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THE GAS-FILLED MAGNET: AN ISOBAR SEPARATOR FOR ACCELERATOR MASS SPECTROMETRY

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Abstract

The most difficult problem for accelerator mass spectrometry is the rejection of stable atomic isobars. The intensity of isobaric interference is expected to become a problem for ^{36}Cl measurements with the use of new high-intensity ion sources. Although better chemical separation may be possible through improved sample preparation, the device expected to help most with this problem is the gas-filled magnet. We tested a gas-filled Enge split-pole spectrograph combined with a multi-plate gas ionization detector for the separation of ^{36}S from ^{36}Cl and obtained an isobar separation of about two orders of magnitude better than that possible with the detector alone.

1. Introduction

Accelerator mass spectrometry (AMS) is characterized by its very high sensitivity for measuring long-lived radioisotopes in natural samples [1]; isotopic ratios as low as a few times 10^{-15} can be determined for $^{36}Cl/Cl$. The main limitation for lower concentrations of ^{36}Cl is the inability to completely separate ^{36}Cl from its stable isobar, ^{36}S. For other long-lived radioisotopes such as ^{53}Mn, ^{59}Ni, ^{60}Fe, ^{79}Se, and ^{126}Sn, interference from the stable isobar prevents AMS detection at natural levels. The Rochester AMS system for ^{36}Cl measurements in the 10^{-15} range requires the interference rate of ^{36}S not to exceed a $^{36}S/^{36}Cl$ counting ratio of 10^5 to 10^6 which is equivalent to a sulphur concentration of about 1 ppm. The use of new high-

intensity ion sources will make possible measurements below the 10^{-15} level provided that ^{36}S, whose counting rate will inevitably increase, can be rejected sufficiently. Improved chemical separation will help, however removal of ^{36}S from very small samples is difficult and reduces the Cl yield. Also, substantial ^{36}S is produced from sample holders once a hole has been sputtered through the sample.

The device expected to help most in reducing background is the gas-filled magnet first proposed by Cohen and Fulmer [2] for fission product identification. The idea of employing a gas-filled magnet in an AMS set-up for isobar separation was successfully used by Kutschera et al. [3] to separate ^{60}Fe from ^{60}Ni and Henning et.al. [4] to separate ^{41}K from ^{41}Ca. However, the ion energies obtained with their tandem-linac combination were far above those attainable at most AMS laboratories. To evaluate the performance of a gas-filled magnet at lower energies, we used the University of Rochester Enge split-pole magnetic spectrograph to separate of ^{36}S from ^{36}Cl.

2. Principle of Operation

Consider a beam of particles of analysed mass and energy impinging on a thin foil which separates the gas-filled region of a magnetic dipole from the accelerator vacuum. The frequency of collisions of the ions with gas molecules is proportional to the gas pressure and the cross section. In each of these collisions the charge state can change due to electron loss or capture. At low gas pressures ($<10^{-2}$ torr) the number of collisions is small and the ion trajectories are determined by the charge state distribution obtained in the entrance window. In the presence of gas, frequent collisions cause the ionic trajectories to oscillate about a trajectory determined by the average charge state (Fig 1). Although the statistical width of the trajectory distribution decreases with increasing collision frequency, multiple scattering processes contribute a broadening at higher pressures. At intermediate pressures of a few torr, there exists a broad minimum in peak width.

Although the ions lose energy transversing the gas, the magnetic rigidity stays approximately constant, because the average charge state $\langle q \rangle$ is

approximately proportional to the ion velocity ($\langle q \rangle \approx vZ^k$ with $k \approx 0.45$ [5]). For isobars (ions having the same mass but different nuclear charge Z), the average charge states and therefore the average trajectories are different. The ratio of the width to the separation of the trajectories is a measure of the resolution. These properties together with computer simulation of heavy-ion trajectories in a gas-filled magnet are discussed in detail by Paul et al.[6].

