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DETECTION OF THE ^{36}Cl RADIOISOTOPE AT THE REHOVOT 14UD PELLETRON ACCELERATOR

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ABSTRACT. A program of accelerator mass spectrometry has been started at the Rehovot 14UD Pelletron Accelerator Laboratory. Part of the initial emphasis has been directed to the detection of the ^{36}Cl radioisotope. We report here on the present status of our work and describe our experimental system. Preliminary results are presented, showing that $^{36}Cl/Cl$ concentrations ranging down to $1x10^{-14}$ could be measured with our system.

INTRODUCTION

A program of experiments in accelerator mass spectrometry was started at the Rehovot 14UD Pelletron Accelerator Laboratory. The high-voltage terminal available at the 14UD Pelletron and its good stability properties are important features which are favorable for detection of heavy radioisotopes. Part of the initial emphasis in our program was therefore directed to the detection of the ^{36}Cl radioisotope. Tandem-accelerator mass spectrometry has indeed been demonstrated to provide a unique method for measuring extremely low ^{36}Cl concentrations (Elmore et al, 1979). The method has also been successfully applied to problems in hydrology and geology (Bentley and Davis, 1981). We report here on the present status of our work and describe the experimental system used in our measurements. Preliminary results on ^{36}Cl detection are presented.

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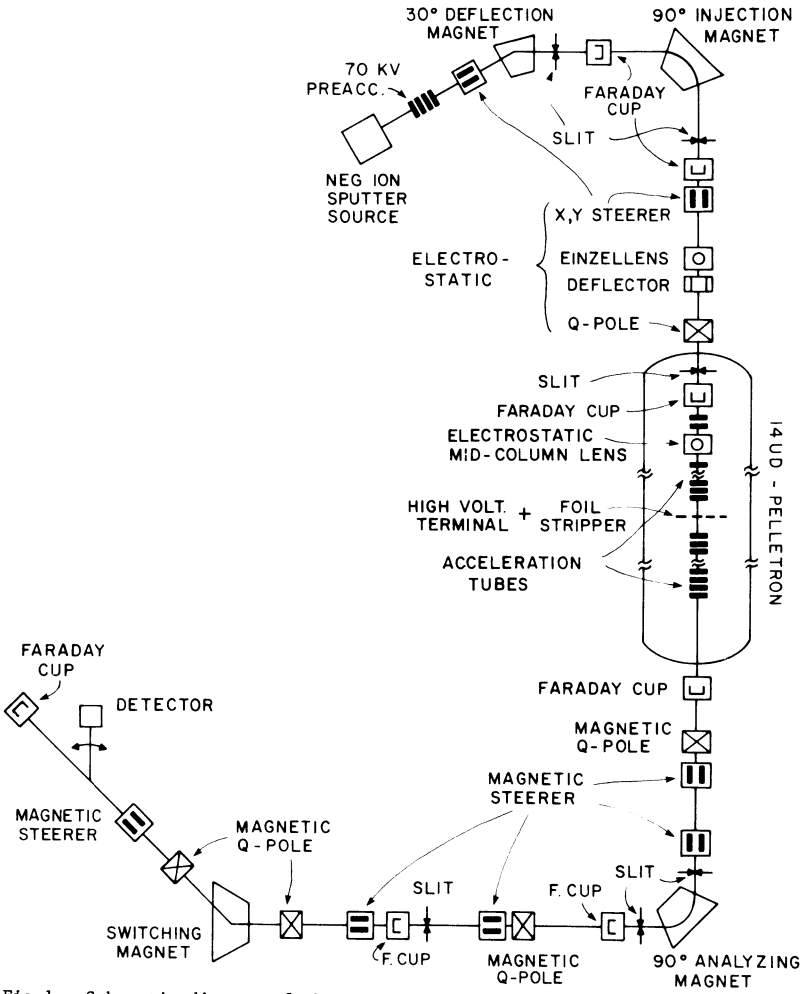

Fig 1. Schematic diagram of the accelerator set-up. The 30° magnetic deflection after the ion source, and the switching magnetic deflection, lie in a horizontal plane; the deflection of the 90° injection and analyzing magnets are in vertical planes. All other ion-optical elements of the LE(HE) sections are electrostatic (magnetic). The Faraday cup at the end of the beam line is used for the ^{37}Cl beam current measurements; the detector (fig 2) can be rotated into the beam for the ^{36}Cl counting.

