

*PHY4902 Work Term Report*

## **Ion Sourcery**

# Generating Ions for Accelerator Mass Spectrometry

Mohammed A. Chamma - 6379153

Submitted to Professor Serge Desgreniers

University of Ottawa

January 8, 2016

# Contents

Introduction . . . . .	1
History . . . . .	2
Cesium Sputter Ion Source . . . . .	3
Problems and Issues . . . . .	5
Strategies for Ion Source Improvement . . . . .	6
Optimize the Cesium Beam . . . . .	6
Improve the Source's Cooling . . . . .	10
Change the Target Base (again) . . . . .	14
Conclusion . . . . .	16

## **Abstract**

This report describes my second COOP term at the A. E. Lalonde Accelerator Mass Spectrometry Lab in the ARC building at uOttawa and details the topic of ion sources for use with accelerator mass spectrometry (AMS). I introduce the development of ion sources and their design and then focus on cesium sputter ion sources, which are now widely used for AMS. I describe the parts of the cesium ion source manufactured by High Voltage Engineering Europa (HVEE) and then talk about the problems we've encountered using it at the A.E.L. AMS lab. I then outline various designs and strategies for solving some of these problems.

# Introduction

Accelerator Mass Spectrometry is used to measure isotopes in a sample that would be otherwise difficult to detect– or difficult to tell apart– from more abundant isotopes. Examples include counting C14 atoms in a sample filled with C12 for radiocarbon dating, or counting U236 in a sample abundant with U238 to track nuclear fallout. The AMS machine accelerates ion beams using a tandem accelerator, and it is at these high energies that AMS is able to separate isotopes from each other using bending magnets and electrostatic analyzers.

The tandem accelerator works by applying a high positive voltage to the center of the beam pipe. The tandem tank at uOttawa applies 3 million volts, and other tandems can go up to 10 million volts. Negative ions formed in the ion source are accelerated towards the tandem, attracted to the high positive voltage. Once in the center of the tandem the electrons are stripped from the ions so that they become positive. These positive ions are now repelled from the high positive voltage and accelerated once again. The tandem technique uses the same field to accelerate ions twice by switching the charge on the ions at the center of the pipe. The reason this is done with negative ions that are then stripped of their electrons is because it is easier to strip electrons from fast-moving negative ions than it is to give electrons to fast-moving positive ions. The stripping of electrons is done by placing a foil or gas in the path of the anions. When the anions collide with the foil or gas electrons are stripped away. At uOttawa, argon gas is used as a stripper, but carbon foils can also be used. Thus the way the tandem works requires that for accelerator mass spectrometry negative ions must be produced.

In AMS beams of negative ions are produced from sample material that people want to study using a cesium sputter ion source. I give a description of how this kind of source works later in the report.

The properties of the ion source determine the shape, size, and quality of the outgoing beam, what molecules and ions are present in the beam, the amount of contamination that is occurring between samples (that is, whether material from a previous sample is being left in the source and coming out as a beam while different samples are inserted),

how much of the sample material available is actually being used, and the magnitude of the current in the beam.

These factors can all have a role in the final analysis of the material: the amount of current you obtain affects how long a sample needs to be measured for a desired precision. A “quick” time per sample is around 5 minutes, while a “long” time can be around an hour. An hour long measuring time becomes impractical when you’d like to analyse hundreds of samples. The molecules and ions that get produced in the source can affect your results for a particular isotope (for example,  $\text{U}^{235}\text{O}_2^{16,17}$  can masquerade as  $\text{U}^{236}\text{O}_2^{16}$  because they have the same mass, which can add a false signal when counting  $\text{U}^{236}$  atoms.) How much of a sample is actually being used (and how deeply the cesium beam penetrates the target) is important when a sample is very small to begin with. How much of it is wasted to vacuum or deposited in the source? How much of it never gets hit by the cesium beam?

This report will largely be a discussion of the impact the properties of the ion source has on the analysis of sample material. This report will also overview problems that arise when measuring difficult samples, like small Carbon samples or Uranium samples. We’ll also discuss how different designs of the ion source (including the geometry and engineering of the components) can solve the problems that we see when operating the source, as well as some other strategies.

