupled system provides higher energies, which allows one to extend AMS to hitherto nearly untouched mass regions. Another important reason is that the complexity, although bothersome for the operation, increases the selectivity of detecting a particular isotope. These arguments are valid for essentially any heavy-ion accelerator combination which is capable of delivering higher energy than a single tandem.

Throughout the world, there exist quite a large variety of different accelerator combinations including tandems, linacs and cyclotrons. The present use of the specific tandem-linac combination for AMS is due to the fact that AMS was almost exclusively developed around tandem accelerators. Therefore the tandem-linac combination presents a natural extension to higher energies. The use of negative ions has some particular advantages in suppressing background from unwanted elements that do not form stable negative ions (e.g. N, Mg, Ar). On the other hand, this limits the detection of isotopes to elements which do form negative ions. For particular problems it

may therefore be advantageous to use accelerators which start with positive ions from the source. What in the end really matters most for choosing one or the other machine is to what extent the entire accelerator system can be operated in a truly quantitative way, from the ion source to the detection system.

In general, AMS is used to measure very small radioisotope-to-stable isotope ratios in an ion source sample. This is done by alternating the system between radioisotope and stable isotope acceleration. The latter provides usually enough beam intensity to measure electric currents in a Faraday cup and use the beam for various on-line diagnostic and feed-back purposes. The radioisotope, however, provides typically only a few ions per second, sometimes masked by orders of magnitude more background ions. The radioisotope intensity is measured by ion counting in a suitable detection system after the acceleration. Although in some cases the background beam is strong enough and ion-optically suitable to provide control signals, in general one runs in a "zero-current" mode for radioisotope detection. This presents a non-trivial problem for the operation of a tandem accelerator and even more so for a tandem-linac combination. The particular operational problem of this combination is that both machines are mass selective, but in different ways. For a constant accelerating field, a linac has a certain mass acceptance for isotopes entering with the same charge and velocity, and for a fixed terminal voltage, a tandem accelerates all isotopes of the same charge to the same energy but different velocity. In addition there are usually magnetic analyzing and focussing elements in the beam line system which are selective to yet another mass dependent quantity, the momentum-to-charge ratio. Finally, there is usually a pulsing system which bunches and chops the dc tandem beam to proper pulses matching the linac rf. This introduces again mass dependency through time-of-flight effects. Without going into further details, it is clear that all this provides on the one hand a very mass-selective device, but on the other hand introduces severe operational problems for alternating between isotopes. Although it is conceivable that the rapidly growing computer-assisted operation of accelerators will eventually allow an automated switching, this is at present not possible at a quantitatively reproduceable level. Therefore manual-tuning is still required, much to the delight of the "tuning-addicts". A particularly careful tuning is required for the measurement of absolute isotopic ratios, as in the ^{60}Fe experiment

described below. If calibrated standards are available the task is much simpler, since isotope dependent effects cancel in a relative measurement between a standard and a sample of unknown isotopic ratio.

II. The Separation of a Radioisotope from a Strong Isobaric Background

The basic goal in AMS is the measurement of very low concentrations ($\gtrsim 10^{-15}$) of long-lived radioisotopes. For every radioisotope with mass $A \leq 209$ there is at least one stable isobar in nature (except for A = 5 and 8). Chemical purification below the ppb level (10^{-9}) is very difficult, and the mass difference between radioactive and stable isobar is in general too small to result in any useful selectivity in the acceleration process. Therefore, the basic problem of the final detector system is to separate the radioisotope from the stable isobar, which arrives with the same mass, energy and charge state from the accelerator. If the count-rate of the stable isobar in the detector is not excessive, dE/dx measurements at sufficiently high energies allow a Z identification of the isobars and an unambiguous detection of the radioisotope. However, quite often the stable-isobar background is very high, in particular if a common element with a high electron affinity is involved. It is then necessary to separate and suppress the intense stable isobar to a level which is acceptable for the final detector system. Three different methods have been used to suppress an excessive background of stable isobars:

