DEVELOPMENT OF ECR HIGH PURITY LINERS FOR REDUCING K CONTAMINATION FOR AMS STUDIES OF ^{39}Ar

Chris Schmitt, Matt Bowers, Philippe Collon, Daniel Robertson, University of Notre Dame, Notre Dame, IN 46556, U.S.A.

Dale Henderson, Cheng-Lie Jiang, Richard Claude Pardo, Ernst Rehm, Robert Scott, Richard Vondrasek, ANL, Argonne, IL 60439, U.S.A.

Frank Calaprice, Ernst de Haas, Cristiano Galbiati, Princeton University, Princeton, NJ 08544, U.S.A.

Michael Paul, Hebrew University of Jerusalem, Jerusalem, 91904, Israel

Walter Kutschera, Vienna Environmental Research Accelerator, Institut fuer Isotopenforschung und Kernphysik, Universitaet Wien, Austria

Abstract

The first application of ^{39}Ar Accelerator Mass Spectrometry (AMS) at the ATLAS linac of Argonne National Laboratory (ANL) was to date ocean water samples relevant to oceanographic studies using the gas-filled magnet technique to separate the ^{39}K-^{39}Ar isobars. In particular the use of a quartz liner in the plasma chamber of the Electron Cyclotron Resonance (ECR) ion source enabled a ^{39}K reduction of a factor ~130 compared to previous runs without liners and allowed for our current lowest detection limit of ^{39}Ar/Ar = 4.2x10^{-17} [1]. We are currently working on improving the AMS method for ^{39}Ar by following two development paths to allow for higher beam currents while lowering ^{39}K rates. The first option is to modify the design of the quartz liner to provide active water cooling. The second option is to use a thick walled liner of high purity aluminum constructed with an interference fit to the plasma chamber wall. The overall driving force for this AMS project is to search for a source of argon that has a low concentration of ^{39}Ar. Such a source of argon would be useful for new liquid argon detectors that are being developed for detecting dark matter WIMPs (Weakly Interacting Massive Particles).

INTRODUCTION

Commercial argon is obtained from the atmosphere and contains ^{39}Ar, which is produced by cosmic ray interactions with ^{40}Ar in the atmosphere. The ^{39}Ar decays by beta emission with an end-point energy of 560 keV and a half-life of 269 years. The atmospheric concentration of ^{39}Ar relative to ^{40}Ar is 8.1×10^{-16}, corresponding to a beta decay rate of ~1 Bq/kg of argon [2].

Argon can also be found as a trace component in gas that comes from deep underground wells. Shielded from cosmic rays it should have lower than atmospheric levels of ^{39}Ar though there are nuclear reaction mechanisms that can produce ^{39}Ar in underground sites. For example, the neutron induced reaction ^{39}K(n, p)^{39}Ar will occur if there is uranium and thorium together with potassium, since the alpha particles of the U and Th chains produce neutrons by (α, n) reactions on light nuclei [3, 4].

The challenges to detect ^{39}Ar at natural levels are great with ^{39}Ar/Ar (= 8.1×10^{-16}) being a thousand times smaller than that of ^{14}C/C (=1.2×10^{-12}). One liter of "modern" ocean water, i.e. water in equilibrium solubility with the atmosphere, contains only ~6500 atoms of ^{39}Ar, requires very high overall detection efficiency [5].

Since argon does not form negative ions, tandem accelerators, the traditional AMS tool, are unsuitable and positive-ion accelerators must be used and a very high background of ubiquitous ^{39}K, the interfering stable isobar ($\Delta M/Q = 1.55 \times 10^{-5}$) must be separated.

The difficulty in this experiment is also evident in a mechanical sense. From ECR II to the spectrograph there is over 120m of equipment that must maintain relatively good stability for long periods of time (see figure 1). Factors that come into play range from: the output of the ion source, the tune of the beam provided by the accelerator, beam line elements like magnets that can drift

as the temperature fluctuates, and thin windows that separates gas filled regions from vacuum that can tear, potentially ruining the vacuum in the beam line.

Another aspect of this overall difficulty is the estimated ^{39}Ar count rate. The highest beam current from the ion source during the last run was between 130-133 eμA so assuming 20% transmission from ECR II to the spectrograph focal plane we get:

- 100% atmospheric Argon ≈ one ^{39}Ar count per minute
- 10% atmospheric Argon ≈ one ^{39}Ar count in 10 minute
- 1% atmospheric Argon ≈ one ^{39}Ar count in 1.7 hours
- 0.1% atmospheric Argon ≈ one ^{39}Ar count in 17 hours.

