RADIOISOTOPE DETECTION WITH THE ARGONNE FN TANDEM ACCELERATOR*

WALTER KUTSCHERA,** WALTER HENNING,
MICHAEL PAUL, E J STEPHENSON, and J L YNTEMA

Argonne National Laboratory, Argonne, Illinois 60439

ABSTRACT. A standard heavy ion nuclear structure facility has been used to detect several long-lived radioisotopes by counting the accelerated ions. The problem of eliminating strong isobaric background beams has been solved by combining the energy loss dispersion through a uniform Al foil stack with the high momentum resolution of an Enge split-pole magnetic spectrograph. Radioisotope concentrations in the following ranges have been measured: $^{14}C/^{12}C = 10^{-12}$ to 10^{-13}, $^{26}Al/^{27}Al = 10^{-10}$ to 10^{-12}, $^{32}Si/Si = 10^{-8}$ to 10^{-14}, $^{36}Cl/Cl = 10^{-8}$ to 10^{-11}.

INTRODUCTION

The Argonne National Laboratory has operated an electrostatic FN tandem accelerator since 1967. In 1973 and 1977, some essential parts of the accelerator were replaced in order to meet the stringent requirements as an injector for the newly developed super-conducting linear accelerator. The conversion of the tandem to metal-bonded ceramic-titanium accelerator tubes with straight electrode geometry, a pelletron charging system and an enclosed corona-tube voltage distribution system, and the use of pure SF_6 insulating tank gas resulted in a machine of ion-optic simplicity, high voltage stability up to 9 MV terminal voltage, and an ultra high vacuum system ($\approx 10^{-8}$ Torr). At present, both the tandem alone and the combined system tandem-linac are almost entirely devoted to heavy ion nuclear physics research. Negative heavy ion beams are routinely produced from an "inverted" cesium-beam sputter source (Chapman, 1976), which can also take small sputter samples of a few milligrams. A major analyzing instrument for heavy ion spectroscopy at the tandem is an Enge split-pole magnetic spectrograph with an ionization chamber focal-plane detector (Erskine, Braid, and Stoltzfus, 1976). This instrument is well suited to identify tandem-accelerated ions by mass and atomic number up to mass 40 and with some improvements probably up to mass 60.

The conditions described above encouraged us to start experiments in the newly revived field of accelerator mass spectroscopy, which is exactly 40 years old (Alvarez and Cornog, 1939a,b). Reviews on the recent developments are given by Muller (1979), Purser, Litherland, and Gove (1979), and Bennet (1979). Our own activity in this field started about nine months ago. The intention was to use as much of the existing experimental facility as possible in order to fit the measurements smoothly into the nuclear physics research program. In addition, rather than concentrating only on ^{14}C and trying to set up a system for radiocarbon dating, we aimed towards developing a technique applicable for a large variety of radioisotopes.

One of the major obstacles in accelerator spectroscopy is the sometimes overwhelming background from stable isobaric beam components. Fortunately, this problem does not exist for ^{14}C measurements with the tandem since ^{14}N does not form stable negative ions. However, the detection of various other radioisotopes of interest suffers severely from the problem. Examples are the isobaric pairs $^{36}_{17}Cl - ^{36}_{16}S$ and, in particular, $^{32}_{14}Si - ^{32}_{16}S$. We will describe briefly our technique to solve this problem, below. A more detailed description will be given elsewhere (Henning and others, ms in preparation).