Although the gas-filled magnet has great promise, early tests showed that it would be hard to improve on the performance obtained for our gas ionization detector, which has a rejection factor for 80 MeV ^{36}S of about 10^6 (20 counts per hour background from 3000 counts per second of ^{36}S with no more than 10% loss of ^{36}Cl). However, the detector performance deteriorates rapidly above 3000 counts per second and new ion sources are expected to produce ^{36}S rates higher than this. Therefore, our only hope is the gas-filled magnet which has the important characteristic of keeping most of the interference from entering the detector. Since the gas-filled magnet alone is not expected to show a rejection factor of better than 10^3 to 10^4, we recognized early on that it was important to minimize to energy loss in the magnet in order to leave as much energy as possible for further separation in the detector. This was done by minimizing the path length in the gas and minimizing the gas pressure within the broad minimum in peak width.

3. Experimental

We used the NSRL Enge split-pole magnetic spectrograph and a split-anode gas ionization detector to test the ^{36}Cl-^{36}S isobar separation at the energy used in previous ^{36}Cl AMS experiments (80 MeV). We modified the "Rochester heavy ion detector" [7] to have five 5-cm wide ΔE plates split diagonally for position measurement (see Fifield et al. [8]). The position spectrum was distorted somewhat by the wires of the Frisch grid which had a spacing 2.3 mm, perpendicular to the rays. Although this had little effect on the overall selectivity, the distorted peak shapes caused difficulty in determining the best magnet gas pressure. We installed an externally-operated shutter in front of the high magnetic rigidity end of the detector to obstruct the ^{36}S. A 5-cm Si position-sensitive detector was used to measure the vertical size of the beam.

The detectors were placed in the spectrograph which is on a beam line with no electrostatic analysis; this allowed passage of considerable isotopic interference. However, by using a high-level ^{36}Cl sample ($^{36}Cl/Cl=33x10^{-12}$) we were able to identify ^{36}Cl in spite of considerable ^{35}Cl and ^{37}Cl counting rates (Fig 2). We used 0.21 mg/cm^2 thick mylar windows for the spectrograph entrance and for the detector entrance. The magnet entrance window was located near the pole face to minimize the region of "dead" gas ("dead" here means a region of negligible magnetic field). An ideal system would have little or no "dead" gas, since this gas contributes to multiple scattering and energy loss but not to Z separation. The detector was moved out of the focal plane and as close as possible to the magnetic field to minimize "dead" gas. This did not create a focussing problem, since the ion beam entering the spectrograph was well collimated. In spite of these measures, the fraction of "dead" gas was about 1/3 of the total path length.

We compared the isobar separation with the detector at each end of the camera box. The ^{36}S and ^{36}Cl peaks were closer together at the low magnetic rigidity end, but were also narrower because of the shorter path length in the gas. The resolving power (the ratio of the difference in position to FWHM of the peaks) was comparable at the two ends (Fig. 3). The low magnetic rigidity end was preferable because the residual energy available for the energy loss measurements in the detector was greater there.

4. Results

We evaluate the performance of isobar separation using the ^{36}S rate divided by the ^{36}Cl-free background at the ^{36}Cl location in the spectra. We set gates on each parameter: four ΔE's, total energy and position. The computer required a six-fold coincidence. We set the gates tight enough to reject only 5-10% of the ^{36}Cl events (less than 1% per tail). To ensure that there was no ^{36}Cl in the beam we used a NiS sample, attenuated by a variable aperture. In past ^{36}Cl runs, we have typically obtained between 10^5 and 10^6 separation between ^{36}S and ^{36}Cl using the gas detector alone at energies in the range of 60-100 MeV. For a gas-filled magnet to be useful we needed to do better than this with the combined system. In our best configuration we obtained 10^8 rejection of ^{36}S. The parameters for this measurement are given in Table 1.

Nitrogen was used in the magnet for the above measurements. We also tried helium gas since it was expected to reduce multiple scattering. The pressure required to reach equilibrium of the average charge state was 20 torr. The separation was similar to that obtained for nitrogen. However, the ions exiting the magnet had only 42 MeV in the gas detector, while when using nitrogen the energy was 54 MeV. The helium configuration showed lower rejection since the detector separation is better at higher energies.