The standard experimental time for attempting measurements is a week. This quantifies the necessity for stability and the need for an ultra pure liner that can provide high source output and low background to allow for the measurement of a "representative" sample.
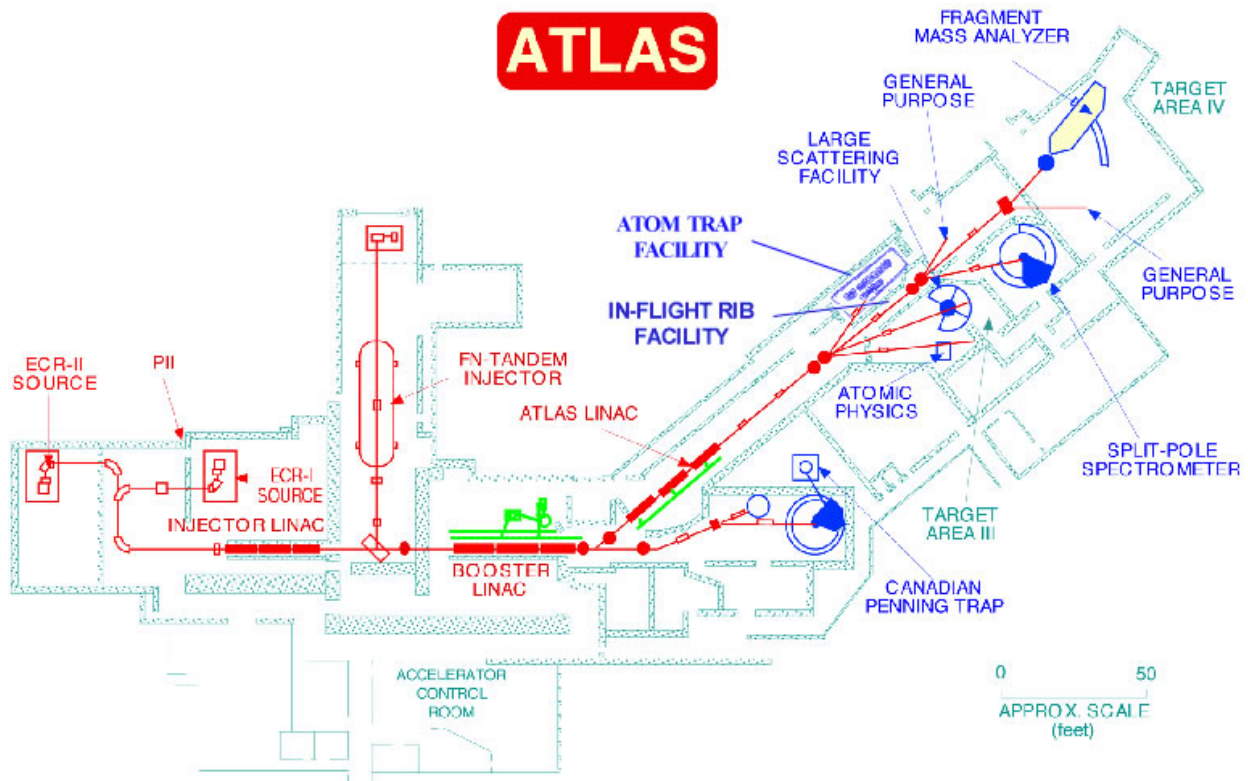

Figure 1: ATLAS facility floor plan at Argonne National Laboratory. From ECR II to the Split-Pole Spectrometer is approximately 400 feet (120 m).

One of the initial goals of our research was to develop and to improve the AMS technique to the stage of obtaining reasonably precise ^{39}Ar measurements from 20 liter water samples for oceanographic studies (see next section) [6, 1]. In the case of ^{39}Ar analysis using AMS, the isobaric separation from ^{39}K was achieved using the gas-filled spectrograph technique [7] (see figure 2). The two key effects combined in this technique, the bending of the ions trajectory in the magnetic field and the loss of ion energy in the gas, have different relations to the charge and mass of the ion, thus allowing the separation of both isotopes and isobars. In particular, in the gas-filled region of the magnet, the discrete trajectories of each of the charge states of the ions coalesce around a trajectory defined by the mean charge state of the ion in the gas [8]. The development of this method and the ^{39}Ar/Ar measurements that followed were performed at ANL using the ATLAS accelerator facility.
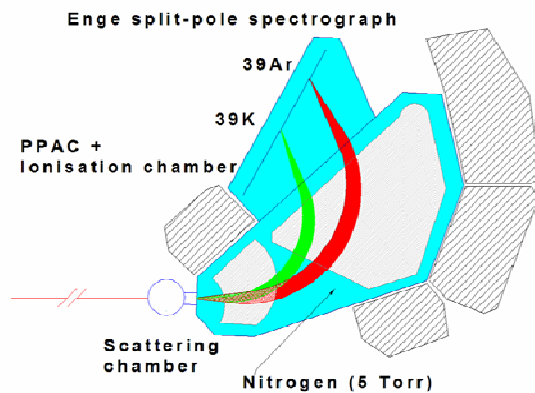