EXPERIMENTAL TECHNIQUE

Figure 1 shows a schematic view of the essential parts of the radio-isotope detection system at Argonne. It illustrates the measurement of $^{36}_{17}Cl$ in the presence of a $^{36}_{16}S$ background. The basic concept of our method is to disperse the energy of the isobaric pair through a uniform foil stack (Clerc and others, 1975) and measure the energy difference
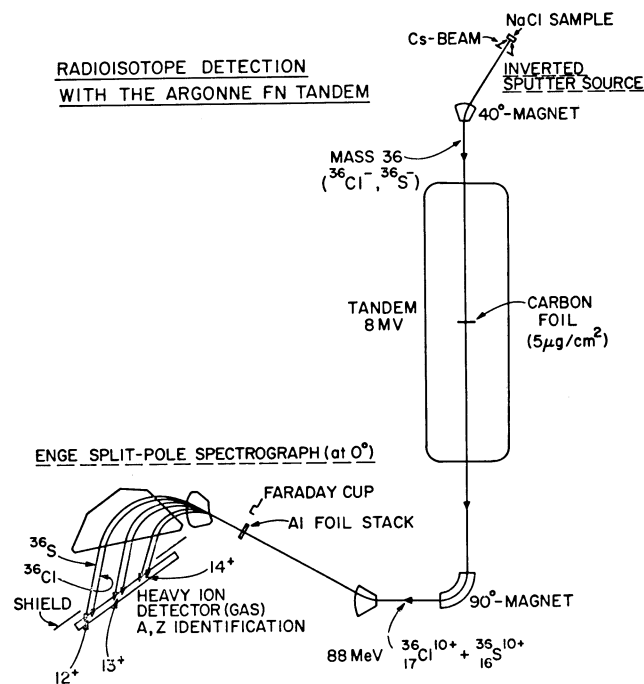

Fig 1. Schematic illustration of a ^{36}Cl measurement with the radioisotope detection system at Argonne. Additional negative-ion background components not shown in the figure are $^{18}O_2^-$ and $^{12}C_3^-$.

accurately with a magnetic spectrograph. The energy-loss difference due to different atomic numbers, results, then, in energy- and spatially-separated components at the focal plane of the spectrograph. If the background lines are high in intensity, they can be conveniently shielded from the focal plane detector. This turned out to be an important option in some cases since the resolution of the detector starts to deteriorate if the total counting rate exceeds 1000 counts/sec. The detector provides additional energy-loss, total energy, and position $(B\rho)$ signals which allow a clear identification of mass and atomic number.

The separation through the foil stack is only useful if the energy straggling is small compared to the energy-loss difference. We investigated several foil materials, including C, Al, Si, and Au in different foil arrangements. Single thick foils (1 to 4mg/cm^2) and stacks with up to 40 thin foils were studied. Non-uniformity and pinholes were the major obstacles. The best peak-to-valley ratio for adjacent components and an energy
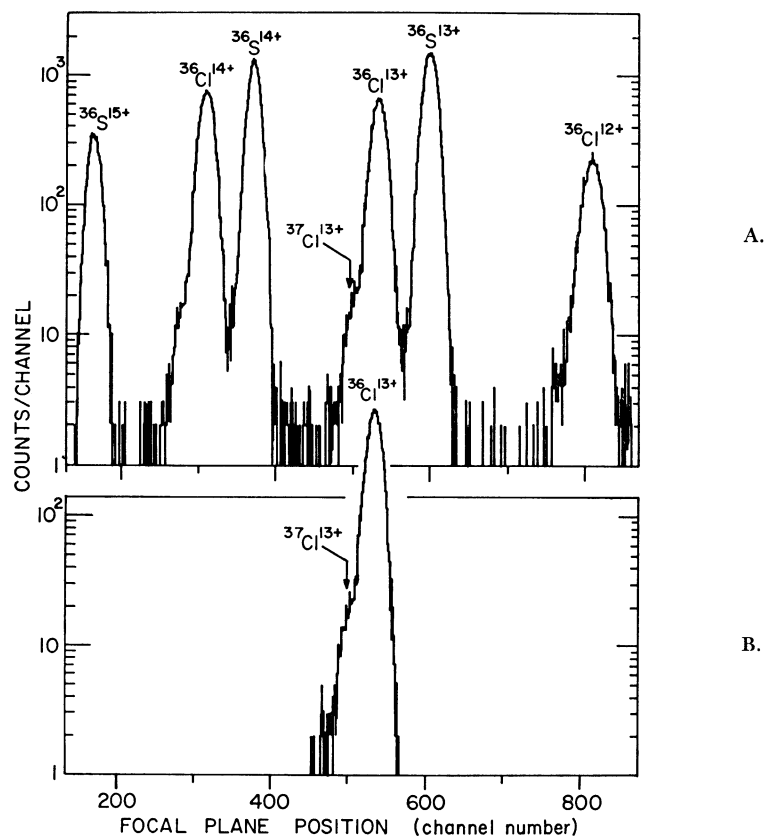
Fig 2. A) Gated position $(B\rho)$ spectrum from an NaCl sample with ^{36}Cl/C$l \approx 10^{-8}$ as measured in the focal-plane detector of the spectrograph. Window set on the ΔE–E spectrum of figure 3. B) Gated position spectrum with an additional window set on Cl^{13+} ions as shown in figure 3.