## History

The first mechanism to produce ions for experiments and fun were gas discharge tubes with a perforated cathode. A tube is filled with some kind of gas (say, hydrogen) and on either side of the tube are an anode and a cathode. By perforating the cathode and applying a potential difference of thousands of volts, Eugen Goldstein in 1886 saw rays shoot out of the channels in the cathode.[1] These were positive ions. This “channel ray tube” helped create the field of mass spectrometry. The energies of the ions in this beam had a large spread.

In 1913 J.J. Thomson put neon inside the gas discharge tube and analyzed the beam of positive ions using the apparatus he used to determine the mass/charge ratio of the

electron. This apparatus bent the ions in the beam and separated them based on their masses. From this he was able to conclude that there were two isotopes of neon present, the first demonstration to show that isotopes existed.[1]

From this basic discovery the field of mass spectrometry began and A. J. Dempster in 1917 used a bending magnet to analyze the ion species in a positive beam. Since the magnet deflected ions based on their mass and not their energy he needed an ion source with a low energy spread, and he built an ion source where electrons heated off of a filament would bombard an alkali salt, releasing a positive ion.[1]

In 1931 K. S. Woodcock generated negative ions by bombarding salts with positive lithium ions from a filament.[1] This led to Secondary Ion Mass Spectrometry (SIMS) which refers to the spectrometry of beams of 'secondary' ions: ions that are ejected from some primary ion beam.

This roughly brings us to 1962, when Victor Krohn, while working at a company that was developing ion propulsion engines for NASA, suggested using a beam of positive cesium ions as the primary beam in a SIMS-style spectrometer.[1] Cesium was chosen because of its very volatile and easily given up valence electron, which could conceivably increase the yield of negative ions. Krohn used this idea to create beams of negative ions and analysed them. This was the beginning of sputtering cesium onto a sample as a negative ion source.

## Cesium Sputter Ion Source

Figure 1 shows a schematic of the cesium sputter ion source used at uOttawa as well as a photo of it from the side.

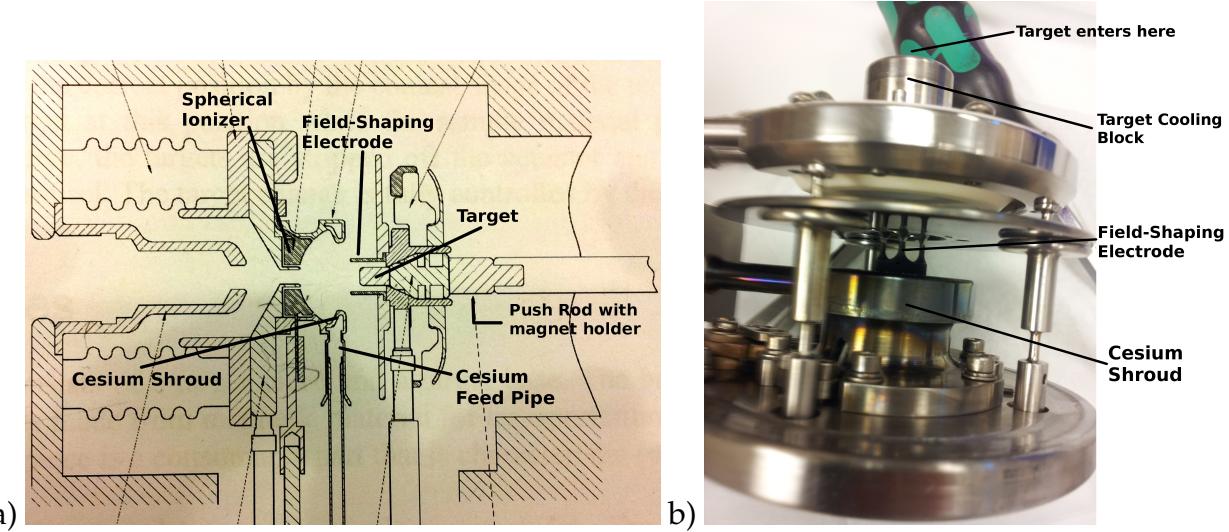


Figure 1: uOttawa's Cesium Sputter Ion Source, manufactured by HVEE

A target containing sample material is pushed into the source by a rod with a magnet on the tip. The target is screwed onto a base made of magnetic steel so that it is held by the rod. Once inside the source the target makes contact with a plate that is held at -35kV. Because the target is conducting, it also takes on the -35kV. This attracts the cesium ions to the target. Around the target inside the source is a field-shaping electrode: a thin ring of conductor that protrudes forward. This electrode is used to 'squeeze' the cesium beam onto the sample material in the target.