Figure 2: In the gas-filled magnetic region, the discreet charge states coalesce around a trajectory defined by the mean charge state of the ion in the gas.

^{39}Ar^{8+} ions were alternately produced in ECR I and ECR II sources, accelerated to 232MeV by the linac, separated from their isobar ^{39}K^{8+} ions in the gas-filled spectrograph, and detected using a position sensitive parallel plate avalanche counter (PPAC) followed by an ionization counter [7, 9] (see figure 3).
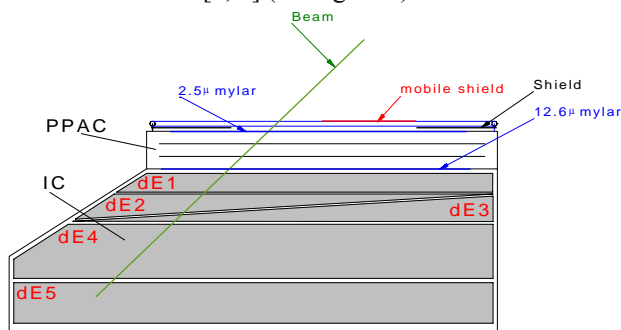

Figure 3. Typical detector setup with a position sensitive PPAC on top and below an ionization chamber.

Although experiments previous to May 2002 obtained a clear separation between the ^{39}K and ^{39}Ar peaks, the intensity of the ^{39}K count rate created signal pile-up in the detector. This pile-up became a major problem when trying to measure samples with lower than atmospheric concentrations and making it impossible to consider increasing the overall beam intensity for AMS measurements as the ^{39}K isobaric background scales with the ^{40}Ar^{8+} beam intensity. A series of experimental runs were initiated to try various plasma chamber liner configurations in order to reduce the amount of potassium coming from the source.

MOTIVATIONS

Oceanographic Applications

With the atmospheric concentration of ^{39}Ar/Ar = 8.1 × 10^{-16}, the practical use of ^{39}Ar as a tracer represents a major technical challenge: 1 L of modern ocean water produces ~17 ^{39}Ar decays per year. As long as large ocean water samples for ^{14}C analysis (~250 L) were routinely collected, low level counting (LLC) of ^{39}Ar extracted from five large volume samples (~1000 L) water samples was feasible [5]. However, with the advent of AMS, only 1 L samples were required for ^{14}C measurements, reducing the routine availability of large water samples. As a consequence, the LLC method for oceanographic ^{39}Ar measurements was practically put on hold and no data have been obtained during the past decade. Therefore it became vital to develop an AMS technique for ^{39}Ar measurements at facilities such as the ATLAS linac facility of ANL.

The first applications of ^{39}Ar AMS at ATLAS to date ocean water samples relevant to oceanographic studies (May 2002, October 2003) were most successful. In particular the use of a quartz liner in ECR II made it possible to reduce the ^{39}K background intensity by a factor of ~130 compared to previous runs. This opened the door to measuring the ^{39}Ar content of actual ocean samples and our measurement demonstrated an excellent detection limit of ^{39}Ar/Ar = 4.2x10^{-17}, corresponding to more than 4 "equivalent half-lives" [1, 6]. However to achieve the original program goal, i.e. to develop a technique that would allow to perform ^{39}Ar/Ar isotope ratio measurements with an uncertainty of the order of 5% using no more than 20 L of ocean water samples, it was necessary to increase the source beam current as well as to improve the overall detection sensitivity while keeping the ^{39}K background at acceptable levels [4]. This has been the focus of the October 2003 run as well as subsequent tests by the ECR group at ANL.