straggling close to the one expected from theory was obtained for a stack of 15 Al foils, each $100\mu g/cm^2$ thick. All measurements, except for ^{14}C for which a thin carbon foil is sufficient, have been performed with this Al stack. Figure 2 shows the total position spectrum in the spectrograph for an incident beam of 88MeV $^{36}Cl^{9+}$ + $^{36}S^{9+}$. The difference in energy loss for the two components is 2.5MeV and corresponds to a spatial separation of 4cm. Additional information is provided by the data in the ΔE vs E and E vs $B\rho$ plane (fig 3). Appropriate windows in these spectra clean up the position spectrum as demonstrated in the lower part of figure 2.

Measurement of the radioisotope concentration from a sample in the ion source, requires a reliable normalization to the beam current of the stable isotope(s). We decided to make this measurement as close to the spectrograph as possible. For this purpose, a small Faraday cup with suppressor ring and a 3mm entrance aperture was interchanged in posi-
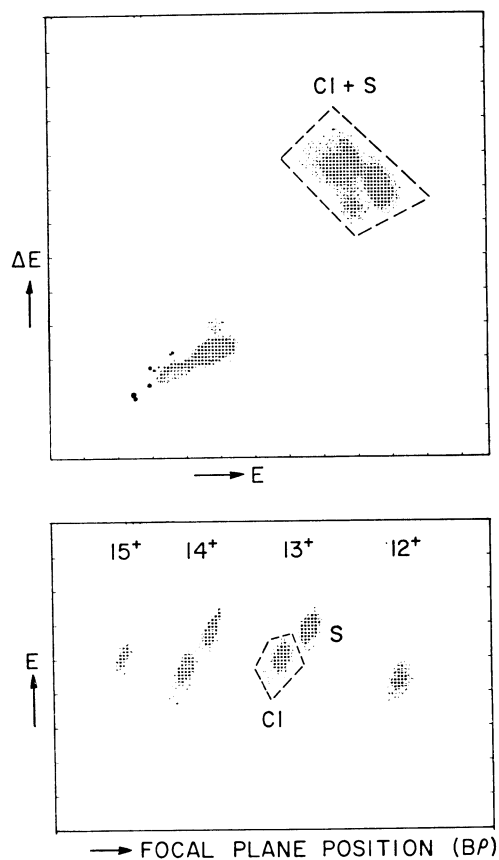

Fig 3. ΔE vs E and E vs $B\rho$ spectra for the same sample as in figure 2. Windows set in these spectra are indicated. The low-lying group in the ΔE vs E spectrum is due to lighter beam contaminants (probably carbon and oxygen).

tion with the energy-loss foil stack in front of the spectrograph (fig 2). The whole focusing system from the ion source to the spectrograph was set up first with the stable-isotope beam by requiring the current on the entrance aperture of the Faraday cup to be close to zero. Then the 40° inflection magnet (see fig 1) was set to the radioisotope mass and the terminal voltage was changed in such a way as to match the magnetic rigidity of the stable-isotope beam. None of the other beam-optical elements were touched. The final settings of the inflection magnet and the terminal voltage were verified by scanning in small steps around the precalculated values and optimizing the counting rate of the radioisotope in the spectrograph. The terminal voltage control was provided by a generating voltmeter (Brookshier, Wallace, and Langsdorf, 1971). The entrance and exit slits of the 90° analyzing magnet were wide open. An exception was the run with ^{32}Si, when the ^{32}S background beam was strong enough to allow tandem stabilization by slit control. In these measurements, the energy was kept constant and the complete accelerator system was adjusted separately for ^{32}Si + ^{32}S and ^{30}Si by optimizing the beam current in the spectrograph Faraday cup.