The cesium is stored in a reservoir outside the source, and is connected to the source by a pipe that feeds into the cesium shroud. When the source is on, a jacket heats the cesium reservoir until cesium vapor forms and travels up the feed pipe to the shroud. The shroud covers the edges of the ionizer and has pores that the cesium vapor exits out of and onto the ionizer.

The ionizer is a metallic spherical surface made of molybdenum that is held at -28kV (along with the shroud) and is heated to hundreds of degrees. When the cesium vapor makes contact with the hot ionizer, it can lose its lone valence electron, leaving it positively charged. This is how the positively charged cesium beam is formed.

The cesium beam is attracted to the target by the field between the ionizer and the target. When the cesium beam strikes the sample material in the target, it is sputtered, and clouds of debris can be ejected. These particles can pick up an electron (from the

surroundings or from the electrons left behind by the cesium) and become negatively charged. These negative ions are pulled the opposite direction of the cesium and can pass through a hole in the center of the ionizer into the extraction cone, which is held at ground voltage. This helps shape the outcoming negative beam, which can now be shaped and focused by einzel lenses, separated based on the energies present in the beam by the curved surfaces of an electrostatic analyzer, and bent even further based on the masses of the particles in the beam by a large magnet.

## Problems and Issues

The source used by the AMS lab at uOttawa is manufactured by HVEE. There are several problems we encountered using it during the months I was here. This section will describe some of those problems and their consequences.

One of the problems that can occur is melting of the target cathode (the part that holds the sample) during sputtering. This is part of the larger issue of target cooling. When the target is so hot that it melts, sample material can be violently ejected and completely cover the source without being ionized. I talked about this in my last report and described a redesign of the target base so that it was mostly made of copper, which cools faster than steel.

Another problem that we noticed was a periodic drop in the current coming out of the source when doing carbon samples. This wasn't considered a serious problem until a set of small samples were put into the machine. Every minute or so, the current would drop to zero and quickly recover. The results of this carbon dating run had high uncertainty because of these drops.

There is also a more general push to obtain higher currents and better efficiency with the source. Higher current means that small carbon samples, as well as iodine samples and uranium samples can be measured more quickly. However, while turning up the amount of cesium in the source (by raising the reservoir temperature) is a good way to increase the beam current, targets are more prone to melting damage and especially in the case of uranium, severe contamination of the source. The purpose of the AMS lab is

to analyse as many samples as possible as accurately as possible, so this cesium 'limit' so to speak becomes a serious obstacle to the growth of the lab.

With this desire for higher current, certain flaws in the source are highlighted. For example, one can examine a target after sputtering and notice that the cesium spot size – the location on the target that the cesium beam strikes– is slightly off center, missing some of the sample material. We also know that the ionizer and the field-shaping electrode serve to focus the cesium beam onto the target, but that the position of the target was only decided upon qualitatively. We already see that the cesium spot size is larger than the diameter of the sample hole. It could very well be that if the target is moved back so that the sample hole coincides with the focus of the cesium beam that the output current will increase.

## Strategies for Ion Source Improvement

### Optimize the Cesium Beam

One strategy to improve the performance of the ion source is to study and understand the cesium beam and how it can be optimized to generate more current.

