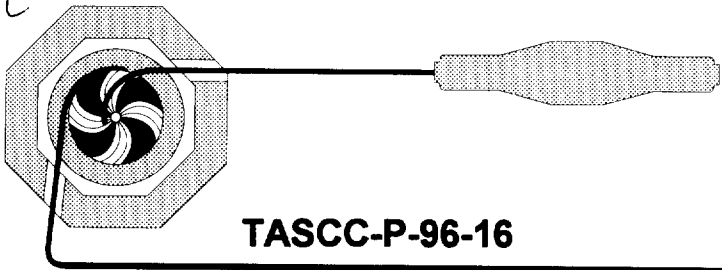EE

THE REDUCTION OF SAMPLE MEMORY EFFECTS IN THE CHALK RIVER AMS ION SOURCE

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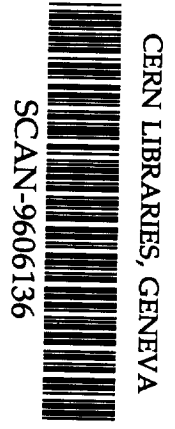
The Reduction Of Sample Memory Effects
In The Chalk River AMS Ion Source

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Abstract

The mechanism underlying Cl and I sample-to-sample interference in the new Chalk River AMS

ion source has been studied. The distribution of sputtered material and its degree of migration

was measured with the radioactive tracer, ^{82}Br. The temperature dependence of the surface

constituents was measured with the elastic recoil detection (ERD) technique and the effect of

sample geometry and ion-source cleaning was studied with elevated (5×10^{-10}) ^{36}Cl/Cl and ^{129}I/I

samples. These measurements indicate that a hot (>350°C) aperture plate ahead of the sample

can prevent the sputtering of contaminated regions near the sample. The plate itself remains

relatively free of Cl or I itself since these elements or their Cs-gettered compounds are desorbed

at this temperature. A small, fixed quantity of Cl or I on this surface is observed, which if

sputtered by Cs^+ ions, may contribute to ion source memory. Relative sample-to-sample

interference is about 10^{-3} after 20 minutes or 10^{-4} after 60 minutes.

1. INTRODUCTION

The AMS program at Chalk River has recently concentrated on $^{36}Cl/Cl$ and $^{129}I/I$ ratio determinations in many types of samples including ground water, precipitation, soil, air, vegetation, seeds, teeth, uranium ore, nuclear fuel and reactor components. The isotopic ratios observed in these samples have varied by several orders of magnitude often exceeding the "dynamic range" of our AMS instrumentation. This limitation arises because less than 10% of the sputtered material is extracted as a beam, the remaining material being distributed throughout the ion source. The distributed sample material then contributes to the ion beam extracted from subsequent samples unless the source is dismantled and cleaned. This effect is known as ion-source memory. Fig. 1 shows the magnitude and time dependence of ion-source memory for Cl and I at Chalk River.

The mechanism of ion source memory is very element specific. In the case of halogen elements their high vapour pressure could lead to rather clean surfaces or perhaps to enhanced migration. They would be expected to readily getter with the sputtering element, Cs, also present in the ion source. Rationalization of these phenomena is further complicated by the large variation in temperature, up to 1200°C, of the ion source components. The purpose of this paper is to describe recent tracer and surface characterization studies with Chalk River's new AMS ion source[1] directed toward better understanding and control of chlorine[2] and iodine memory.

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2. DESCRIPTION OF THE ION SOURCE

The new ion source, illustrated in Fig. 2, has substantially better cooling than its predecessor. Peripheral regions of the source are at ambient water temperature and the internal support structures are fabricated from aluminum that is characterized by high thermal conductivity.

One unique feature of the Chalk River ion source is its hot aperture plate. This thin (0.25 mm)Ta plate is sandwiched between zirconia ball bearings and mica and radiatively heated by the 1200°C ionizer to 400°C. As shown in Fig. 2, other regions of the ion source are relatively cooler with temperature not exceeding 100°C.