EXPERIMENTAL METHOD

ACCELERATOR SYSTEM. A schematic diagram of the Rehovot 14UD Pelletron Tandem Accelerator Facility is shown in figure 1. The ion source used in our experiments was a General Ionex Hiconex 834 sputter source operated with the Cs^+ beam in the reflected mode. Twelve positions for sputter samples are available. The injection of the negative ion beam is achieved by two magnets with 30° and 90° deflection angles. All other ion-

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optical elements of the low-energy (LE) transport system (fig 1) are electrostatic. A thin C foil was used to strip the negative ions at the terminal. The vacuum in the acceleration tubes is normally in the region of 10^{-8} Torr. The high-energy (HE) ion-optical system, entirely magnetic, transports the beam after a 90° momentum analysis from the accelerator tower to the target room. A terminal voltage stabilization system (Ben-Zvi *et al*, 1981), based on a Generating Voltmeter, was built in our laboratory to stabilize the terminal voltage during the radioisotope measurement, when the slit system cannot be used. The GVM system stabilizes the terminal voltage within ±3kV at any value in the working range. All other elements of the accelerator used in our measurements were existing equipment routinely operating at the laboratory.

SAMPLE PREPARATION. Samples enriched in ^{36}Cl for testing and calibration purposes and several samples of natural origin were used in the ion source. The ^{36}Cl-enriched samples were produced by neutron irradiation of NaCl at the Soreq Nuclear Center and the Argonne CP5 Reactor. The integrated neutron flux was monitored by measuring the 15-hour activity of ^{24}Na simultaneously produced in the irradiation. The $^{36}Cl/Cl$ concentration can then be estimated from the known $^{35}Cl(n,\gamma)$ ^{36}Cl cross section. Great care was taken in a chemical procedure to reduce the level of sulphur impurity in the material, the ^{36}S stable isobar being a major source of background in the identification of ^{36}Cl (see below). The NaCl samples were first dissolved in triple distilled water. $Ba(NO_3)_2$ and $Cu(NO_3)_2$ were used to precipitate respectively SO_4^{--} and S^{--} ions. After filtration, AgCl was precipitated by $AgNO_3$ and further purified by multiple dissolution in concentrated NH_4OH and reprecipitation by HNO_3. The final AgCl precipitate was dried in darkness and pressed directly into a 4mm diameter hole in the sputter sample holder. The use of holders made of 99.99% Al was shown to reduce the ^{36}S level even further.

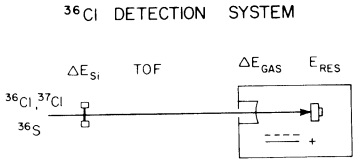

Fig 2. Schematic diagram of the detection system used for ^{36}Cl detection. Differential energy-loss signals ΔE_{Si} and ΔE_{GAS} are produced respectively by a Si detector and a gas-ionization chamber filled with isobutane at 30 Torr. The residual energy E_{RES} of the ions is measured in the last Si detector. The time-of-flight TOF of ions between the two Si detectors, ca 40cm apart, was measured.

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TUNING PROCEDURE. The complete ion-optical system of the accelerator from the ion source to the final Faraday cup was set up with the $^{37}Cl^{+}$ stable isotope beam at a terminal voltage of 10.22 MV. The 10^{+} charge state was selected to obtain sufficiently high energy for the identification system. Charge currents measured in the final Faraday cup ranged from 40 to 80 nA. After tuning, the terminal voltage was raised to 10.50 MV corresponding to a $^{36}Cl^{10+}$ beam of equal magnetic rigidity, and the source magnets (30° deflection and 90° injection) were adjusted for M=36. The detector was rotated into the beam and fine tuning of the terminal voltage was then achieved by maximizing the counting rate of the residual ^{36}S beam in the detector. This counting rate, typically of 100 to 500 sec^{-1}, was monitored during the ^{36}Cl counting time to check the stability of the system.

DETECTION SYSTEM. The detection system is designed to count the ^{36}Cl accelerated ions, while discriminating efficiently against residual ^{36}S ions and other parasitic beams degenerate in magnetic rigidity with ^{36}Cl and thus transported identically to the detector. The system, illustrated in figure 2, is a telescope composed of a Si surface-barrier detector, 13μm thick, a gas-ionization chamber (Fowler and Jared, 1975), and a 300mm^2 Si detector. The ΔE_{Si} signal (fig 3) provided by the first detector measures the differential stopping power of the ions and gives excellent discrimination between Cl and S ions. Lighter ions from parasitic beams (not shown in the figure) are easily separated. The gas chamber, filled with isobutane at 30 Torr, was used both to provide an independent energy-loss signal ΔE_{GAS} and to act as a very homogeneous passive