A simple hypothesis is that the most current is obtained from a sample when the focus of the cesium beam coincides with the surface of the target. Since we already know that the cesium focus is not quite coincident with the position of the target a simple experiment to find the focus can be performed: first produce some targets (out of aluminum for example) that do not have a hole in the center for any sample material, but can otherwise be screwed onto the target base. When these targets are inserted into the source, the cesium will sputter onto it and leave a very clear circular spot on the target surface. We can now shave off different amounts off the top of the target to produce targets of differing heights. When we look at the sizes of the cesium spots we expect targets of the same height to have the same size while the shorter targets will either have smaller or larger spot sizes on them. This can give us an idea of which way the target needs to move (further in or pulled back) to coincide with the cesium focus. This experiment hasn't been done yet but we expect the cesium focus is around 5mm behind where the target cur-

rently sits. This kind of experiment is simple to do and gives valuable information that is otherwise difficult to obtain: the source is built so that the target is pushed by a rod until the target base makes contact with a cooling cone inside the source, so setting it up so that the target can be pulled back while its base still makes contact with the cooling cone could require a lot of reengineering.

Another strategy in finding the cesium focus is to model the electrical fields in the source and to simulate the trajectories of cesium and other ions in the source. In my last report I gave an explanation of simulations of the beam line using a program called Simion, a program which does not take into account the effect of space charge: the phenomenon where ions in a beam interact with each other electrically. For the beam line this limitation is acceptable, but inside the source the density of charge is much higher and the space we're interested in much smaller, and ignoring the effect ions have on each other could leave us with a simulation that doesn't correspond to what's observed. It has been observed by R. Middleton that after a few minutes of sputtering a small pit in the sample material is dug out and a blue plasma ball forms in the pit, after which the output current increases greatly.[2] There are also ideas that the presence of the positive cesium ions right next to the sample helps to pull negative ions and focus the beam. These considerations make a proper ion simulation with space charge very desirable.

During this work term we worked to learn a software package called Lorentz2E that can do such simulations and to create a basic model of our source. Lorentz2E works by letting you draw some geometry on a two-dimensional plane and then letting you specify the voltages present on the surfaces and shapes you've drawn. Ion emissions are specified by selecting a surface and denoting it as an emitter. You can then say what kinds of ions are being emitted from that surface. So for example, to specify the cesium beam you could draw the arc of a circle to represent the spherical surface of the ionizer and then specify that curved surface as an emitter of Cs+ ions. Once the geometry and emitters are defined, Lorentz2E will calculate the field in the open spaces between the geometry and then the trajectories of the emitted ions. Lorentz2E lets you specify whether or not space charge plays a role. When you tell it to calculate space charge an iterative process begins where the field is calculated, the ions are moved a small distance, the field is recalculated with

the new positions of the ions and so on and so forth. Lorentz2E does not let you specify 3-dimensional geometry, but does let you draw images that are then rotated around an axis. This is for systems that are cylindrically symmetric, which is the case for this ion source. To start, we created a model that was just a 2-dimensional cross section of the source. Figure 2 shows the display from Lorentz2E after a simulation with Cs<sup>+</sup> and C<sup>-</sup> ions.