The vertical size of the beam at the detector was measured with the position-sensitive silicon detector under slightly different conditions. The magnet gas pressure was 4.5 torr nitrogen and the magnet entrance window was 37 cm from the entrance pole face. Under these conditions the FWHM of the ^{36}S peak as 17 mm, the separation of the isobars was 52 mm and the vertical FWHM was 14 mm.

5. Conclusions

The isobar separation we obtained for the ^{36}Cl-^{36}S system is about two orders of magnitude better than the best we could obtain with the gas detector alone. It should be possible to improve this somewhat by reducing the "dead" gas region. We plan to install a dedicated gas-filled magnet at Rochester to overcome the expected problems associated with the use of a high-intensity ion source for ^{36}Cl, and to attempt detection of heavier isotopes such as ^{53}Mn, ^{59}Ni, ^{60}Fe, and ^{79}Se.

Several specific design parameters for the incorporation of a gas-filled magnet in our AMS set-up can be obtained from our tests with the Enge spectrograph. A 90° dipole magnet is probably adequate. Magnets with a larger angle of bend like the 300° magnet used in the isotope separator JOSEF [9] will lead to a wider spacial separation; however, long flight paths will result in a lower energy in the detector. We found a better rejection using a radius of curvature of 47 cm instead of 85 cm, on the spectrograph. It would be difficult to go much lower than this because of the field limitation of a room temperature magnet. For the detection of ^{79}Se at an energy of 130 MeV, a 50-60 cm radius would be needed. A pole face gap of 3-4 cm should be sufficient. The beam transport and magnet optics should allow focussing the

beam onto the detector entrance. The effects of a field index and angles to the entrance pole face should be considered. It is probably not possible to obtain converging rays following the magnet, so the best configuration is to place the detector at the magnet exit.

The performance of a gas-filled magnet is not accurately measured by the FWHM/separation ratio but by the counting rate of the interfering isobar at the location of the radioisotope. The low energy tail is especially a problem for the case where the radioisotope has the higher Z. It could be that smaller values of FWHM/separation, which one gets at higher pressures result in more intense tails.

Since several radioisotopes are being measured sensitively with small tandems (below 3 MV), it would be of considerable interest to be able to measure ^{36}Cl with these also. Ordinarily, we measure ratios in the 10^{-15} regime by using high energies (60-100 MeV) to achieve an isobar separation of 10^5 to 10^6. Elmore et al.[10] report that at 18 MeV the detection limit with our gas detector alone is only about $^{36}Cl/Cl = 10^{-12}$. At 80 MeV, the gas-filled magnet improved the separation by only 2 orders of magnitude; we would expect even less improvement at 18 MeV. Since 3 orders of magnitude are needed, it is unlikely that low-level ^{36}Cl measurements will be competitive with a 3 MV accelerator. When we have installed a gas-filled magnet on our AMS beam line, we will pursue this problem further.

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Table 1

Parameters for ^{36}S-^{36}Cl separation with the Rochester Enge split-pole spectrograph containing nitrogen gas

magnetic rigidity at center of detector	47 cm
magnetic field	12.3 kG
beam	$^{36}S^{7+}$, 80 MeV
magnet gas pressure (nitrogen)	3 torr
detector gas pressure (isobutane)	33 torr
window thicknesses	0.21 mg/cm^2 mylar
distance of entrance window from pole face	3 cm
distance of detector from magnet exit	39 cm
energy at detector	54 MeV
^{36}S counting rate	900,000 cps *)
counting rate in detector	3000 cps
counting rate inside ^{36}Cl gates	0.008 cps
total rejection 900,000/0.008	10^8

*) measured with a 30x attenuator in place

Figure captions:

Fig 1: Charge state distribution of a ^{36}S beam (80 MeV incident energy) at the exit of the Enge split-pole magnetic spectrograph as a function of gas pressure: a) with 0.6 torr, b) with 1.8 torr and c) with 4.0 torr of nitrogen. The intensity (in arbitrary units) is plotted versus the position (channel numbers) of the particles in the split-anode gas ionization detector. At 4 torr all charge states have converged towards the average charge state.

