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TO A CS SPUTTER ION SOURCE FOR THE  
ACCELERATION OF RADIOACTIVE IONS WITH  
THE CHALK RIVER MP TANDEM ACCELERATOR***

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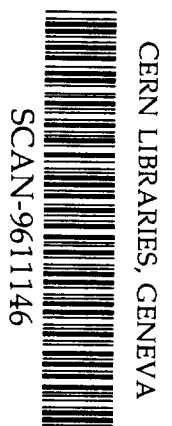
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***Coupling of a He-jet Transfer System to a  
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with the Chalk River MP Tandem Accelerator***

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**Abstract**

The concept of coupling a He-jet transfer system to the negative-ion sputter source of the Tandem accelerator at Chalk River has been tested. Negative-ion beams of the radioactive isotopes:  $^{34m}\text{Cl}$ ,  $^{56}\text{Co}$ ,  $^{60}\text{Cu}$ , and  $^{103}\text{Ag}$  were extracted from a  $\text{Cs}^+$  sputter ion source by sputtering cathodes that were covered with radioactivity-laden aerosol deposits from a gas-jet transfer system. Efficiencies of 1 to 25% were observed with about 50% of the deposit sputtered in less than one minute. A fast ion-source sample changer and a pneumatically-positioned target chamber have been installed at the MP Tandem accelerator. They are coupled by a He-jet transfer system, permitting the alternate acceleration of stable and radioactive ions.

## 1. INTRODUCTION

At Chalk River we are investigating the merits of coupling a gas-jet transfer system to the Accelerator Mass Spectrometry (AMS) sputter ion source in order to prepare, via stringent AMS selection techniques, an isotopically-pure, accelerated, radioactive ion flux that can be counted, identified and accumulated into samples for subsequent  $\beta$ -decay studies. This would lend itself to precise studies of  $0^+ \rightarrow 0^+$  superallowed  $\beta$  emitters, for which the degree of sample purity has often been of major concern in half-life and branching-ratio measurements. It would also provide experimental versatility since the selected accelerated ions could be directed to one of several existing large detection systems.

In the following we describe details that cover the production and acceleration of radioactive beams with the MP Tandem accelerator and measurements that provide evidence that a  $\text{Cs}^+$  sputter ion-source can be effectively coupled by a He-jet gas transfer system to a target chamber.

## 2. CONCEPT

The concept for the production, acceleration and detection of radioactive ions is analogous to that employed in Accelerator Mass Spectrometry (AMS)<sup>[1]</sup> where an isotopic ratio for an element such as carbon ( $^{14}\text{C}/^{12}\text{C}$ ) or chlorine ( $^{36}\text{Cl}/^{35}\text{Cl}$ ) is determined by alternately measuring the flux of a rare (often unstable and long-lived) isotope and that of the abundant isotopes. An AMS measurement involves: (i) chemically preparing from an environmental specimen a solid 1-to-10-mg sample containing the desired element; (ii) installing the sample as the cathode of a negative-ion  $\text{Cs}^+$  sputter ion source and sputtering

the sample with 10 to 100 $\mu$ A of 5 to 10 keV Cs<sup>+</sup> ions; (iii) sequentially injecting the low-energy (200 keV) mass-separated negative-ion beams into the Tandem accelerator; (iv) directing the accelerated beams of the abundant isotopes into Faraday cups for current measurements; (v) directing the flux of the rare isotope through rigidity, velocity and Z-filters in order to remove or reduce the flux of undesired background ions; and finally (vi) detecting, identifying and counting the filtered ions in a sensitive, charged-particle detection system.