1. Outranging

If $Z_{\text{Radioisotope}} < Z_{\text{Stable Isobar}}$ an absorber of suitable thickness can stop the stable isobar completely and allow only the radioisotope to enter the final particle detector. This method is widely used[2,3] to suppress $^{10}_{5}B$ in the detection of $^{10}_{4}Be(T_{1/2} = 1.6 \times 10^{6}$ y). For heavier elements it becomes increasingly difficult to separate and identify the radioisotope cleanly by outranging the stable isobar. This is due to the smaller relative difference in Z between radioisotope and stable isobar, combined with an increase in range straggling. Higher energy helps to some extent, but extremely uniform absorbers are needed, as discussed in the separation of ^{14}C from ^{14}N using a cyclotron.[4]

2. Fully Stripping

If $Z_{Radioisotope} > Z_{Stable\ Isobar}$ and sufficiently high energy is available, a very effective separation can be achieved by fully stripping the ions in a thin foil and separating the highest charge states in a magnetic field. Only ions of this highest charge state are allowed to enter the final detector. This method has been applied to several radioisotope-stable isobar systems including $^{26}_{13}Al$ - $^{26}_{12}Mg$ (ref. 5), $^{36}_{17}Cl$ - $^{36}_{16}S$ (ref. 6), $^{41}_{20}Ca$ - $^{41}_{19}K$ (ref. 7), and $^{59}_{28}Ni$ - $^{59}_{27}Co$ (ref. 8). In all cases coupled accelerator systems were used to achieve energies sufficiently high to fully strip the ions. For ^{26}Al and ^{41}Ca the linac-cyclotron system ALICE at Orsay was used. For the ^{36}Cl and ^{59}Ni detection the tandem-linac systems of Munich and Argonne, respectively, were used. A simple estimate for the ion energy needed to strip off all electrons with a probability of about 10% or higher can be obtained from the so-called Bohr criterion.[9] This states that all electrons with orbital velocities smaller than the ion velocity are stripped off. As a rule of thumb we therefore need ion velocities equal to or higher than the orbital velocity of the last electron (hydrogen-like atom). This velocity is $v_{ion} = Z\ v_o$, where Z is the nuclear charge and v_o is the velocity of the electron in the hydrogen atom ($v_o = c/137$). This means that for an effective stripping of all electrons the ion energy for heavier elements has to increase proportional to Z^2.

For the experiments mentioned above the following energies were used (in brackets we give the Bohr estimate): ^{26}Al, 200 [109] MeV; ^{36}Cl, 153 [258]; ^{41}Ca, 310 [407]; ^{59}Ni, 324 [1148]. The demand for very high energy of the heaviest isotopes is evident. For example, for ^{59}Ni we measured[8] at 328 MeV a fraction of only 10^{-4} $^{59}Ni^{28+}$, three orders of magnitude lower than the one expected at the "Bohr" energy of 1.1 GeV. Such high energies will be available in the near future for this mass range at the full ATLAS facility at Argonne. From recent experiments with fully stripped ions of ^{197}Au (ref. 10) and ^{238}U (ref. 11) at the Bevalac in Berkeley, one finds that 40 to 50% of the ions are fully stripped at 79 and 104 GeV, respectively. The corresponding Bohr estimates give energies of 31 and 50 GeV. Although at present these largest heavy ion machines are not used for AMS, this could well happen some day in the future, when a problem worth the effort can be found.

3. Energy Loss Difference

This method is the most general one, since it can be applied for $Z_{\text{Radioisotope}} \neq Z_{\text{Stable Isobar}}$. Isobaric ions passing through a foil or gas layer of suitable thickness loose different amounts of energy. This can be used to spatially separate isobars in an electric or magnetic field. The intense stable isobar component can then be physically blocked from entering the final detector system. We have used this technique at Argonne for the systems $^{32}_{14}Si$ - $^{32}_{16}S$ (ref. 12) and $^{60}_{26}Fe$ - $^{60}_{28}Ni$ (ref. 13), and at GSI Darmstadt for the very heavy system $^{205}_{82}Pb$ - $^{205}_{81}Tl$ (refs. 14,15). For an efficient separation the incident energy should be well above the energy for the maximum in the dE/dx curve. This maximum shifts in energy for heavier ions approximately proportional to $Z^{4/3}$. Compared to the full-stripping technique, which requires an energy increase proportional to Z^2, the energy loss separation can be applied at much lower energies. For example, the difficult ^{205}Pb-^{205}Tl separation was performed at 2.3 GeV whereas fully stripping, which is in principal applicable for this case, would require about 20 times higher energies.