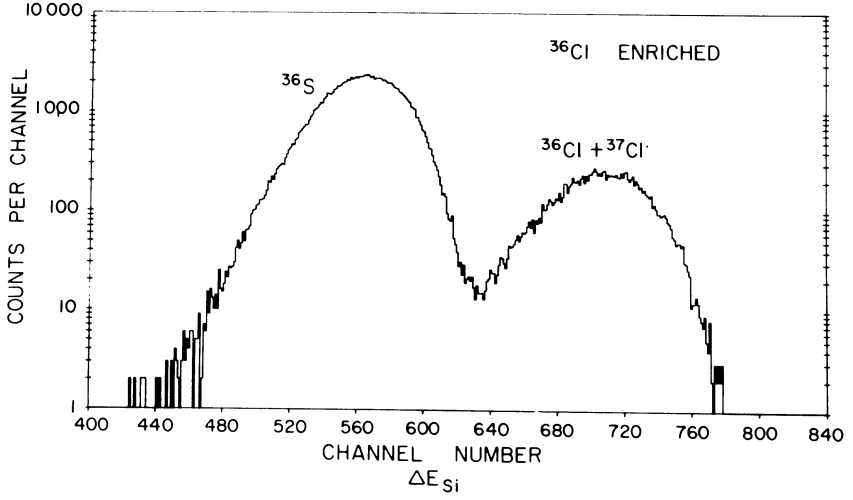

Fig 3. Spectrum of the ΔE_{Si} signal (fig 2) for S and Cl ions

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energy absorber. The spectrum shown in figure 4, obtained by adding the ΔE_{Si} signal and the residual energy E_{RES} of the last Si detector, reflects the different energy losses experienced by ^{36}Cl and ^{36}S ions in the gas: indeed, it should be noted that these ions have equal energies before entering the detection system and the separation observed in figure 4 results only from the difference in the energies absorbed in the gas. The ^{37}Cl ions, identified in figure 4, result from a tail of $^{37}Cl^-$ ions injected by the source magnets and which, by means of charge-exchange processes in the residual gas along the accelerating tube, are accelerated to a magnetic rigidity equal to that of ^{36}Cl. They are separated from ^{36}Cl by their

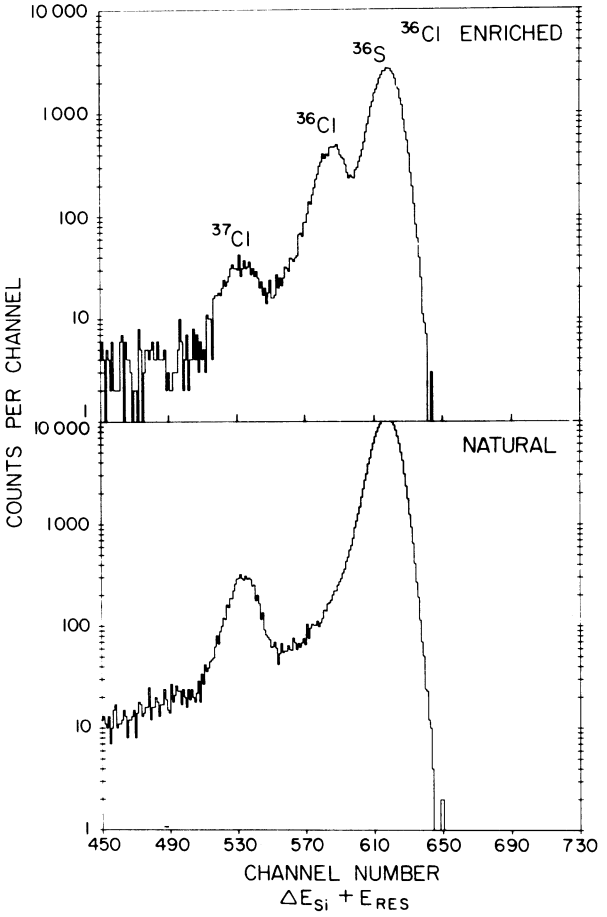

Fig 4. Spectra of the summed energies $\Delta E_{Si} + E_{RES}$, deposited in the first and last Si detectors (fig 2) for a ^{36}Cl-enriched sample and a sample of natural origin. The summation was done through software on-line treatment of the ΔE_{Si} and E_{RES} information by a PDP-11 data acquisition computer.

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difference in energy, enhanced in the spectrum of figure 4 by a slightly different energy loss in the gas. A time-of-flight signal TOF was extracted from the first and last Si detectors to obtain an independent mass identification, but this gave only marginal additional information in the present measurements. Figure 5 shows a final spectrum of identified ^{36}Cl ions obtained by filtering the events through software gates on the ΔE_{Si}, ΔE_{GAS} and TOF signals.