The extracted ^{35}Cl and ^{127}I currents are typically 5 and 2 μA respectively for about 15 μA of 8 keV Cs^+ ions. The Cs^+ current is measured on a segmented Faraday cup located behind the sample. When the sample is removed, a profile of the Cs^+ beam is registered on this cup and read-out through a 4-channel CAMAC-based electrometer constructed at Chalk River. The anode in this source differs from our earlier ion source in that the Cs vapour now enters the ionizer region through six holes equally spaced around the perimeter of the Ta ionizer and that a Cs focus lens has been added to control the focal length of the Cs optics.

The vacuum at the sample (cathode) is measured with a hot-filament ionization gauge to be 10^{-6} Torr; the pumping speed is limited by the small apertures surrounding the aperture plate and ionizer assembly.

3. SURFACE DISTRIBUTION AND MIGRATION OF SPUTTERED MATERIAL

The radionuclide ^{82}Br ($t\frac{1}{2} \simeq$ 1.47 d) is an ideal tracer for determining the surface distribution of sputtered halogen salts like AgCl or AgI in an ion source. In the form of LiBr, it is easily activated in Chalk River's NRU reactor. Its half life permits day-long measurements but it is short enough that the ion-source is free of contamination after a few days. The ^{82}Br can be easily purified by precipitating AgBr out of solution thereby preventing the contamination of the source by unwanted longer-lived activities that are present in the activated salt.

To determine the distribution of sputtered material, the Ag^{82}Br was diluted with Ag powder and packed into a Cu sample holder. Its initial β-decay rate was about 120 kBq. The sample was sputtered for four hours during which 50% of the sample was consumed. This was determined by γ-ray spectroscopy of the sample before and after sputtering.

Before disassembly of the ion source, the degree of Br migration in the ion source, under operational conditions, was determined by inserting an unused, empty sample holder into the source and sputtering it for half an hour. It was then removed and γ assayed for ^{82}Br. It registered a ^{82}Br decay rate equal to 0.08% of the initial ^{82}Br intensity.

Another empty sample holder was inserted for half an hour but not sputtered (cathode bias off), then removed and γ assayed. No ^{82}Br was observed, an upper limit of about 0.005% being set by the background events in the spectrum. This procedure was repeated during the next four hours while the ion source cooled in preparation for disassembly and β assaying. Again no ^{82}Br was observed in this sample holder.

After disassembly, the components were β assayed with a 50 mm^2, 2000 μm thick Si surface-barrier detector. As before[2], the activity was observed to be quite localized at (see Fig.

2) the beam stop, the region on the sample transfer rod immediately adjacent to the sample holder, and the heat shield. The aperture plate, found to be heavily contaminated in earlier experiments when the plate was relatively cool, had small but perceptible amounts of ^{82}Br contamination, amounting to about 6% of that found on the heat shield and 0.8% of that on the transfer rod.

4.   SURFACE CHARACTERIZATION IN AN OPERATING SPUTTER ION SOURCE

In the Elastic Recoil Detection (ERD)[2] measurements, a high-energy (350 MeV) Au beam from the TASCC facility was used to forward Rutherford-scatter surface atoms off an ion-source surface into a ΔE-E detector. These data were recorded event-by-event on magnetic tape along with time-of-occurrence from the beginning of the run, substrate temperature and ion-source operating conditions for later analysis. The ΔE signal provides elemental identification while the total energy (ΔE+E) E defines the depth from which the atom is scattered[3].

The Au beam was scattered off a temperature-controlled Cu heat shield in a functioning sputter ion source that contained either a AgCl or AgI sample mounted on a Cu sample holder. Unlike the earlier studies[2], Rb^+ instead of Cs^+ ions were used to sputter the sample in order to prevent Cs from interfering in the identification of I. As with Cs and Cl, for substrate temperatures below 200°, surface contamination of Rb and I was observed to increase with time, but they were desorbed above 350°C (see Fig. 3). The surface abundance of other elements such as Ag or Cu also increased during sputtering but they were not affected by substrate temperatures as high as 600°C.

As observed with the [82]Br tracer, a residual amount of Rb and I, as well as K, Na and Cl, remained fixed on the substrate and could not be desorbed at temperatures up to 600°C.