Until now, all changes were made manually as in the routine operation of the tandem. It takes approximately 5 to 10 minutes to switch between radioisotope and stable isotope running condition. Although a

TABLE 1

Summary of radioisotope measurements

Radio-isotope	Sample	Ratio	Measured (10^{-12})	Expected (10^{-12})
^{14}C	Charcoal	^{14}C/^{12}C	1.06 ± 0.15	1.144 ± 0.011*
	"Fermi" graphite**	^{14}C/^{12}C	4.5 ± 0.7	5.137 ± 0.041*
	Graphite	^{14}C/^{12}C	0.20 ± 0.05	low
^{26}Al	$^{27}Al(\gamma, n)$	^{26}Al/^{27}Al	290 ± 60	410 ± 50***
	$^{27}Al(^{16}O, ^{17}O)$	^{26}Al/^{27}Al	91 ± 30	~100†
	^{27}Al	^{26}Al/^{27}Al	~4	low
^{32}Si	Si (t, p)	^{32}Si/Si	$\sim10^4$	10^3–10^4‡
	Si $(^{18}O, ^{16}O)$	^{32}Si/Si	0.095 ± 0.050	<0.1§
^{36}Cl	NaCl (n, γ)	^{36}Cl/Cl	$14,900 \pm 2200$	$12,900 \pm 700$¶
	NaCl (n, γ)	^{36}Cl/Cl	1360 ± 200	1450 ± 100¶
	NaCl (n, γ)	^{36}Cl/Cl	100 ± 20	36.8 ± 2.0¶
	NaCl	^{36}Cl/Cl	60 ± 20	low

* Measured from β activity with a sample from the same material by D D Coleman, Illinois State Geological Survey, Urbana.

** Graphite probably originating from the first reactor pile in Chicago built by E Fermi in 1942.

*** Al beam stop from the 20MeV electron linac in Argonne. ^{26}Al content determined independently from activity (1.8MeVγ-ray) and $^{27}Al(\gamma, n)$ cross-section.

† Al sample irradiated with 1×10^{17} ^{16}O ions (56MeV) from the tandem. ^{26}Al content estimated assuming $\sigma(^{16}O, ^{17}O) = 1$mb.

‡ Part (1.3mg) of a K_2SiF_6 sample produced by D Alburger and G Harbottle, Brookhaven National Laboratory, from the irradiation of $^{30}SiO_2$ with 2.58×10^{19} tritons (3.4MeV). ^{32}Si content estimated from β activity.

§ Si single crystal irradiated with 4×10^{15} ^{18}O ions (56MeV) from the tandem. ^{32}Si content estimated assuming $\sigma(^{18}O, ^{16}O) = 1$mb.

¶ NaCl samples irradiated in the Argonne CP5 reactor. ^{36}Cl content determined from ^{24}Na activity, ^{23}Na(n, γ) and $^{35}Cl(n, \gamma)$ cross-section.

computerized system might improve the precision in the switching procedure, other factors such as the current measurement of the stable-isotope beam and the background subtraction are the dominant contributions to the errors given in table 1.

RESULTS

Table 1 summarizes the results of our measurements. As mentioned previously, our aim was to determine how well our method works for a variety of radioisotopes. We shall briefly discuss each radioisotope.