The separation of isobars through energy loss difference is applicable to all long-lived radioisotopes in nature. Energies in the range from 5 to 15 MeV/nucleon suffice to separate even the heaviest isobars by this method. Heavy ion energies of this range are available at a number of accelerators throughout the world.

Although the universality of the energy loss separation technique is a great asset for AMS, the separating power is somewhat lower than for the full-stripping method. The main reason for this is that the absorber needed to get the appropriate energy-loss difference has to be approximately 20 times thicker than a typical stripper foil. This introduces a comparatively large line broadening through energy-loss straggling. Great uniformity of absorber thickness is mandatory.[16] In addition, the spatial separation of isobars in a magnet due to the nuclear charge difference is in general larger than the one that can be achieved through energy loss difference. All this leads to the conclusion that fully stripping results in a cleaner separation, but it is, of course, limited to cases where the radioisotope has a higher nuclear charge than the stable isobar.

In the following I should like to discuss briefly two experiments performed with fully stripped radioisotopes (^{36}Cl, ^{59}Ni) and one with the energy loss technique (^{60}Fe). All three experiments were performed with tandem-linac systems.

III. Detection of ^{36}Cl and ^{59}Ni by Fully Stripping

The detection of natural concentrations of ^{36}Cl ($T_{1/2} = 3.0 \times 10^5$ y) was one of the first goals of AMS.[17] The tandem laboratories of the University of Rochester[17] and the Weizmann Institute in Rehovot[18] have both shown that $^{36}Cl/Cl$ ratio measurements down to a level of about 10^{-15} can be performed by chemically purifying the samples from ^{36}S. After careful chemistry the ^{36}S background was tolerable for direct acceptance in the final detector system. A group in Munich[19], however, developed recently a ^{36}Cl detection method through fully stripping, by boosting tandem accelerated ions with a linear post accelerator from 87.5 MeV to 153 MeV. The ions were then stripped in a 110 $\mu g/cm^2$ thick carbon foil which left 7% of the ^{36}Cl ions in the 17^+ charge state. These fully stripped ions were subsequently separated from the lower charge state in a magnetic deflection system and counted in a Bragg-curve ionization chamber.

Rather than reducing ^{36}S in the Cl samples, this method allowed to deliberately add about 0.1% ^{36}S to the samples. The resulting ^{36}S beam was used as a pilot beam for slit stabilization of the tandem terminal voltage and for various beam monitoring purposes. The suppression of the ^{36}S pilot beam by the combination of stripping and magnetic deflection was larger than 10^7. Figure 1 shows two spectra of natural samples measured[19] in the ionization chamber, when the deflection system was set to detect $^{36}Cl^{17+}$ ions. Figure 1a shows the full range of energies vs. nuclear charge. Besides the clearly separated $^{36}Cl^{17+}$ ions with the full beam energy, there is a tail of lower charge state ions whose energy matches the magnetic rigidity of $^{36}Cl^{17+}$. A small natural $^{18}O_2^-$ contribution extracted from the ion source leads to a $^{18}O^{8+}$ component. Figure 1b shows an expanded view of the region of highest charge states for a measurement of a groundwater sample from a well used by the Löwenbräu brewery in Munich. The $^{36}Cl/Cl$ ratio of $(6.3 \pm 0.8) \times 10^{-13}$ found for this water is not unusual and hence does not reveal the secret of the famous taste of Munich beer.