5. TIME DEPENDENCE OF [36]Cl AND [129]I

The magnitude and time-dependence of ion source memory for [36]Cl and [129]I were investigated for full, and half-full, double-depth sample holders. These deeper, partially-filled holders confine the sputtered particles to travel in a narrow cone that might limit the distribution of contamination in the source. Samples of AgCl and AgI containing 5×10^{-10} [36]Cl/Cl or [129]I/I were sputtered for either five or ten minutes and quickly replaced with either Si, AgCl or AgI blanks. The [36]Cl or [129]I counting intensity from these blanks was followed for up to two hours. In one case the effect of thoroughly scrubbing the transfer rod with a NaOH solution about thirty minutes after removal of the contaminating sample was studied. The results are shown in Fig. 4, along with previous data.

The results indicate that:

(a)   cleaning the highly contaminated region of the transfer rod (identified with the [82]Br tracer), did not significantly reduce the [36]Cl memory;

(b)   the rate of decay was not affected by the duration for which the contaminated samples were sputtered (not shown in Fig. 4);

(c)   the half-full and full cones behave identically;

(d)   the time-dependence of [129]I decay was similar to that observed with the full [36]Cl cone; and

(e)   the decay of [36]Cl with time in this ion source is not as rapid as it was for its predecessor.

6.   SUMMARY

The decay-time of ion-source memory for Cl and I samples has been shown to be comparable (see Fig. 3). There are two highly-contaminated regions of the ion source (see Fig. 2) but the aperture plate effectively screens the region adjacent to the sample, since the cleaning of the transfer rod had no appreciable effect on ^{36}Cl memory following contamination with the 5×10^{-10} ^{36}Cl/Cl sample. Migration of sputtered material, as observed with ^{82}Br, is <0.1%. Both ERD and ^{82}Br studies indicate that small amounts of sputtered Cl, Br or I remain fixed to the heated aperture plate. Limiting the distribution of sputtered material with partially-filled double-depth sample holders did not affect the decay-time of ion-source memory.

The heated aperture plate has reduced ion-source memory by about a factor of ten from earlier values obtained with a cool aperture plate but the performance of the new ion source in this regard is not as good as the ion source it has replaced. This may be due to the larger Cs^+ halo that strikes the contaminated aperture plate. Efforts are underway to reduce the size of this halo through structural changes that eliminate hot peripheral regions of the ionizer assembly. Lastly, Fig. 4 shows the lowest-background counting rates observed during recent runs. For the last three runs shown (post 3/95) the ion source was equipped with a heated aperture plate. These runs are characterized by a lower ^{36}Cl background rate than the pre 3/95 runs. The ^{129}I background rate remains unaltered. This latter point is not understood considering the similarities in behaviour of I and Cl observed in these measurements.