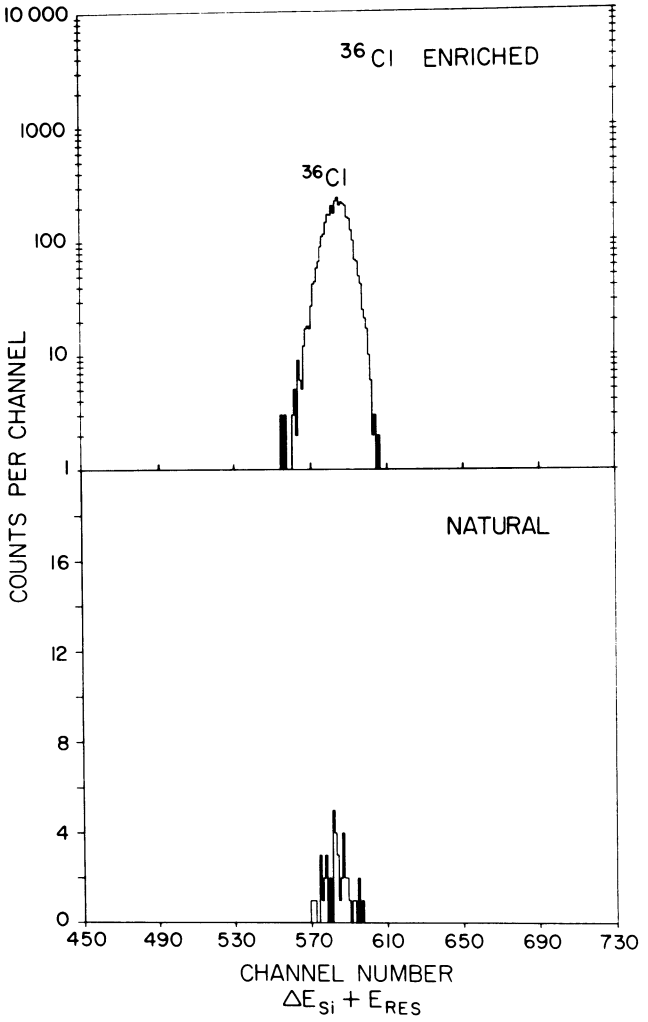

Fig 5. Spectra of summed energies $\Delta E_{Si} + E_{RES}$ for events filtered through software gates on the ΔE_{Si}, ΔE_{GAS} and TOF signals corresponding to ^{36}Cl for a ^{36}Cl-enriched sample and a sample of natural origin

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PRESENT STATUS

The overall stability of the accelerator and the beam transport elements during the radioisotope counting was found to be adequate. Also, it should be emphasized that the switching between ^{37}Cl and ^{36}Cl beams could be accomplished quickly and reliably by changing only the fields of the source magnets and the charging conditions of the accelerator. Figures 4 and 5 show spectra obtained using a ^{36}Cl-enriched sample and a sample of natural origin. The low intensity of the parasitic beams such as ^{37}Cl (see fig 4) and lighter ions is attributed to the high vacuum in the accelerator tubes and to a good mass resolution of the injection magnets. Losses resulting from angular straggling of ions in the ΔE_{Si} and gas detectors were measured at ca 10%. Table 1 show our results for enriched and natural samples. The values indicate that measurements of concentrations of the order of $1x10^{-14}$ could be attained with the present system. We have not, up to now, assessed the importance of systematic errors in the measurements such as isotopic fractionation effects due to stripping efficiency, machine ion-optics, or detection efficiency. Consequently, all $^{36}Cl/Cl$ concentrations given in Table 1 have been measured relative to the G9 enriched sample. The level of the background and possible cross-contamination effects between source samples have yet to be established for our system.

TABLE 1. Preliminary results of $^{36}Cl/Cl$ concentration measurements

Sample	Counting time (sec)	$^{37}Cl^{10+}$ stable isotope charge current (nA)	^{36}Cl counts	$(^{36}Cl/Cl)^{*}_{meas}$	$(^{36}Cl/Cl)_{expected}$
G9 (Reactor enriched)	260	52	12100	$1.45x10^{-9}$*	$(1.45\pm0.10)x10^{-9}$
E11 (Reactor enriched)	600	36	124	$(1.0\pm0.4)x10^{-11}$	$(1.49\pm0.13)x10^{-11}$
S2 (Natural)	1530	80	80	$(5\pm2)x10^{-13}$	-
Z2 (Natural)	3600	41	21	$(1\pm0.5)x10^{-13}$	-
BRC2 (Natural)	3000	22	10	$(8\pm3)x10^{-14}$	-

*Normalized relatively to a value $1.45x10^{-9}$ for the G9 enriched sample. The values are not corrected for fractionation effects. The errors given include statistical and other random errors, estimated from reproducibility of the values obtained in the small number of available measurements.

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CONCLUSION

The present results on the sensitivity of our system for ^{36}Cl detection are encouraging and may allow for concentration measurements of cosmogenic ^{36}Cl in nature. Interesting problems related to geology and hydrology are under study and could be attacked with the present system.

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