At Argonne we performed[8] a fully stripping experiment with the much heavier radioisotope ^{59}Ni ($T_{1/2} = 7.5 \times 10^4$ y). In this experiment the energy of the tandem accelerated ions was boosted from 93.5 MeV to 328 MeV with our superconducting linac. The ions were then stripped in a 100 $\mu g/cm^2$ thick carbon foil which left only 0.014% of the ^{59}Ni ions in the 28+ charge state. The fully stripped ions were subsequently separated in a dipole magnet and counted in a ΔE-E silicon surface barrier telescope. A pilot beam of the stable isobar ^{59}Co was produced by mixing approximately equal amounts of ^{59}Co with Ni. The Ni contained ^{59}Ni with a ratio of $^{59}Ni/Ni = 1.3 \times 10^{-7}$ produced by neutron activation in a reactor. Figure 2 shows spectra similar to the one of the ^{36}Cl measurement discussed above. The charge state distribution of the ^{59}Co ions clearly indicates that the beam energy is far below the one required to efficiently strip off all electrons. As mentioned earlier, this energy is about 1.1 GeV and will be available at the full ATLAS system in the near future. The good separation of $^{59}Ni^{28+}$ from $^{59}Co^{27+}$ is seen in Fig. 2b. The ^{59}Co background at the stripper was about 10^6 times higher than ^{59}Ni.

The use of a ^{59}Co pilot beam was convenient for tuning purposes, tandem slit stabilization and provision of a beam-phase pick-up signal between tandem and linac. The latter is required to synchronize the pre-tandem buncher to the linac rf phase. Fortunately, it turned out during the ^{60}Fe experiment described below that the tandem-linac system can also be operated in a stable mode without the use of the pre-tandem bunching system. The post-tandem bunching system does not require a phase-lock signal from the beam. Compared to the full pulsing sytem in operation one looses a factor 2 in transmission through the linac and gets a beam slightly degraded in energy and time resolution. However, this loss in beam quality is irrelevant for the AMS experiments. Since the tandem can be stabilized with a generating voltmeter we now operate the tandem-linac system without a pilot beam. This means that AMS experiments can be extended to radioisotopes for which a separation from a pilot beam would not be possible. In the next section I describe such an experiment where our tandem-linac system was used for the first time to measure absolute radioisotope concentrations.

IV. Detection of ^{60}Fe by Energy Loss Difference

The radioisotope ^{60}Fe is an interesting long-lived isotope, whose half-life was only poorly known before our measurement[13]. The half-life had been measured once before by Roy and Kohman[20], who found a value of $T_{1/2} \simeq 3 \times 10^5$ y, uncertain by a factor of 3. We have used[13] the Argonne tandem-linac system to measure the concentration of artificially produced ^{60}Fe in an Fe sample. From the ^{60}Fe/Fe ratios of $(9.54 \pm 1.40) \times 10^{-8}$ and the decay rate which we measured by gamma counting of the daughter product ^{60}Co, we found a half-life of $T_{1/2} = (1.49 \pm 0.27) \times 10^6$ y. The half-life was calculated from the relation $dN/dt = -\lambda N$. The new half-life value is significantly longer than the previous one and may have a number of interesting astrophysical implications as discussed in ref. 13. Details of the experimental technique used for the AMS measurement are also described in this paper.[13] Here, I should like to discuss some of the technical aspects of the ^{60}Fe/Fe ratio measurement which relates to using a coupled tandem-linac system.

The basic problem for the AMS measurement was to suppress a strong ^{60}Ni background. Since Ni is a common constituent of ion source components, chemical purification of the Fe sample did not significantly reduce the background. In addition, ^{60}Ni was used between ^{60}Fe measurements to check the timing status of the accelerator and beam-line system. This resulted in a background of ^{60}Ni ions after acceleration, which was about 10^6 times higher than ^{60}Fe. Since ^{60}Fe has Z = 26 and ^{60}Ni has Z = 28, the full-stripping technique cannot be used to remove ^{60}Ni. Instead we used a stack of 28 uniform Al foils, each 100 $\mu g/cm^2$ thick, to disperse the energy of the isobars by their difference in energy loss. At an incident energy of 320 to 360 MeV the ^{60}Fe ions loose about 63 MeV and the ^{60}Ni ions about 70 MeV. The difference in energy after the stack resulted in a spatial separation of 4 cm between ^{60}Fe and ^{60}Ni in the focal plane of an Enge split-pole spectrograph. Despite considerable energy straggling the bulk of the intense ^{60}Ni ions could be blocked from entering the focal-plane ionization chamber by appropriate Ta shields.