REFERENCES

1. See accompanying paper on *"The New Chalk River AMS Ion Source, Sample Changer and External Sample Magazine"*.

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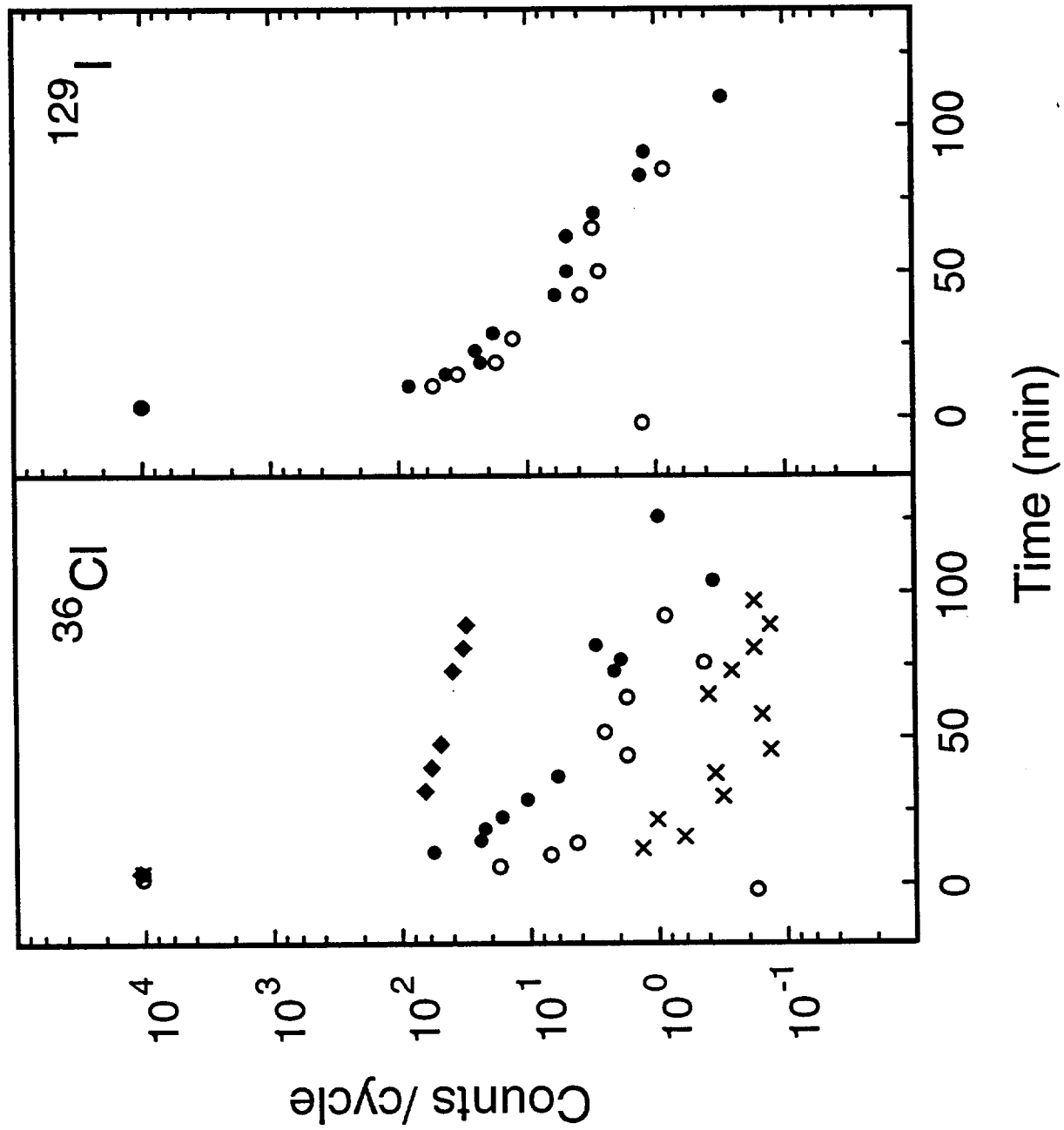
FIGURE CAPTIONS

Figure 1: The counting intensity for ^{36}Cl and ^{129}I versus time from blank samples after a high-level contaminating sample (~5 x 10^{10} isotope ratio) is sputtered. The ion-source memory observed prior to these investigations for ^{36}Cl is indicated by diamonds. In this case the aperture plate of the old ion source was relatively cool (<200°). The x represent data for a hot aperture (>350°C) in the old ion source. Circles represent data for the new AMS ion source[1]. Open circles represent conditions where the sample-holder well, for the contaminating sample was double-depth but half-full. The background before contamination is indicated in the lower left of each panel, at time zero, by open circles and corresponds to 10^{-15} and 10^{-14} for $^{36}Cl/Cl$ and $^{129}I/I$ respectively. Thirty minutes into the ^{36}Cl measurement shown with filled circles the sample transfer rod was thoroughly cleaned and measurements were resumed about forty minutes later. The effect of cleaning on ion-source memory was negligible.

