

Progress on a Gas-Accepting Ion Source for Continuous-Flow Accelerator Mass Spectrometry

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Abstract

A gas-accepting microwave-plasma ion source is being developed for continuous-flow Accelerator Mass Spectrometry (AMS). Characteristics of the ion source will be presented. Schemes for connecting a gas or liquid chromatograph to the ion source will also be discussed.

1. Introduction

The National Ocean Sciences AMS (NOSAMS) facility at Woods Hole Oceanographic Institution is constructing a compact AMS system optimized for the analysis of ^{14}C in a continuously flowing gas stream. Because it will allow the continuous monitoring of chromatographic effluents, the new system will enable dramatic expansion of significant and well established lines of inquiry including: (i) surveys of the distribution of radiocarbon among natural products and thus of the sources of those materials^[1], (ii) quantitation of ^{14}C tracers at extraordinary levels of dilution, and (iii) recognition of fossil-fuel-derived pollutants in natural systems by exploiting their zero content of ^{14}C as a ‘negative label’^[2].

The key component of this new system will be a gas-accepting microwave-plasma source first built at Atomic Energy of Canada Limited, Chalk River, Ontario, Canada and subsequently developed at NOSAMS^[3,4,5,6,7]. A schematic of the source is shown in Figure 1. The source uses 2.45 GHz microwaves and continuously flowing gas feed to sustain a plasma. Positive ions are extracted from the source and converted to negative ions by passage through a magnesium-vapor charge-exchange canal located at the exit of the ion source.

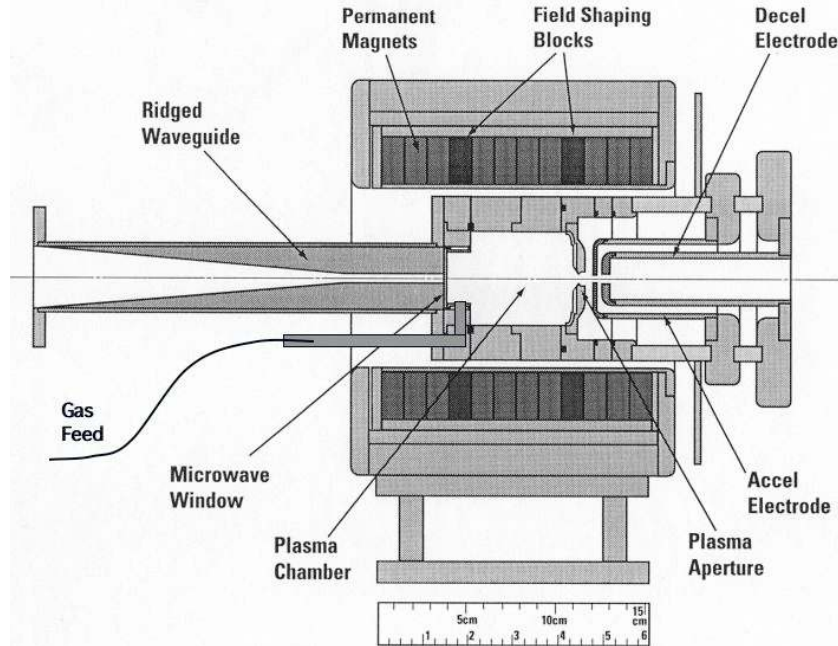


Figure 1. The AECL gas-accepting microwave-plasma ion source. In our case, the length of the plasma chamber was shortened from 5 cm as shown to 2.5 cm to reduce the volume.

2. Performance of the Gas Ion Source

For characterization, the gas ion source was mounted on an ion-source test stand. The arrangement, shown in Figure 2, consisted of a combustion system, a gas-transfer system, the ion source, the magnesium-vapor charge-exchange canal, a 90° double-focusing magnet, two off-axis Faraday cups for the simultaneous measurement of mass-12 and mass-13 beams, and a rotating-wire beam-profile monitor. The double-focusing magnet had a 70 mm aperture with object and image points about 69 cm from the effective field boundary. This setup was useful in optimizing the gas-transfer system, the ion source, and charge-exchange canal.