An energy loss vs. total energy spectrum measured in the ionization detector is shown in fig. 3. Although there is still a fairly intense ^{60}Ni component scattered into the focal plane region of ^{60}Fe, the ^{60}Fe particles

are clearly separated from ^{60}Ni through their different energy-loss and total energy signals. The spectrum shown in the figure was measured from a sample containing ^{60}Fe/Fe = 1 x 10^{-7}. Background runs with blank samples indicated that our present detection limit lies at 5 x 10^{-12}. Improvements in ion source output, in ^{60}Ni suppression in the ion source, and in the straggling effects of the absorber are required to lower the detection limit further. The goal is to reach the sensitivity where ^{60}Fe/Fe ratio measurements in iron meteorites are possible (~ 1 x 10^{-14}).

Figure 4 shows a summary of all ^{60}Fe/Fe ratio measurements performed for the half-life measurement. The three runs indicated in the figure were separated by 9 and 4 months, respectively. This means the operating conditions were quite different. Therefore the consistency of the results is very satisfactory. The normilization of ^{60}Fe to the bulk Fe of the sample was achieved by measuring the ^{56}Fe current at the entrance of the linac. At present, the linac plus the beam-line system to the spectrograph cannot be alternated quantitatively back and forth between ^{60}Fe and ^{56}Fe. We therefore measured the transmission of ^{60}Fe from the linac entrance to the spectrograph using the ^{60}Ni beam. We are convinced that a careful tuning of all relevant parameters by different people under various operating conditions provides a rather reliable method to establish data for such an absolute ratio measurement with the claimed uncertainty of about 15%.

The ^{60}Fe experiment was a first step towards using even more complex accelerators of higher energy for quantitative AMS experiments. The largest machine used so far for AMS is the UNILAC accelerator at GSI Darmstadt, where we performed[14,15] an exploratory experiment to detect the very heavy radioisotope ^{205}Pb ($T_{1/2}$ = 1.5 x 10^{7} y) and separate it by the energy loss technique from ^{205}Tl. I am confident that the higher energies available at future tandem-linac combinations and other even larger heavy ion accelerators of various kinds provide an exciting opportunity for AMS experiments with very heavy radioisotopes.

The author should like to thank W. Henning and P. W. Kubik for discussions on the subject of this paper. This work was supported by the U. S. Department of Energy under Contract W-31-109-Eng-38.