Dark Matter Searches

Recently work done by Frank Calaprice and Cristian Galbiati from Princeton University has opened up a new motivation to develop this AMS technique for ^{39}Ar in groundwater [10]. The driving force for the use of AMS is to search for a source of argon that has a low concentration of ^{39}Ar. Such a source of argon would be useful for new liquid argon detectors that are being developed for detecting dark matter WIMPs.

Standard liquid argon, which is obtained from atmospheric argon gas, has a small but important component of ^{39}Ar owing to cosmic ray reactions. Argon gas is also found underground in natural gas and CO_2 gas wells. Because the cosmic ray flux is suppressed, underground argon may be low in ^{39}Ar. However, underground processes can also produce ^{39}Ar by a sequence of (α,n) and $^{39}K(n,p)^{39}$Ar reactions in rock with high concentrations of U, Th, and K. A mantle source of argon may fit these requirements. We therefore propose a set of measurements on argon gas samples from different sites to search for a suitable source. A source of argon with ^{39}Ar at <1% the atmospheric concentration would be highly desirable for WIMP searches.

If WIMPs do exist they will collide with ordinary nuclei and the collisions may be detected by observing the recoil atoms. The recoils will have a continuous energy spectrum ranging up to ~100 keV. The event rate expected could be as low as a few events per ton per year with a projected cross section of 3×10^{-45} cm^2. Detecting WIMPs therefore requires a large detector with low background and a low energy threshold.

The rare gas atoms- neon, argon, and xenon - have excellent properties for detecting rare WIMP-induced nuclear recoils. The rare gasses can detect nuclear recoils down to a few keV by scintillation and/or ionization. Moreover, the beta and recoil signals differ, which provides powerful discrimination for separating events due to recoil atoms from ubiquitous gamma and beta events. The difference in the stopping power between recoils and betas leads to significant difference in the ratio of ionization charge to scintillation light detected and also produces different scintillator pulse shapes that are quite significant in argon. It is also expected that the technology can be scaled up to detectors with a mass of hundreds of kilograms.

For a liquid argon detector, the beta-recoil discrimination is particularly important because of background from the atmospheric ^{39}Ar. Preliminary studies of the beta-recoil discrimination with the WARP 2.3 kg liquid argon detector indicate that the discrimination is marginally sufficient to permit a sensitive WIMP search with a 100 kg liquid argon detector produced from atmospheric argon. Argon with a concentration of ^{39}Ar at or below 1% the atmospheric

concentration would permit a very sensitive search for dark matter.

The AMS technique developed at ANL for measuring ^{39}Ar was well suited for the original oceanographic applications however the proposed measurements on underground argon related to WIMP searches requires improved sensitivity. The limiting factor to the AMS sensitivity is the background beam of K. Reducing the K background beam is therefore part of the goal of the proposed measurements.

PRESENT STATUS OF THE ^{39}K BACKGROUND REDUCTIONS

The successful use of a quartz liner in the plasma chamber to significantly reduce the amount of K background has been demonstrated. However, this is linked to a limitation of beam intensity. The October 2003 run, as well as more recent tests by the ion source group, strongly indicate that the use of a quartz liner limits the argon beam intensities to a present maximum achieved current around 80eμA. Previous tests using metal liners or silicon oxide coatings of the plasma chamber showed significantly higher ^{40}Ar^{8+} currents but were always hampered by extremely high levels of ^{39}K.

The Princeton group is involved in the Borexino solar neutrino experiment and has extensive experience in low background counting. Measurements of the naturally occurring radioactive elements K, U, and Th have been made on a number of materials and a data base has been accumulated that will be helpful for choosing materials to eliminate bulk sources of K in the ECR source. Precision cleaning of critical parts of the ECR source to eliminate surface contamination is also important. Precision cleaning methods with suitable low K cleaning agents have been developed for Borexino and can be applied to reduce the K background in the ECR source.

We are currently pursuing the development of the ^{39}Ar AMS technique at ATLAS using a powerful cleaning technique using detergents with very low metal content (i.e. potassium) developed for removing both particulates and other materials from surfaces, largely driven by the semiconductor industry. Additionally, at Argonne and other labs working on RF superconductor technology, high pressure ultra-pure water rinsing techniques have also been developed. This technique has been used very successfully with the ATLAS resonators to remove surface particulates and improve surface fields. A number of these cleaning techniques have been adopted for low-level counting chambers with particular attention to potassium.