Figure 2. The ion source mounted on the ion-source test stand. The ion source is to the left inside the cage and the double focusing magnet is back center.

Gas flowed to the ion source via a 100-micron diameter, fused-silica capillary. By varying the length of the capillary, different gas flow rates could be selected. Maximum ion-source output of C^+ from CO_2 gas was obtained with a capillary length of 2.2 m at an extraction voltage of 30 kV. At this length, the measured CO_2 gas flow rate was 240 μ L/min, which agrees well with a calculated flow rate of 230 μ L/min using Poiseuille's equation. The volume of the plasma discharge chamber is 40cc. The chamber is coupled to the external, turbo-pumped vacuum system through a 5mm diameter extraction aperture. The starting, or base pressure, is about 2×10^{-6} T. The conductance through the 5 mm aperture is about 2 l/s, but the gas flow introduced by the capillary results in a pressure differential across this boundary. As the static pressure builds up during gas flow, a discharge starts at about 10^{-5} T, reaching a maximum about 3×10^{-5} T, as measured externally to the discharge chamber. The actual internal pressure of the plasma chamber is unknown.

Figure 3 shows a mass scan of emitted ion beams when pure CO_2 was used as a feed gas. With the charge-exchange canal turned off, and using a flow rate of 240 μ L/min, C^+ ion currents of 400 μ A were obtained at an extraction voltage of 30 kV and 125 watts of RF power. With the charge-exchange canal turned on, C^- currents of 63 μ A were obtained. This implies an ion source and beam transport efficiency of 2.3% for the conversion of CO_2 ions to C^+ ions, a charge-exchange canal efficiency of 15% for the conversion of C^+ ions to C^- , and an overall efficiency of 0.4% for the conversion of CO_2 ions to C^- ions.

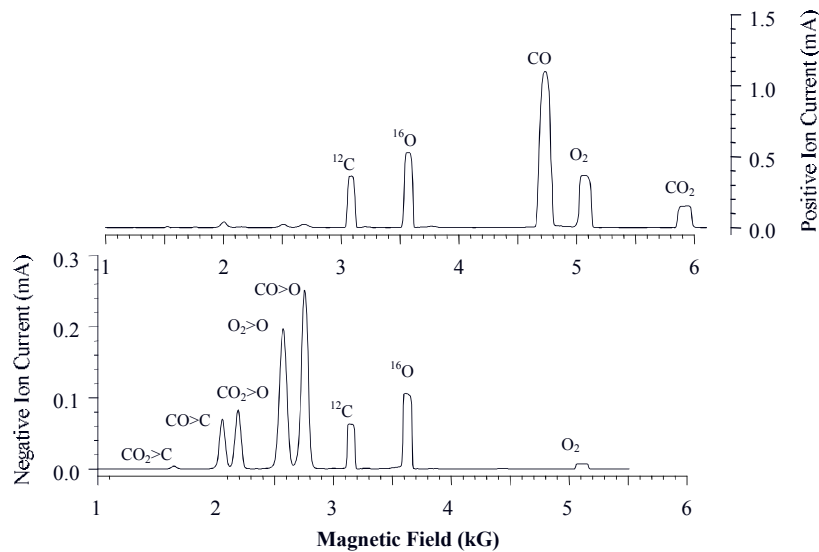


Figure 3. Mass scan using CO_2 as a gas feed. The upper scan is with the charge-exchange canal turned off (positive ions) while the lower scan is with the charge-exchange canal turned on (negative ions). The extraction potential was 25 kV.