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DISCLAIMER

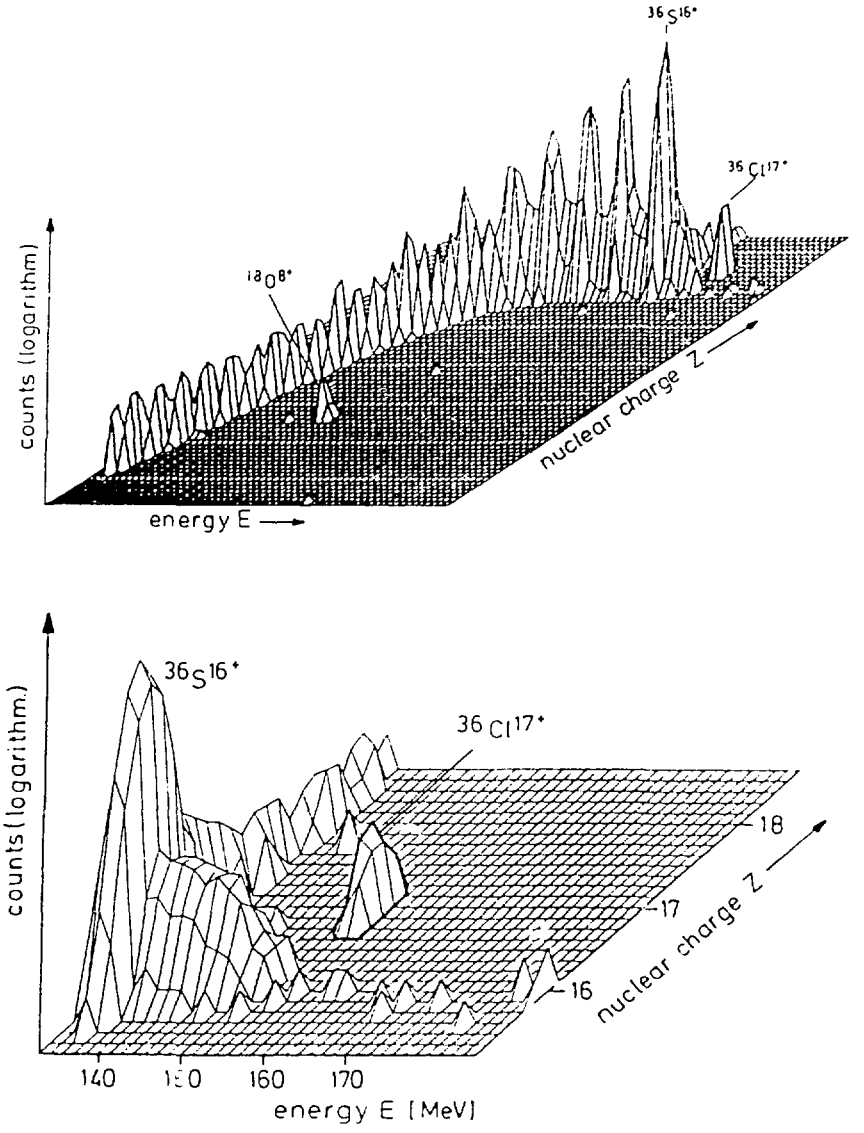

Fig. 1 Three dimensional spectra of $^{36}Cl + ^{36}S$ ions stripped at 153 MeV in a carbon foil (from ref. 6). The spectra were measured with a Bragg-curve spectrometer, which was located after a magnetic deflection system at the position of $^{36}Cl^{17+}$ ions.

a) Complete ion spectrum of a natural Cl sample with $^{36}Cl/Cl$ of about 10^{-12}.

b) Expanded view of the highest charge state region for a ground-water sample from the Löwenbräu well with $^{36}Cl/Cl = 6.3 \times 10^{-13}$.