We adapted these techniques to carry out a series of tests to determine if the potassium background in the ECR II ion source can be significantly reduced. A first

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experiment using these techniques was run June 2007 where we tested a cleaned ultra pure Al liner in ECR II resulting in a $^{40}Ar^{8+}$ output of 210 eμA, a clear improvement over the quartz liner case. Without a liner in place, the available beam intensity has been demonstrated up to 340 eμA of $^{40}Ar^{8+}$. These tests were hampered however by extremely high levels of $^{34}S^{7+}$ which made it through the accelerator and spectrograph. These were probably due to the thin ultra pure Al liner having lost thermal contact with the chamber wall. The loss of thermal contact resulted in the melting of part of the aluminum liner during the run (see figure 4). During the June 2007 tests using the high purity Al liner plus cleaning technique resulted in a ^{39}K count rate as low as: $3.6x10^4$ cps, which showed the promise of using Al liners.

Figure 4: The ultra pure aluminum liner after losing thermal contact from the plasma chamber wall.

This opens up the possibility of combining increased argon beam intensity, associated with the use of a more robust high purity aluminum plasma chamber liner or coating, with a suppressed K background resulting from the above mentioned cleaning techniques.

The April 2008 tests with ECR II were dedicated to further investigate techniques using ultra pure aluminum from Hydro Aluminium Deutschland GmbH. Testing both the ultra pure aluminum coated wall to avoid the thermal contact problem previously encountered and then in combination with open quartz liners to try and achieve the coupled outcome of increased Ar beam intensity with lower levels of ^{39}K background.

The spare plasma chamber from ECR II was sent to Princeton for cleaning, using the techniques they have developed, as well as coating the wall with ultra pure Al, which has K at less than 1ppb. These tests were made to avoid the thermal contact problems from the previous run. In addition to this, a new extractor cathode, bias disk, and injector snout were fabricated out of this ultra pure Al material and were used during the run.

Overall the results were very promising. During this run we initially used the open-ended quartz liner in combination with the Al coated chamber. However later on, this liner was removed and the source was run in an "ultra-pure Al plasma chamber configuration" alone. In both configurations no significant ^{34}S contamination was detected. This is a "first" as during previous runs when no

liners were used there was always a detectable ^{34}S component.

Background suppression in the ECR ion source

The use of the ultra pure Al coated chamber also showed marked improvements in ^{39}K levels, both in combination with the open quartz liner as well as without any liner using only ultra pure materials in the plasma chamber.

The levels of ^{39}K using an open ended quartz liner during the August 01 run were: ^{39}K count rate: $1.3x10^6$ cps with an ion source currents of 80eμA. During this run ^{39}K count rates in identical conditions were $3.6x10^5$ cps with a clean separation between the ^{39}K tail and the ^{39}Ar peak. The lowest levels of ^{39}K were achieved during the May 2001 run (Closed quartz liner): ^{39}K count rate: 9800 cps. It is clear though that closed quartz of that configuration is limited to extracted currents of ~80eμA $^{40}Ar^{8+}$. Many subsequent tests have shown that increasing this beam current is not possible with closed quartz. The count rate of ^{39}K without any liner, but using only the high-purity Al coated chamber and end-pieces made from the same material was $1.5x10^6$ cps only hours after the source had been opened twice over a 6 hour period. This is similar to the lowest count rate achieved with open-ended quartz in August 01 and is the lowest level ever detected without a quartz liner. Though some beam line alignment issues prevented us from making reliable measurements to lower levels. We have strong evidence that the ^{39}K count rate was significantly decreasing over time (a factor of 10 over 9 hours) to levels where measurements below our current level would be possible.