Divergence of the ion beam was measured by using a National Electrostatics Corporation (NEC) rotating-wire beam-profile monitor in a setup shown schematically in Figure 4. Using CO_2 as a feed gas, a 30 keV positive-ion beam was extracted from the 5 mm ion source aperture and passed through the 10-mm diameter charge exchange canal. At 410 mm from the exit of the charge exchange canal, the negative-ion beam had a diameter of 23 mm. This gives a divergence of approximately 16 mrad and an approximate beam emittance of $80\pi\text{-mm}\cdot\text{mrad}$. This is significantly more than the emittance of a typical solid-sample cesium sputter-source ($8\pi\text{-mm}\cdot\text{mrad}$ for the NEC SNICS source^[8], $40\pi\text{-mm}\cdot\text{mrad}$ for the Lawrence Livermore National Laboratory sputter source^[9]). For our measurements, it is important to note that the positive- and negative-ion beams were not momentum analyzed. Reported beam sizes include all ions (e.g., C, O, CO, O_2 , CO_2 , etc.).

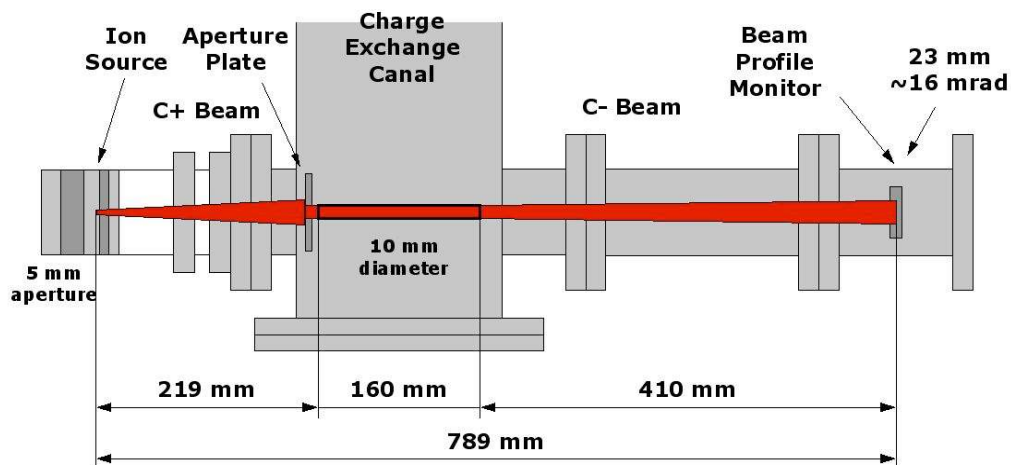


Figure 4. Schematic of the setup used to determine beam divergence and emittance. Beam emittance is approximately $80\pi\text{-mm}\cdot\text{mrad}$ at 30 keV.

A magnesium charge-exchange canal was used to convert C^+ ions to C^- ions. In our case, maximum momentum-analyzed C^- currents were obtained at magnesium temperatures of about $490^\circ C$. Unfortunately, a large energy spread was introduced into the beam during the charge-exchanging process. Calculations with SRIM^[10] indicated an average energy loss of 325 eV with an energy spread of ± 323 eV at a nominal beam energy of 30 keV. Using the test stand magnet as an energy analyzer, this energy spread was also confirmed by experimental observations. To efficiently transport a beam with large energy spread, the associated AMS system will need to be energy-stigmatic (or achromatic) at the low-energy end.

To test the ion source for memory and stability effects, the gas ion source and charge-exchange canal were temporarily mounted in place of one of the standard sputter ion sources on the current AMS system. Unfortunately, the angular divergence and energy spread of the ion beam was large relative to that from the standard source. To avoid scattering of ions and charging of surfaces, a beam-limiting aperture of 5 mm diameter was installed at the object point of the injector system. Even with this limitation, C^- beams as large as $22 \mu A$ were obtained and the entrance to the accelerator. When pulses of CO_2 produced by combustion of the Ox-I and Ox-II oxalic-acid standards were analyzed sequentially, the observed ratio of $^{14}C/^{12}C$ ratios was 1.27. Given the low total numbers of ^{14}C ions collected, this result does not differ significantly from the accepted value of 1.29. Figure 5 shows the response of the AMS system when alternating between a ^{14}C containing CO_2 gas stream and 'dead' CO_2 gas stream into the ion source. The absence of any appreciable retention time for ^{14}C is clearly evident.