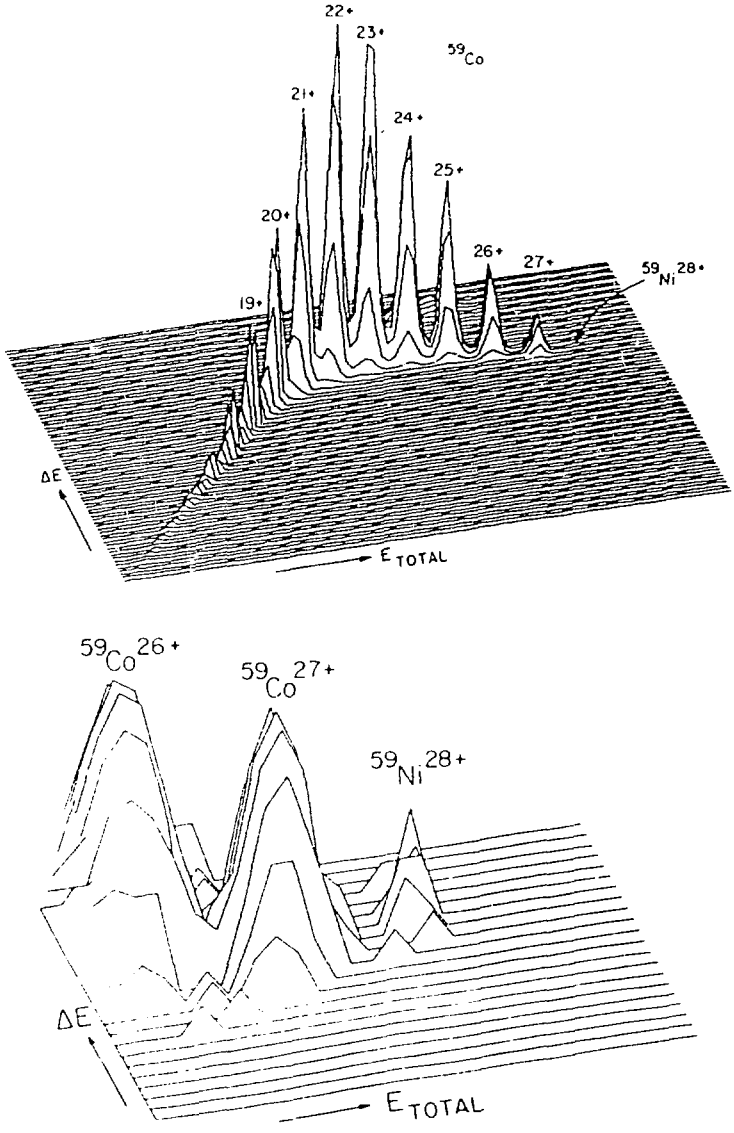

Fig. 2 Three dimensional spectra of ^{59}Ni + ^{59}Co ions stripped at 328 MeV in a carbon foil (from ref. 8). The spectra were measured with a ΔE-E silicon surface-barrier telescope, which was located after a dipole magnet set to deflect $^{59}Ni^{28+}$ ions into the detector.

a) Complete ion spectrum for a sample with $^{59}Ni/Ni = 1.3 \times 10^{-7}$. The intensity scale is linear.

b) Expanded view of the highest charge state region for the same sample. The intensity scale is logarithmic.

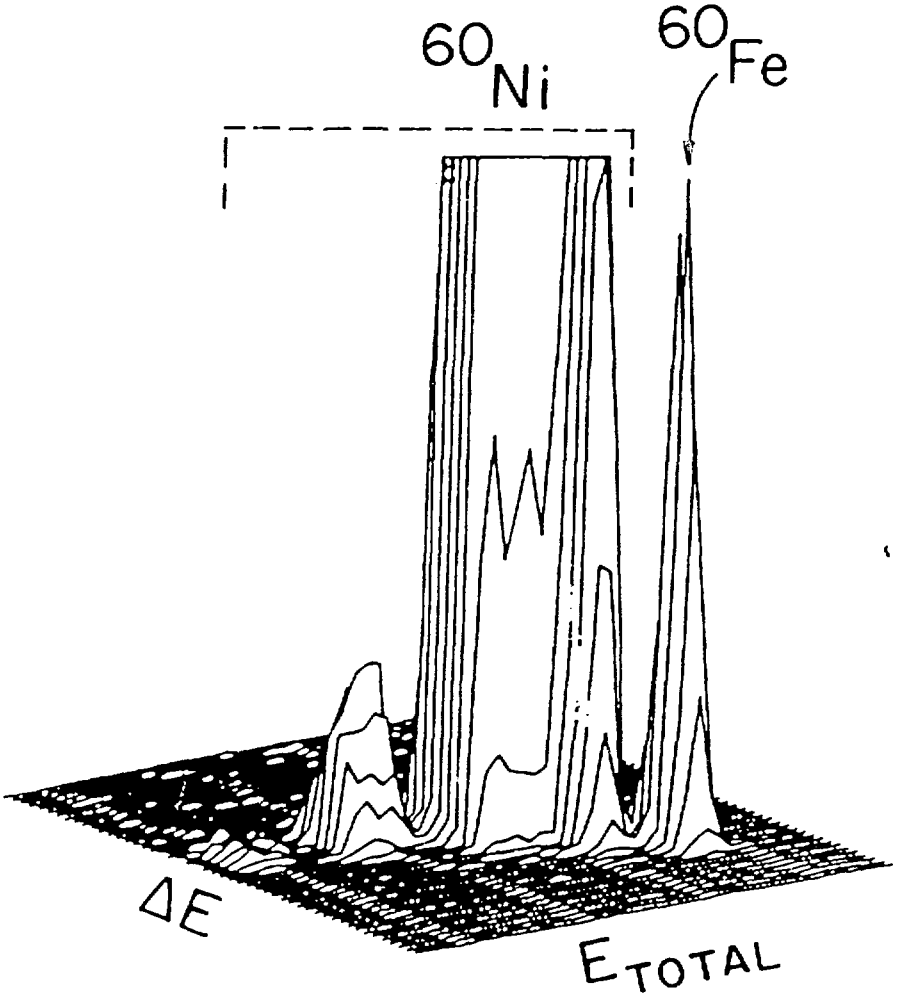

Fig. 3 Three dimensional spectrum of $^{60}Fe + ^{60}Ni$ ions after passing through a 2.8 mg/cm^2 thick Al absorber (from ref. 13). The spectrum was measured in the focal-plane ionization chamber of the split-pole spectrograph, with an aperture in front of the detector selecting the region of $^{60}Fe^{21+}$ ions. The $^{60}Fe/Fe$ ratio in the sample was 1×10^{-7}. The intensity scale is linear.