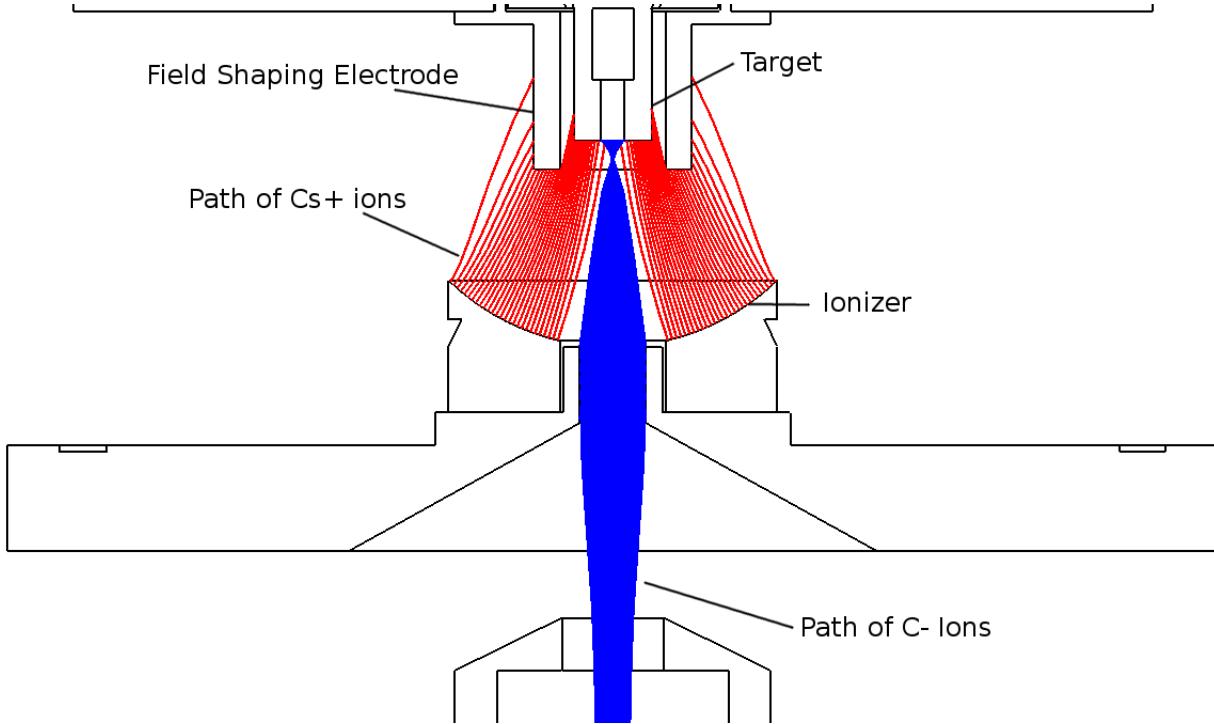
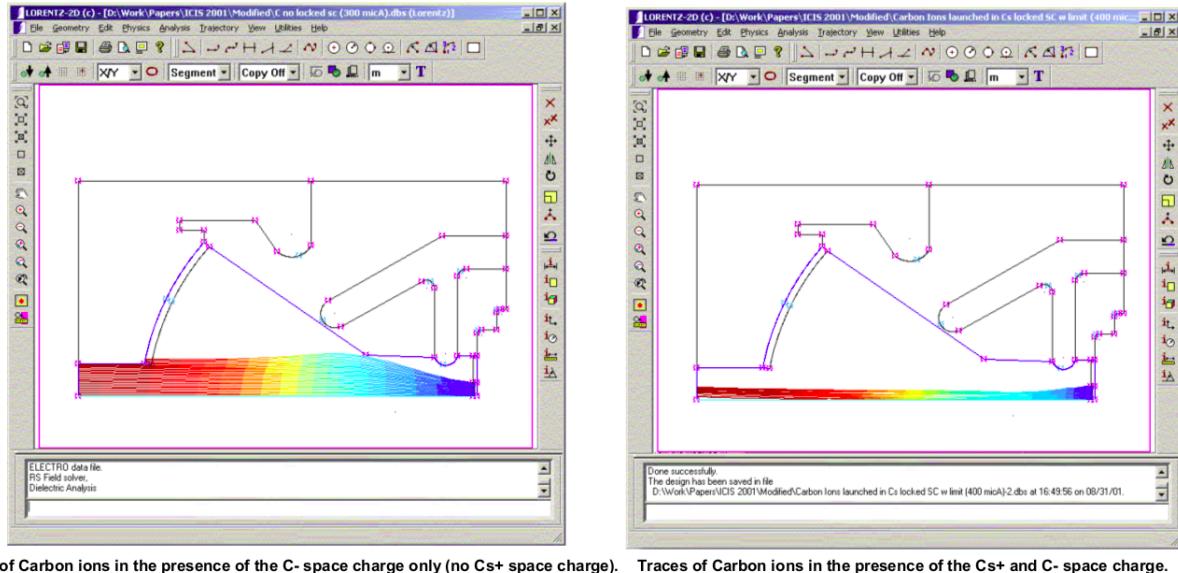


Figure 2: The HVEE ion source in Lorentz2E

In the figure the carbon ions are moving downward from the target and the cesium ions move upward from the curved surface of the ionizer, focused somewhat towards the target.