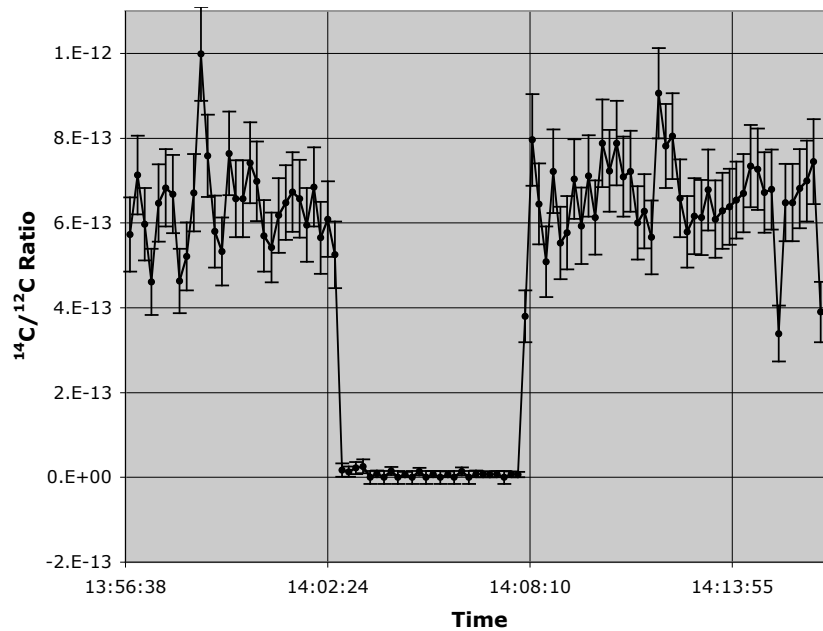


Figure 5. System response to alternating ^{14}C -containing and 'dead' CO_2 gas streams.

3. Future Improvements

In collaboration with Atomic Energy of Canada Limited, Chalk River, Ontario, Canada, we are presently designing an improved version of the ion source. A drawing of the new ion source is shown in Figure 6. Details of the extraction geometry and magnetic fields were optimized through the respective use of the software PBGUNS^[11] and Infolytica^[12] MagNet. In comparison to the current ion source, the new version has several improvements including: i) better coupling to the magnesium-vapor charge-exchange canal, ii) improved control of the magnetic field (by using a solenoid magnet rather than permanent magnets), iii) improved alignment capability, iv) the ability to operate at potentials higher than 30 kV, v) the elimination

of internal water seals with O-rings, and vi) the use of commercially available brazed insulators (as opposed to a glued ceramic/metal extraction column).