^{14}C. The charcoal sample originated from a piece of wood burned in a fireplace. Agreement with the β activity measurement is satisfactory. The so-called "Fermi" graphite sample was obtained from stock graphite at Argonne. Originally intended to be used for background determination, it turned out to be fairly radioactive. If all the ^{14}C was produced from $^{13}C(n, \gamma)$, this graphite must have been exposed to a total thermal neutron
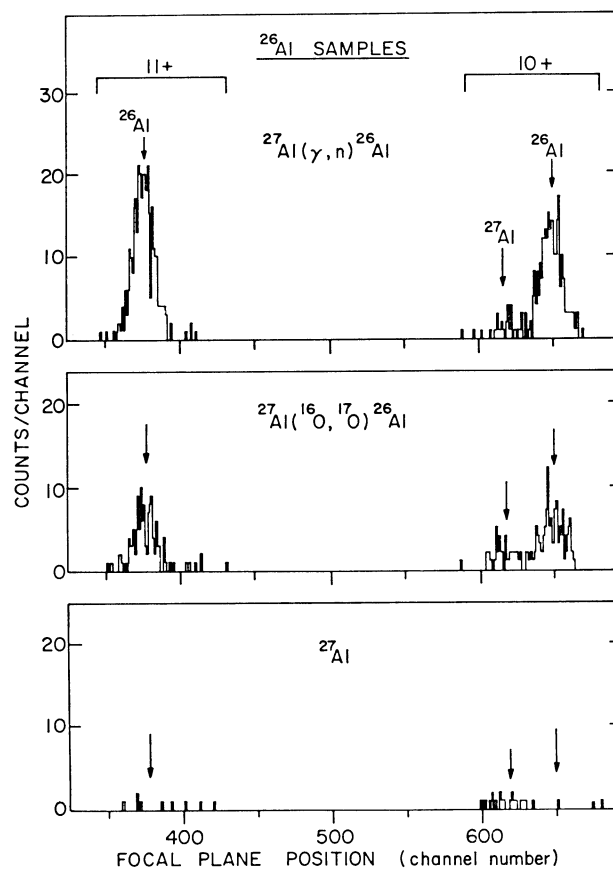

Fig 4. Gated position spectrum from the three Al samples (see table 1). Windows have been set on $^{26}Al^{11+}$ and $^{26}Al^{10+}$ ions, respectively. Some ^{27}Al probably originating from double charge exchange in the tandem can be seen.

flux of 10^{17} neutrons/cm^2. Another graphite sample showed a relatively high ^{14}C background. It seems unlikely that this is the inherent background from the ion source. Typical running conditions were: $^{12}C^- =$ 1 μA, TV (terminal voltage) = 6 MV, $^{13}C^{4+} = 15$ nA in the spectrograph cup (used for normalization).

^{26}Al. Figure 4 shows ^{26}Al spectra measured for the three Al samples. Typical running conditions were: $^{27}Al^- =$ few nA, TV = 8 MV, $^{27}Al^{8+}$ = 2 nA. The spectra shown in figure 4 were obtained in about 30 minutes each. The low background observed in the blank sample suggests that measurements below 10^{-12} are feasible. However, for such measurements, it is desirable to increase the output of the ion source by at least an order of magnitude. No indication of the existence of $^{26}Mg^-$ has been observed.

^{36}Cl. Several samples with known ^{36}Cl concentrations were prepared. Results on the highly enriched samples agree well with expected values. The origin of the ^{36}Cl content in the blank sample is not known. The $NaCl$ material was taken from a window for infrared spectroscopy. Typical running conditions were: $^{37}Cl^- = 40$ nA, TV = 8 MV, $^{37}Cl^{10+} =$ 1.5 nA. For the highest enriched sample, the $^{36}Cl^{13+}$ counting rate in the focal-plane detector was 10 counts/sec while the $^{36}S^{13+}$ rate was about twice as high (see fig 2).
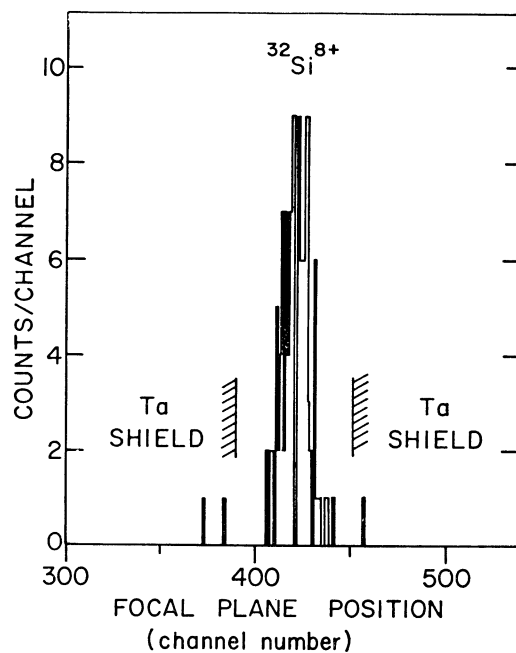

Fig 5. Gated position spectrum of $^{32}Si^{8+}$ ions from the K_2SiF_6 sample (see table 1). A 4cm wide Ta slit in front of the focal-plane detector shields the ^{32}S background. Windows are set on $^{32}Si^{8+}$ ions (see fig 6).