Fig 2: Two-dimensional spectrum (ΔE3 versus position) of a high-level ^{36}Cl sample ($^{36}Cl/Cl=33x10^{-12}$). ^{36}Cl can be identified between peaks of ^{35}Cl and ^{37}Cl. The $^{35,37}Cl$ ions are transmitted through the system because of lack of electrostatic analysis on the spectrograph beam line. ^{36}Cl is spacially very well separated from the isobaric interference of ^{36}S. Peak shapes are distorted by he grid (see text).

Fig 3: Position spectrum of the high-level ^{36}Cl sample of Fig 2. The ratio of FWHM to separation between the ^{36}Cl and ^{36}S maxima is 0.35. Peak shapes are distorted by the grid (see text).

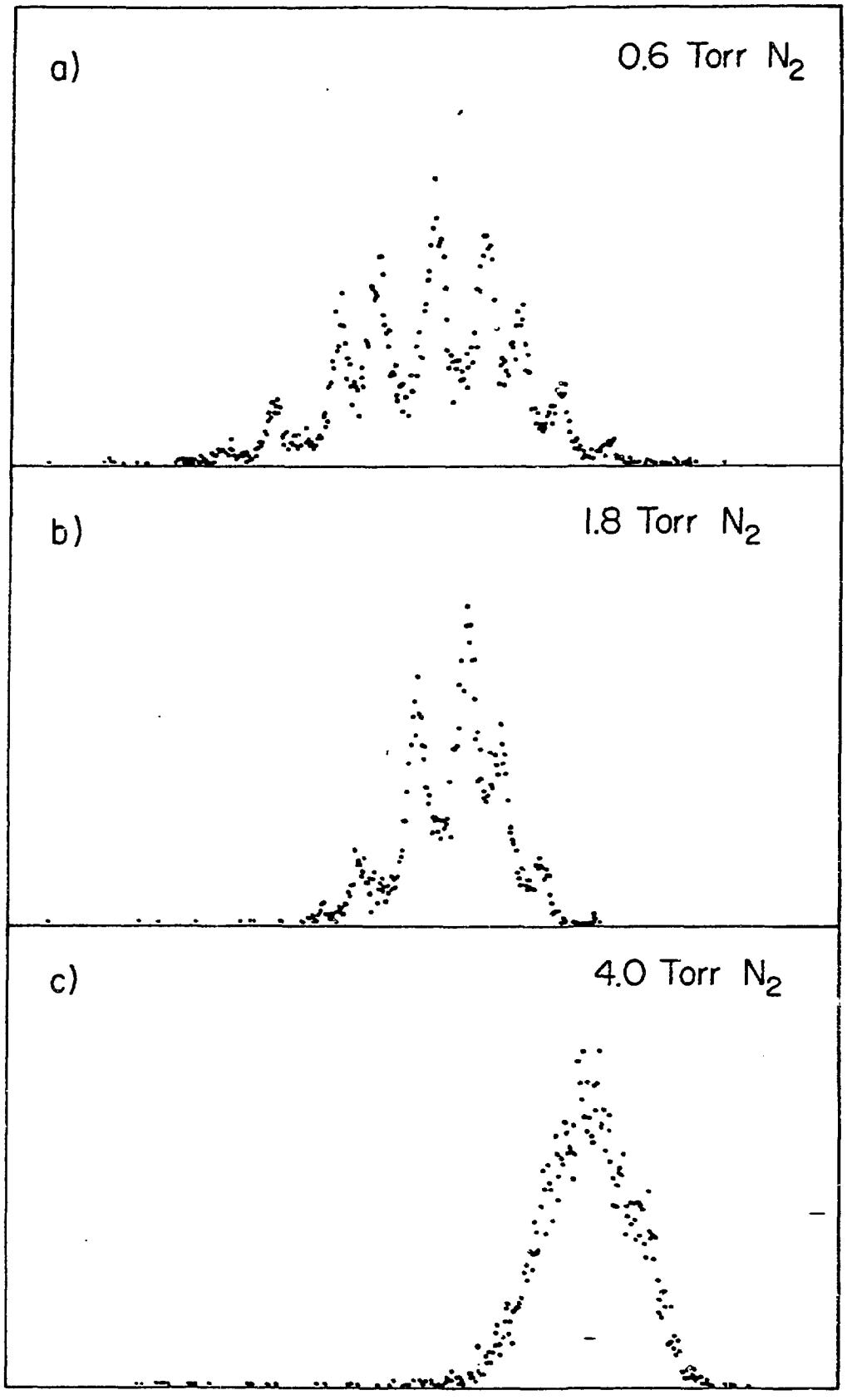

Fig 1

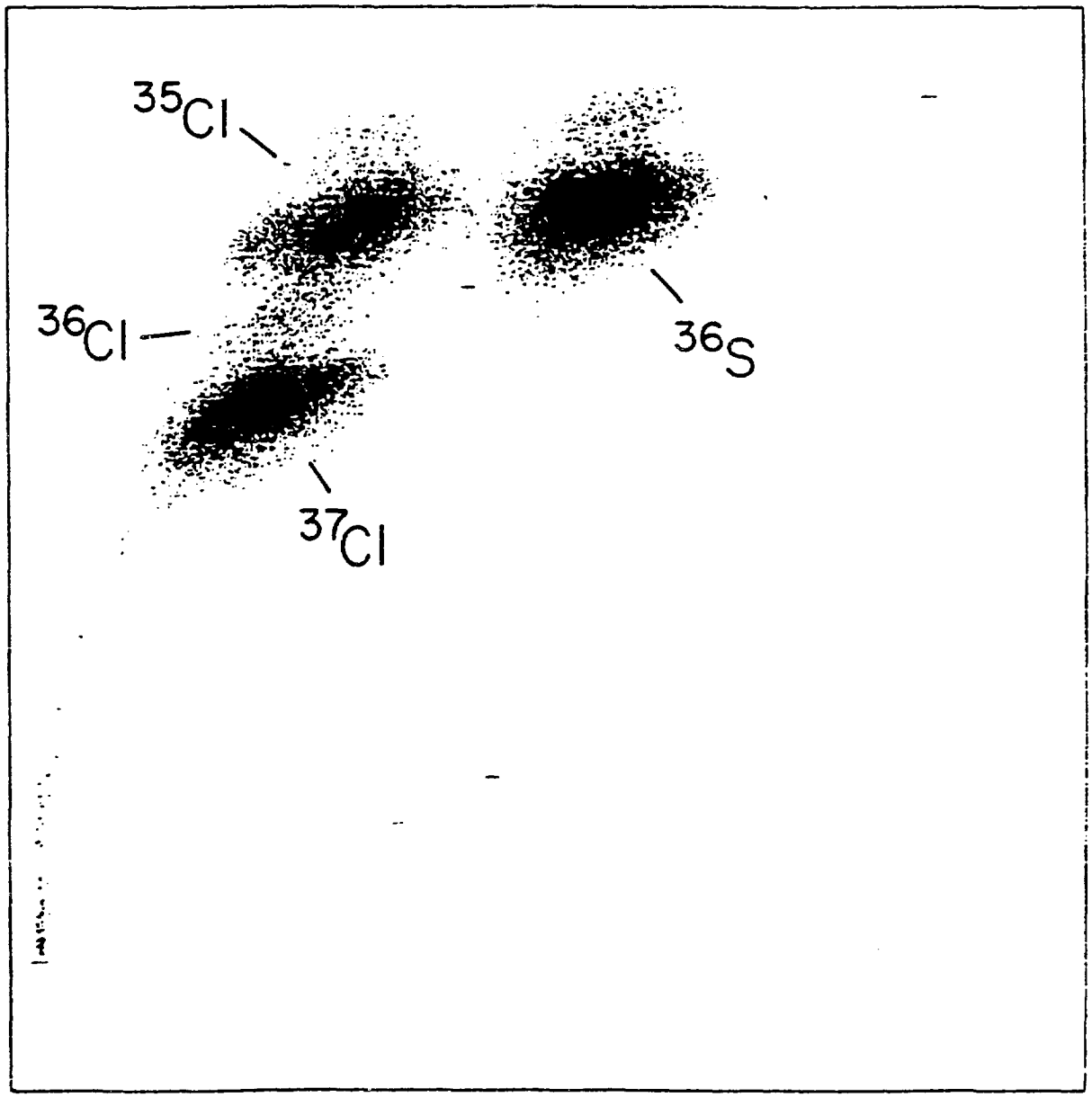

Fig 2

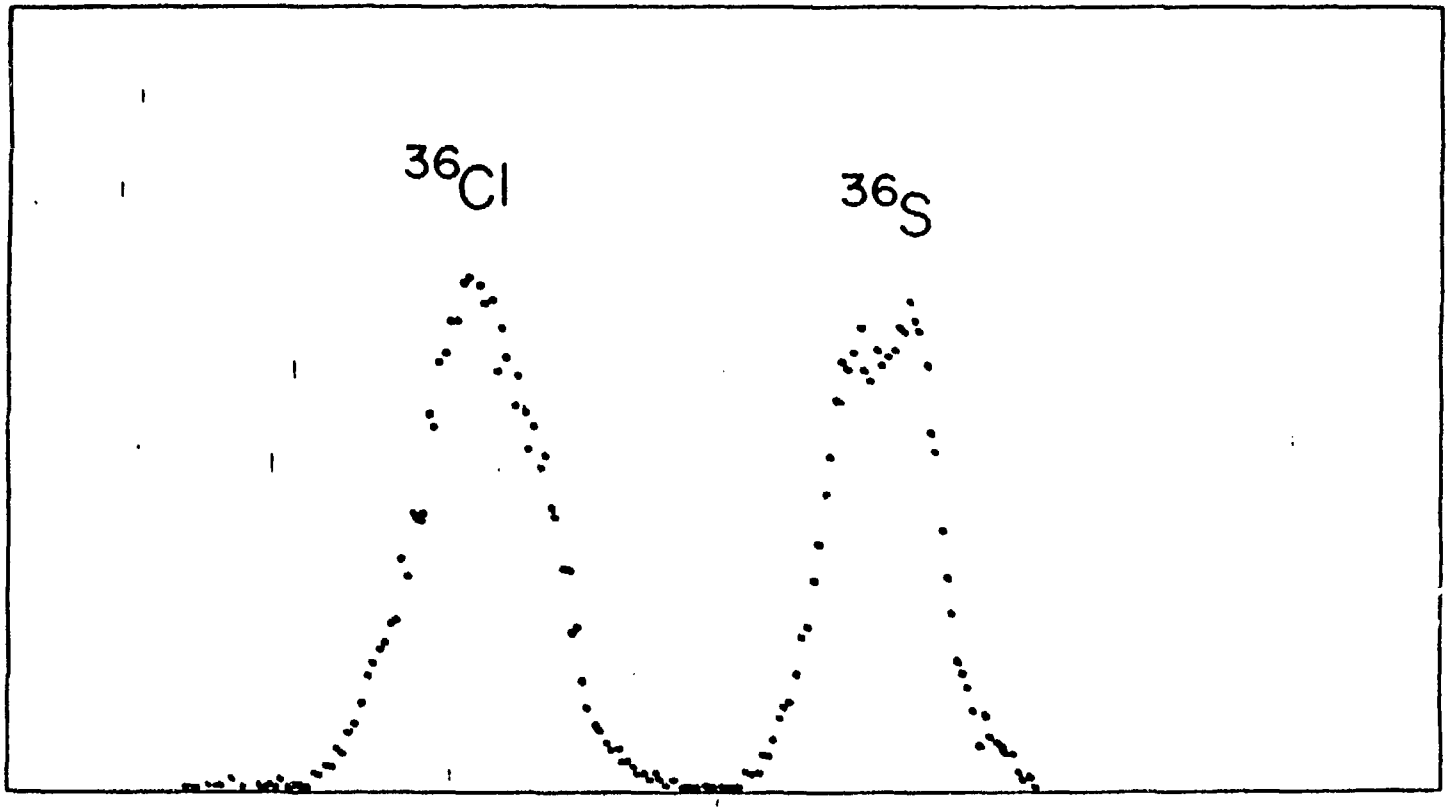

Fig 3