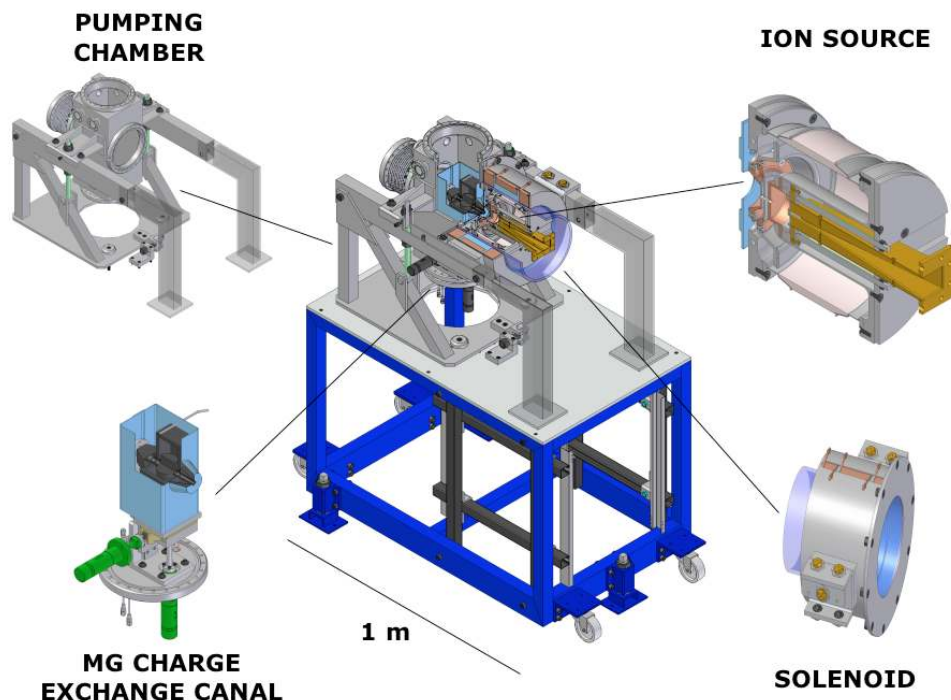


Figure 6. Drawing of the ion source being designed in collaboration with Chalk River Laboratories.

In addition to the listed operational improvements, the new ion source will also be designed so that the magnesium charge-exchange canal can be ‘floated’ to high voltage. The plan is to operate the charge-exchange canal at negative 30 kV while maintaining the ion source at entrance to the low-energy spectrometer at ground potential. The resulting ‘tandem’ charge-exchange canal will yield ion beams with a total energy of 60 keV. This doubling of beam energy will reduce the relative energy spread by a factor of two and the beam emittance by $\sqrt{2}$.

4. Sample Size Requirements

A continuous-flow AMS system is practicable only if means are available for getting samples into a gas stream compatible with the gas-accepting ion source. It will not work if the sample processing devices (e.g., a gas chromatograph) deliver high flow rates of helium and CO₂ while the AMS system requires low flow rates of CO₂ and argon (argon will be used as a carrier gas to buffer CO₂ flow to the ion source to maintain a constant overall flow rate). To address this issue, we must first estimate sample requirements. As stated above, the demonstrated efficiency of the ion source / charge-exchange canal for the conversion of CO₂ ions to C⁻ ions is 0.4%. The AMS system should yield 50% of the incoming C⁻ ions as C⁺ ions at the detector. This gives an overall efficiency of 0.2% for collected ions per molecule of CO₂ introduced.

In AMS measurements, we measure the ratio of ¹⁴C ions collected to ¹²C ions collected:

$$R = \frac{{}^{14}\text{C}}{{}^{12}\text{C}} \quad (1)$$

and the error in the ratio is given by:

$$\frac{\sigma_R}{R} = \sqrt{\frac{1}{^{14}n} + \frac{1}{^{12}n}} \approx \sqrt{\frac{1}{^{14}n}} \quad (2)$$

where ^{14}n and ^{12}n are the number of ^{14}C and ^{12}C ions collected. To achieve a 10% statistical uncertainty ($\sigma_R/R = 0.1$) we need 100 ^{14}C ions. For a half-modern sample ($R \approx 0.5 \times 10^{-12}$) this requires 200×10^{12} ^{12}C ions. Using an overall efficiency of 0.2%, this requires admission to the ion source of 1×10^{17} molecules of CO_2 or approximately 0.2 μmol of CO_2 (2 μg of C) be admitted to the ion source.

5. Summary

A gas-accepting ion source suitable for continuous-flow Accelerator Mass Spectrometry has been characterized. The ion source charge-exchange canal combination produces reasonable ion currents with low memory. Unfortunately, the combination also produces a beam with large emittance and large energy spread. To avoid excessive transmission losses, the AMS system to which this source will be connected will need to have large-gap optical elements and a low-energy, energy-stigmatic transport system (or achromat). An ion optical modeling of an AMS system with these characteristics is described elsewhere in these proceedings^[13].

6. References

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