^{32}Si. All Si samples produced a $^{32}S^{9+}$ current between 0.03 to 0.3 nA at the spectrograph cup. Since this corresponds to 10^7 to 10^8 ^{32}S ions, careful shielding of the focal-plane detector is mandatory. Even so, the tails of the sulfur peaks forced us to select a low intensity charge state emerging from the Al foil stack. We chose the $^{32}Si^{8+}$ component which contains only 0.76 percent of the incident $^{32}Si^{9+}$ ions (80MeV). Figure 5 shows the ^{32}Si peak as obtained with the K_2SiF_6 sample [see footnote ‡ in table 1]. A total of 122 $^{32}Si^{8+}$ ions were counted in 56 minutes. A normalizing current of only 0.074 nA of $^{30}Si^{9+}$ was measured in the spectrograph cup. The isotopic abundance of ^{30}Si in this sample was approximately 5 percent. A background sample of enriched $K_2{}^{30}SiF_6$ material (95.6 percent ^{30}Si) produced ≈500 times higher ^{30}Si currents. In 15 minutes, *zero* counts were observed in the region for $^{32}Si^{8+}$ ions (see fig 6) which corresponds to an upper limit of $^{32}Si/Si < 5 \times 10^{-12}$. The highest source current, 10 μA of $^{28}Si^-$, was obtained from the Si single crystal sample [see footnote § in table 1]. From this sample with natural Si abundance (*ie*, 3.1 percent ^{30}Si) a normalizing current of 35 nA $^{30}Si^{9+}$ was measured in the spectrograph cup. Since the ^{32}S background was lowest with this sample, we measured $^{32}Si^{9+}$ ions (6.7 percent charge state probability) emerging from the Al foil stack. In 33 minutes, 10 counts of $^{32}Si^{9+}$ ions were detected. The $^{32}Si/Si$ ratio from these values (table 1) indicates that measurements in the 10^{-14} range are possible for ^{32}Si under favorable conditions.

CONCLUSION

It appears that accelerator mass spectroscopy can be performed at a tandem accelerator used normally for heavy ion nuclear physics research without major additional investments in equipment. The precision and sensitivity obtained with the described technique should allow the investigation of a number of interesting problems.
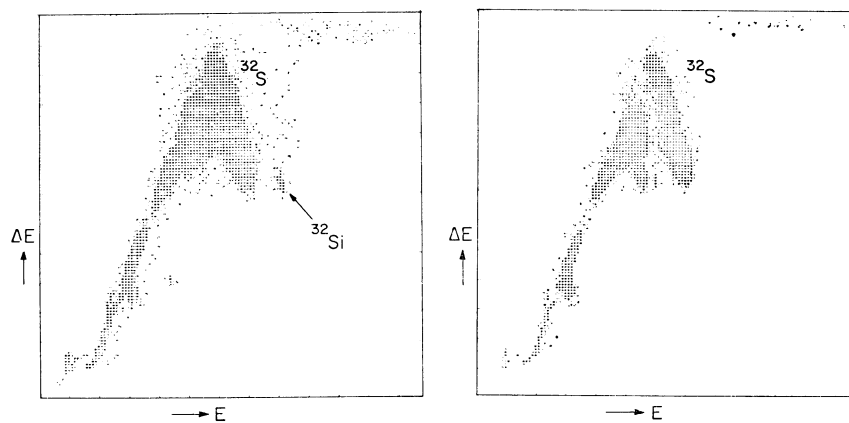

Fig 6. ΔE vs E spectra for the K_2SiF_6 samples. The irradiated sample (left) shows a clear ^{32}Si peak, whereas the background sample (right) has zero counts in the same region.

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