Figure 2: The new Chalk River AMS ion source. Circled numbers refer to component temperatures (°C). Uncircled numbers refer to the degree of ^{82}Br contamination present on the surfaces (number of β events detected per 100 s in a 50 mm^2 surface barrier detector at the surface).
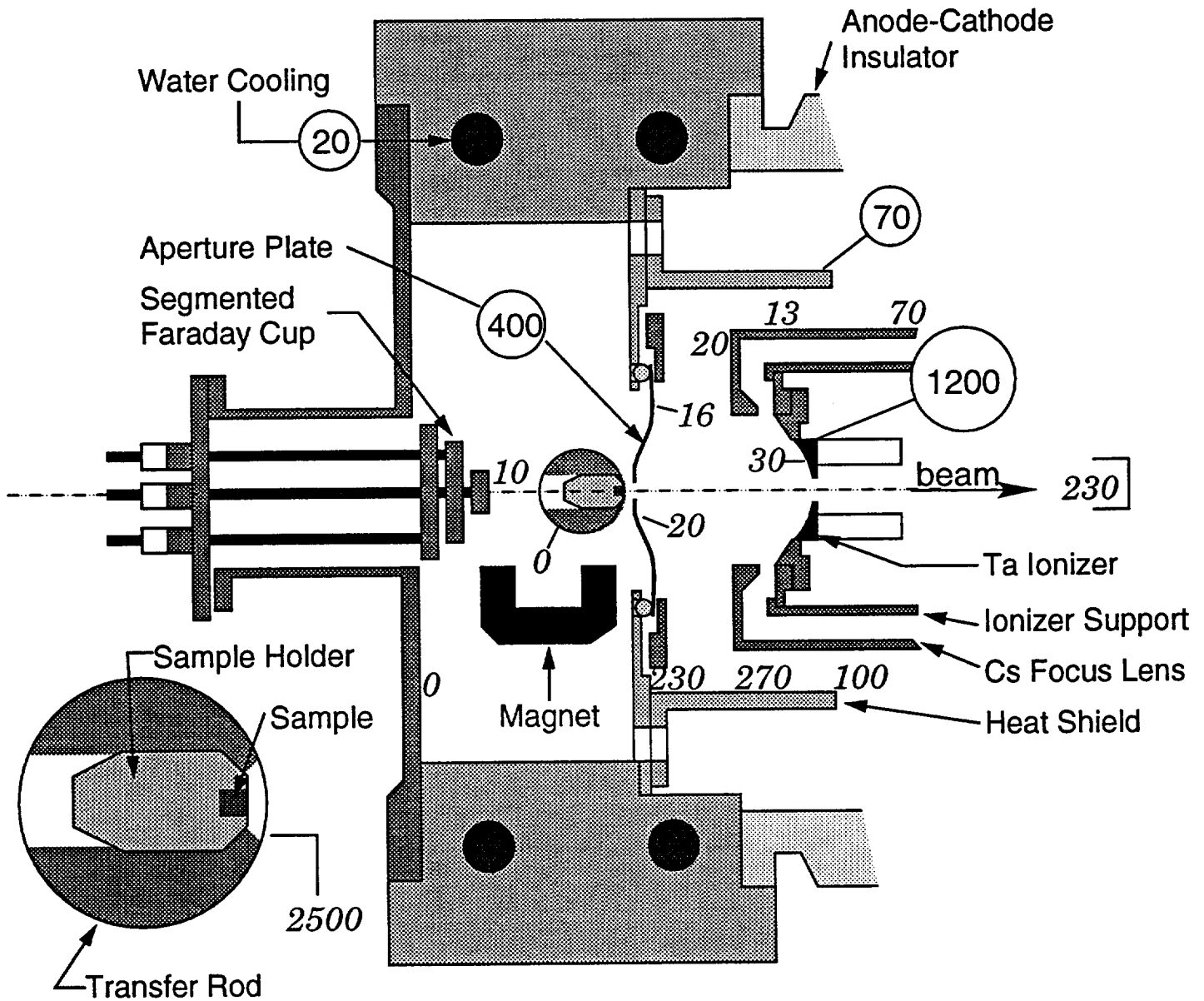
Figure 3: The desorption of I and Rb as a contaminated Cu surface is heated to 600 °C.
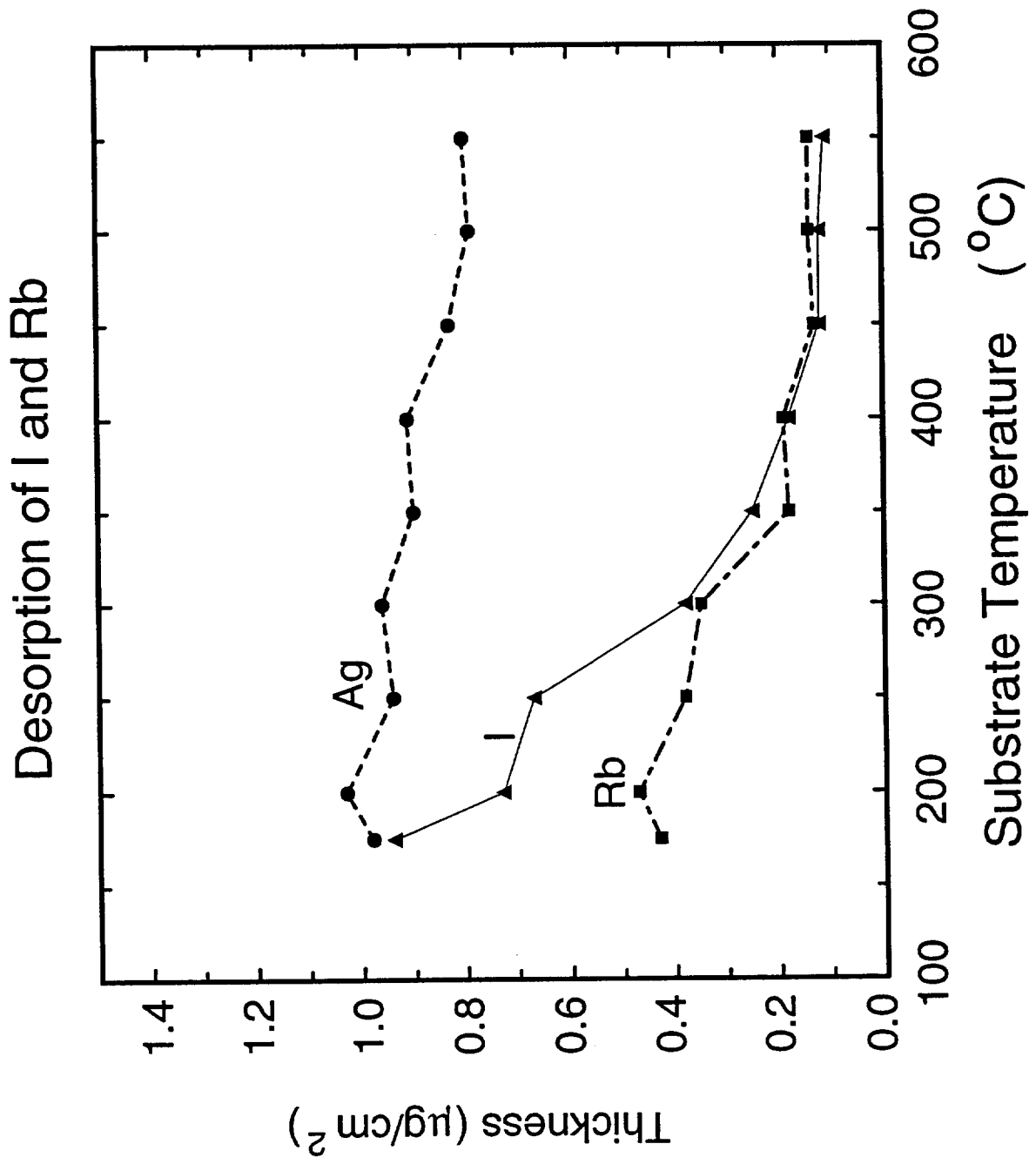
Figure 4: The lowest background counting-rate per run observed from a blank sample for
^{36}Cl *an* ^{129}I *measurements dating back to 9/93. The* ^{36}Cl *data point for run 6/95 is an upper limit since no background events were observed over a 90-cycle (~80 minutes) period. The measurements, post 3/95, have used a heated aperture plate. The last two runs shown involved the new AMS ion source. Improvements in* ^{36}Cl *background are clearly visible but the* ^{129}I *background has not been reduced. This effect is not understood.*

Ion Source Memory

^{129}I

^{36}Cl

Time (min)

Counts /cycle

Fig 1

The Chalk River AMS Ion Source

Anode-Cathode Insulator

Water Cooling
(20)

(70)

Aperture Plate

Segmented
Faraday Cup

(400)

13   70

20

(1200)

16

30

beam   230

10

Ta Ionizer

20

0

Ionizer Support

0

Sample Holder

Cs Focus Lens

Sample

230   270   100

Heat Shield

0

Magnet

2500

Transfer Rod

Fig 2

Fig 3

Desorption of I and Rb

Substrate Temperature (°C)

Thickness (μg/cm²)

Ag

I

Rb

Lowest Background per Run

Fig. 4