At Chalk River this technique has been extended to permit the sequential acceleration of primary and secondary beams with the Tandem accelerator by installing two special devices: the first is a sample changer that permits fast exchanges of AMS sample holders in the ion source<sup>[2]</sup>; the second is a pneumatically-positioned target chamber, located at the high-energy exit of the accelerator, that is He-jet coupled to an AMS sample holder. While the target chamber is inserted, the targets are bombarded by the primary (stable) beam produced from an AMS sample holder in the ion source, and the resultant radioactivity is transported by He-jet to a second AMS sample holder outside the ion source. After a suitable irradiation/collection time, the second holder, with the gas-jet-deposited radioactivity, is exchanged with the first holder; the target chamber is also withdrawn from the beam line and the negative-ion injector optics are adjusted to select the radioactive isotope. Accelerated radioactive ions are then directed through the AMS beam-line filters to the detection apparatus. The cycling between primary and secondary modes of operation has been automated through computer control. Only the injector settings need be altered. The Tandem accelerator voltage is chosen to provide an appropriate activation energy. The secondary ion energy is not critical since it need only

be high enough to offer adequate  $\Delta E - E$  identification or Z discrimination in a gas-filled spectrometer. Table 1 summarizes the facility operating parameters for the production and acceleration of  $^{14}\text{O}$  from 14 MeV proton bombardment of nitrogen.

### 3.0 TECHNICALITIES OF THE ION SOURCE/GAS-JET COUPLING

A He-jet transfer system and a sputter ion source have many compatible characteristics. Gas transfer systems are an element-insensitive, efficient and fast method of transporting aerosol particles. When swept through a target chamber, the aerosol particles emerge laden with reaction products<sup>[3]</sup>. They can then be transported through a capillary over long distances<sup>[4]</sup> and deposited onto an AMS sample holder.

The  $\text{Cs}^+$  sputter ion sources<sup>[2,5]</sup>, commonly found at Tandem accelerator and AMS laboratories, are efficient at producing negative-ion beams from nearly any solid material. Their emittance and energy spread are well matched to a Tandem accelerator and the ion source is able to operate for long periods with a minimum of maintenance. Briefly, ion-beam formation involves sputtering (with 5 to 10 keV  $\text{Cs}^+$  ions) a caesiated cathode that is fabricated from a (conductive) material that contains the element of the desired ion. The  $\text{Cs}^+$  beam is easily formed by surface ionization on a hot anode. The efficiency of creating negative ions of the sputtered atoms is high when an atom leaves a metal surface with low work-function (caesiated cathode) at a velocity higher than the thermal velocity<sup>[6]</sup>. For many metallic elements, the efficiencies can be more than 10%<sup>[7]</sup>.

The maximum production of radioactive ions from the sputtering of NaCl aerosol deposits is achieved when the deposits are efficiently sputtered before a substantial amount of activity decays. The negative-ion emission efficiency for trace atoms in a non-metallic

matrix offers adequate beam intensity and the proportion of negatively-ionized molecular ions is minimal.

The dwell time and efficiency can be affected by factors such as the chemical stability of the aerosol deposit when subjected to heat, the diameter and depth of the deposit, the size of the  $\text{Cs}^+$  ion beam and the  $\text{Cs}^+$  sputtering rate. Shown in Fig. 1 are time profiles of  $\text{Cl}^-$  and  $\text{Cu}^-$  currents extracted from  $\text{NaCl}$  and  $\text{CuCl}_2$  aerosol deposits inserted into a Model 860 HICONEX ion source<sup>[5]</sup>, which is in regular use at Chalk River for the production of ion beams. It indicates that (1) larger  $\text{Cl}^-$  currents are extracted from heavier aerosol deposits, the integrated current being approximately proportional to the deposition time; (2) larger  $\text{Cl}^-$  currents are extracted with higher  $\text{Cs}^+$  sputtering currents; and (3) most of the deposit can be sputtered off the sample holder within ten seconds, although the absolute fraction sputtered is difficult to gauge because of the extent of the long tail in these time profiles and uncertainty in the initial size of the sample. The tail could originate in two ways:  $\text{Cl}$  recycling in a hot ion source<sup>[8]</sup> and/or implantation of some sample material into the sample holder by the  $\text{Cs}^+$  bombardment. Since both  $\text{Cu}$  and  $\text{Cl}$ , with characteristically different vapor pressures, have similar ion-current time profiles, the tail likely originates from the latter mechanism.