Improved sensitivity in the detector

The use of higher gas pressures both in the spectrograph and in the Ionization Chamber allowed us to optimize the separation between the tail end of the ^{39}K peak and the ^{39}Ar peak due to their specific ranges in the detector (see figure 5). Slight modifications to the detector (i.e. different Mylar window support grids) will allow us to further increase this and thereby improve our sensitivity.
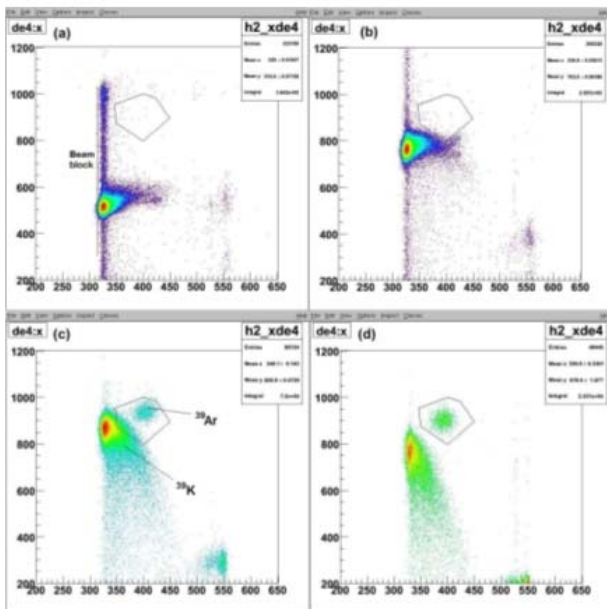
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Figure 5. Position vs dE$_4$ spectrum taken for varying IC and spectrograph pressures. The 232MeV ^{39}Ar + ^{39}K beam was unchanged in these runs. A beam block was placed to block most of the ^{39}K peak and only the tail-end of the peak is observed. In (a) and (b) ionization pressure is changed and data collected for a couple of minutes. P_{IC} = 14.8 torr and P_{IC} = 20.8 torr, respectively. In (c) the pressure is change to P_{IC} = 24.7 torr, but the data collection time is longer approximately an hour. Here we see ^{39}Ar separated from ^{39}K, but there is some signal pile-up. In (d) the N$_2$ pressure (P_{SPEC}) increased from 12 to 13 torr and the separation between the peaks is made much clearer.

DEVELOPMENT PATHS

There are two development paths that will be pursued. The first is a quartz liner that is aluminum coated and actively water cooled. The best results have always come with the closed ended quartz liner and the hope is that the Al coating will allow us to increase the source output (see figure 6).
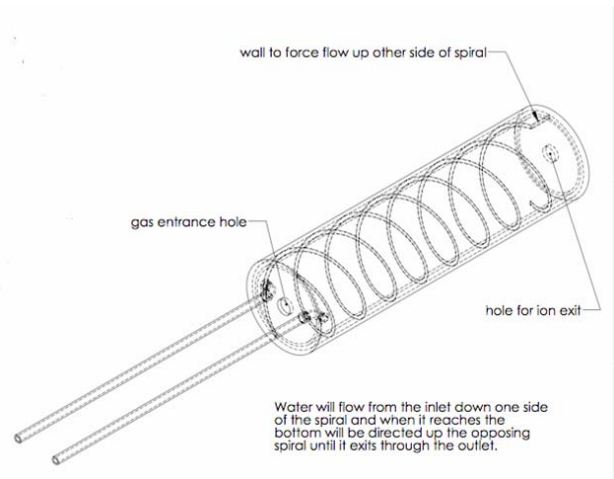

Figure 6: A schematic of the closed quartz liner that is actively cooled.

A several mm thick ultra pure Al liner will be constructed with an interference fit. The chamber will be heated and the liner chilled with liquid nitrogen. When the liner warms up it will make maximum surface contact to ensure continuous cooling.

CONCLUSION

The application of AMS toward ^{39}Ar at ANL began as a way to date ocean water samples relevant to oceanographic studies and now a new motivation for dark matter WIMP searches. There is currently active development to increase the detection sensitivity to improve the AMS method for ^{39}Ar. The principal, though not the only, difficulty is the high ^{39}K background. To reach our goal we need to be able to reduce the background and increase the ion source output.

There are two development paths to allow for higher beam currents while lowering ^{39}K rates. The first option is to modify the design of the quartz liner to provide active water cooling and the other path is to use a thick walled liner of high purity aluminum constructed with an interference fit to the plasma chamber wall. The present results show great promise as is summarized in Table 1.

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Table 1: Summary of Results.

	Date	^{39}K full peak	I_{ECR} (^{40}Ar^{8+})	Comments
^{39}K no treatment	June 07	4.2x10^6 cps	83 eμA	Baseline (a)
Open Quartz	Aug 01[5]	1.3x10^6 cps	83 eμA	Factor 3.2
Closed quartz	May 01 [5]	9800 cps	83 eμA Max output	Factor 430, best ever but limited in current
Open ultra-pure Al	June 07	4.5x10^4 cps	98 eμA	Factor 110, higher beam
	June 07		210 eμA	Max beam output in these conditions
Ultra-pure Al coated chamber	April 08	1.5x10^6 cps	55 eμA	Factor 1.8 (normalized to 83eμA)
	April 08		130 eμA	Max beam output in these conditions

(a) In the above table this run was used as the baseline for ^{39}K count rate.

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