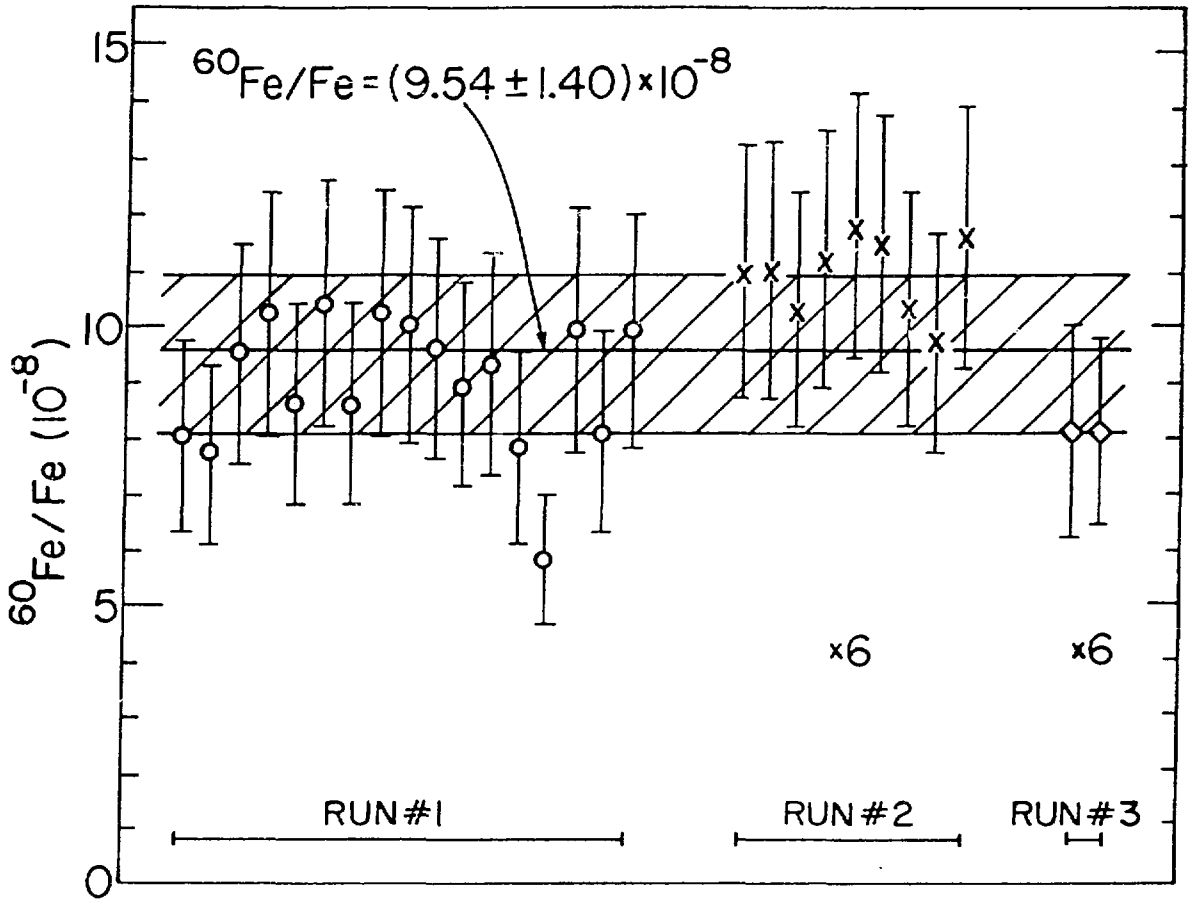

Fig. 4 Summary of the $^{60}Fe/Fe$ ratio measurements (from ref. 13). Run #1 was performed in February 1983 with 360 MeV ^{60}Fe ions, Run #2 in November 1983 and Run #3 in March 1984, both at 320 MeV. The ^{60}Fe content of the samples used in Run #2 and #3 was chemically diluted by a factor 6. The measured $^{60}Fe/Fe$ ratio has therefore been multiplied by the same factor to compare them with the result of Run #1.