Absolute sputtering yield and negative ionization efficiencies were measured with radioactive aerosol deposits containing  $^{34\text{m}}\text{Cl}$ ,  $^{56}\text{Co}$ ,  $^{60}\text{Cu}$  or  $^{103}\text{Ag}$ . These isotopes were selected because their half-lives (between 32 min and 77 d) permit manual handling; each have characteristic  $\gamma$ -rays, and these elements are known to produce copious negative-ion beams. They were prepared by bombardment of  $\text{Pb}^{34}\text{S}$ ,  $^{\text{nat}}\text{Fe}$ ,  $^{\text{nat}}\text{Ni}$  or  $^{\text{nat}}\text{Pd}$  targets, respectively, with 17 MeV protons. The activities were transported by He-jet through

20 m of 2.25 mm diameter capillary tubing and deposited on ion-source sample holders. Each sample was subsequently assayed with a HPGe detection system prior to its being sputtered in the ion source and finally re-assayed after removal from the ion source. The extracted ion beam for each sample was mass-analyzed and implanted into an aluminum paddle that was also removed and assayed. The sputtering yield was calculated by comparing the decay-corrected activity on the sample holder before and after sputtering. The negative ionization efficiency was determined by comparing the decay-corrected activity on the paddle with that sputtered off the sample holder.

In one series of measurements we obtained an absolute sputtering-yield time profile by repeatedly sputtering and assaying an aerosol deposit that initially contained about 100 Bq of  $^{56}\text{Co}$ . The results are shown in Fig. 2. Like the stable  $\text{Cl}^-$  current time profiles of Fig. 1, the  $^{56}\text{Co}$  results also indicate that the sputtering yield saturates with sputtering time. The proposal that sample material is implanted into the holder is corroborated by a series of measurements that compared the amount of radioactivity remaining on both sputtered and unsputtered samples after a hot-water rinse and ultrasonic cleaning. Washed, unsputtered samples no longer carried radioactivity while sputtered and washed samples retained 50-75% of the radioactivity found on the sample holder prior to rinsing.

The measured negative ionization is shown in Table 2. Bulk negative-ion emission efficiencies were determined by Middleton<sup>[9]</sup>, who measured the integrated negative-ion current extracted from a sample prepared from a stable isotope and weighing the sample before and after several hours of sputtering. Generally the negative-ion emission efficiency of the aerosol or salt deposit agree well with the bulk efficiency determinations

and the fraction sputtered in one or two minutes is 50-90% of the aerosol deposit. This illustrates that trace and matrix atoms are identically affected by sputtering.

The diameter and intensity profile of the  $\text{Cs}^+$  spot can only be estimated by the appearance of a sputtered sample holder. A 1-mm-diameter hot spot usually exists in the center while the full diameter is about 5-6 mm. This is reasonably well matched to a 5-mm-diameter aerosol deposit made by a 2.25 mm diameter capillary, 20 m long. With a  $^{56}\text{Co}$  tracer the deposit has been radially profiled and 75% of the activity is within a 3-mm diameter.

Finally, we tested the stability of the aerosol deposit against deterioration by ion-source heating by measuring the  $^{56}\text{Co}$  activity remaining on an aerosol deposit after it was placed in a vacuum oven for several minutes. No activity was lost for temperatures below  $600^\circ\text{C}$  and insertion times of twenty minutes. Radiant heating by the Cs ionizer does not induce these temperatures at the ion-source sample location and consequently the sample must remain stable until sputtering begins.

#### 4.0 CONCLUSION

The gas-jet transfer system has been shown to provide a solid, radioactive sputtering sample, well-matched to a  $\text{Cs}^+$  sputter negative-ion source. Radioactive atoms have been produced, delivered to the ion source, ionized, accelerated and mass-analyzed with measured efficiencies ranging from 1 to 25%. With improvements in sputtering currents, ion-source dwell-times below a few seconds could be achieved. Together with the new AMS sample-changing mechanism and pneumatically positioned target chamber,



the system should be able to produce radioactive ion yields of modest intensity ( $10^3$ - $10^7$  per sample) for precise  $\beta$ -decay experiments.

**TABLE 1**

**Facility Operating Parameters for the Production of  $^{14}\text{O}$**

	<b>Beam</b>	
	<b>Primary</b>	<b>Secondary</b>
Cathode Material	TiH	NaCl aerosol deposit
Negative Ion Selected By Injector	H <sup>-</sup>	$^{14}\text{C}^-$ , $^{14}\text{O}^-$
Tandem Accelerator Volts	7 MV	7 MV
Charge State of High-Energy Ions	+1	+5
Energy of Ions	14 MeV	42 MeV

**TABLE 2**

**Measured Aerosol Depletion Rates and Negative-Ion Emission Efficiencies**

Isotope	$t_{1/2}$	Efficiency (%)		Type of Sample	Fraction Sputtered (%)	Sputtering Interval(s)
		This work	bulk <sup>†</sup>			
<sup>34m</sup> Cl	32 min	25	16	NaCl aerosol	75	180
<sup>56</sup> Co	77 d	1	1	NaCl aerosol	33	30
<sup>60</sup> Cu	24 min	1	8	NaCl aerosol	90	120
<sup>103</sup> Ag	1.1 h	1		NaCl aerosol	85	120
-----						
<sup>82</sup> Br	1.5 d	25		NaBr deposit*	28	120
					50	300
<sup>14</sup> O	71 s	1		AlN deposit**	15	90

<sup>†</sup> Bulk negative-ion efficiency measurements by Middleton[9] were determined by his sputtering stable isotopes for several hours and comparing the weight loss of the sample to the integrated extracted beam current.

\*

\* NaBr depositions were prepared by drying a small aliquot of salt solution on ion-source sample holders.

\*\* <sup>14</sup>O was prepared by 13 MeV proton bombardment of 0.5 mg of AlN powder that was pressed into a gold substrate of an ion-source sample holder.

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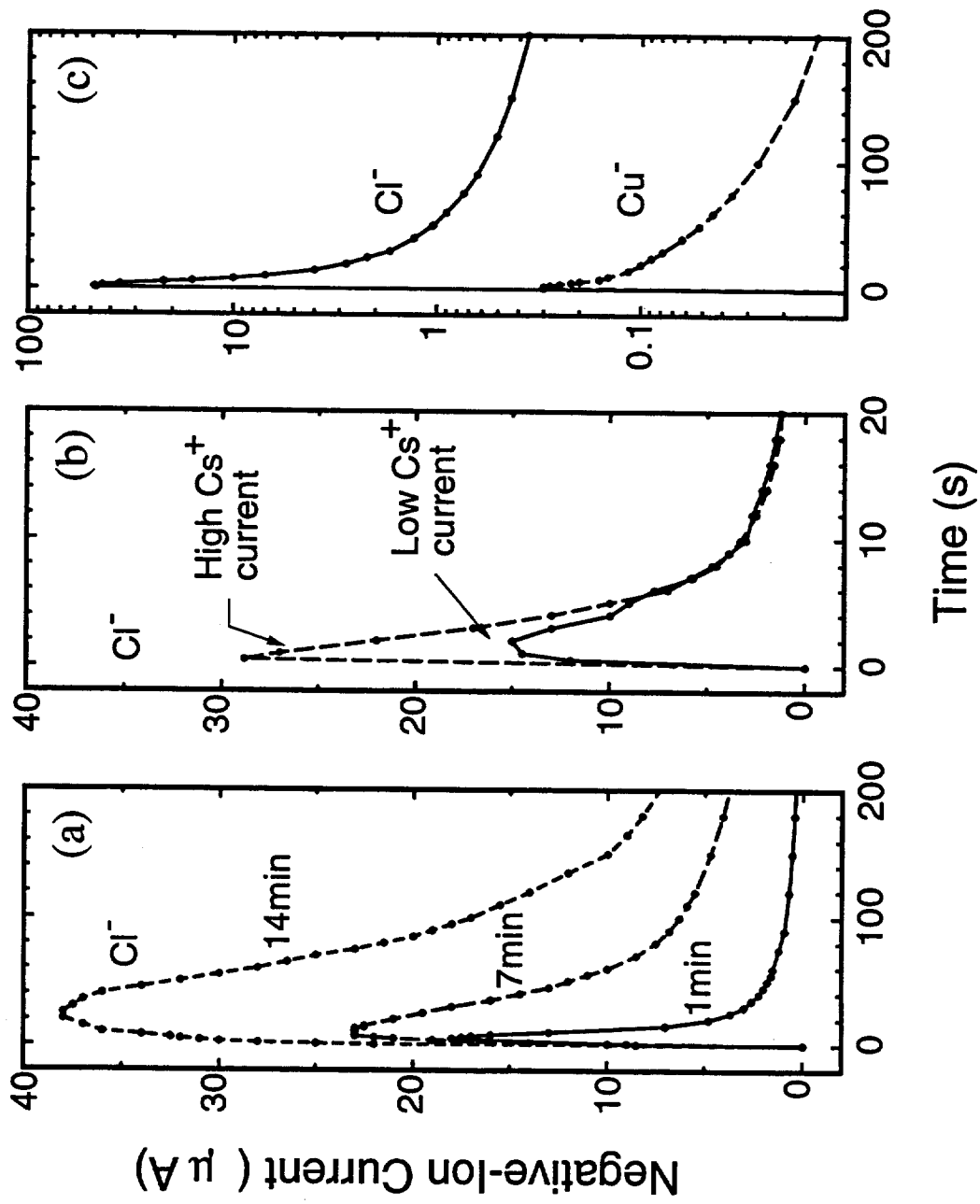
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## FIGURE CAPTIONS

*Figure 1: Time profile of negative-ion currents. Time-zero is when sputtering began. Panel (a) shows the extracted, mass-analyzed  $\text{Cl}^-$  beams from NaCl aerosol depositions of three sizes that were prepared by collecting the deposits over 14, 7 and 1 minute periods on Cu sample holders. The integrated currents are approximately proportional to the deposition periods. Panel (b) indicates that a  $\text{Cl}^-$  beam from two identical NaCl depositions can be intensified if the sample is sputtered with a higher  $\text{Cs}^+$  current. Panel (c) shows the extracted and mass-analyzed  $\text{Cu}^-$  and  $\text{Cl}^-$  beams from two identical  $\text{CuCl}_2$  aerosol deposits prepared on Ta sample holders. The  $\text{Cu}^-$  beam current is lower in intensity than the  $\text{Cl}^-$  beam current because of a substantially lower negative-ion efficiency, but both exhibit tails that probably arise from the secondary process of sputtering Cu and Cl atoms previously forward-scattered and implanted into the sample holder.*

*Figure 2: Absolute sputtering yield versus time from a  $^{56}\text{Co}$ -laden NaCl aerosol deposition. Also shown is the integrated  $\text{Cl}^-$  current (normalized to unity at a time of 10000 s) observed in the time profiles of Fig. 1. Both are observed to saturate as the aerosol deposit becomes depleted since it is less productive to sputter  $^{56}\text{Co}$  or Cl target atoms that have been buried in the sample holder by forward scattering.*

# Time-profile of Negative-Ion Currents



# Sputtering Yield .vs. Time