There are also ideas that after a certain density of cesium ions something exciting happens: a “super beam” forms that is much higher in current and much more focused. This is likely related to the plasma ball that Middleton mentions seeing and the increased current that accompanies it. Testing this experimentally is difficult, increasing the cesium means adding more heat which means the rest of the source needs to be able to handle it. This idea can be seen more readily in simulations however, as done by A. Asi using

Lorentz2D in a model of a slightly different cesium sputter source (Figure 3).



Traces of Carbon ions in the presence of the C- space charge only (no Cs+ space charge). Traces of Carbon ions in the presence of the Cs+ and C- space charge.

Figure 3: No cesium space charge (left) and With cesium space charge (right)[3]

The figure compares a negative ion beam that is not affected by the space charge of cesium and a beam that is affected and we see that the beam is squeezed narrower by the cesium ions, cancelling some of the self-repelling of the negative ions. Asi also reports that the current in this simulation is at least twice as much with the effect of the cesium space charge.

In addition to exploring ideas like the effect of the cesium beam on the extracted current Lorentz2E is also useful in designing new geometries for the source and testing them before producing them. We can use Lorentz to find the optimal distances between components but we can also use it to model different shapes of components. The field-shaping electrode we currently use for example, a thin ring that surrounds the space slightly in front of the target and serves to 'squeeze' the cesium beam, has some sharp surfaces that causes the field to change sharply around its edges. These sharp points can throw ions off course, so redesigning the field-shaping electrode is as simple as redrawing its shape in Lorentz and running the simulation. Other important components Lorentz lets us play with are the extraction electrode, the ionizer, and the target cathode.

## Improve the Source's Cooling

In the quest for higher current, improving the cooling system is worthwhile because it allows the source to handle a higher amount of cesium without overheating. The essence of the cooling system is this: the target's base has a cone-like surface that makes contact with the source cooling block, which has a matching cone surface, is made of steel, and is hollow. A heat transfer fluid called syltherm flows into the source cooling block to absorb heat. The syltherm is then pumped to a heat exchanger that is cooled by water and the cycle repeats with the syltherm in a closed loop.

The target base is normally made of steel. During my last work-term we tried redesigning the target base to have a copper jacket so that the thermal conductivity between the base and the source cooling block is higher. This turned out to be helpful but overall not a groundbreaking improvement.

This term we worked on implementing another idea: chill the syltherm to much lower temperatures so that heat flows more quickly into it. Normally the syltherm is cooled to, at best, the temperature of the water cooling the system, which is around 17-20°C. If we could cut into the cycle and add an additional heat exchange step to get the syltherm even colder then we can have cooler temperatures in the source. To do this we used an immersion chiller: a smallish box-like refrigerator that has a cold finger on a hose connected to the box. The cold finger is meant to be dumped into a bath of some liquid which is then cooled that way. We had two chillers to work with: the first could cool its finger to -40°C and the second one we borrowed could reach -100°C.

We could have set it up so that the syltherm comes from the main pump, is dumped in a bath and cooled by the chiller, and then pumped out of the bath into the source. We didn't have any handy pumps on hand however so we opted to coil 1/4" diameter copper pipe around the cold finger as tightly as we could, then wrap that with foamy insulation material making sure to have an entry and exit pipe sticking out that we could connect to. This was all held together with tape. We then cut into the loop by connecting our coil ends onto the syltherm pipes with Swagelock connections (a way of fastening pipes together), turned on the chiller and turned on the source. The syltherm

would now flow from the pump, into the coil being cooled by contact with the cold finger, and onto the source. This failed however because the coiling increased the impedance of the flow and the pump couldn't keep the flow rate as high as was required by the software, which tripped an alarm and shut down the source when it detected that the flow was too slow.

We undid our work and tried a second idea. We made a syltherm bath in a dewar and stuck the cold finger into the bath. Then, we shortened the copper coil to have a lower impedance and immersed the copper coil inside the syltherm bath. The syltherm circulating in the coil would be cooled by the syltherm in the bath being cooled by the chiller. Though the cold finger could get to  $-100^{\circ}\text{C}$ , the syltherm won't freeze. This took a few tries to get right; we had to keep shortening the coil until the flow was fast enough that the pump wouldn't trip. Of course, shortening the coil means the circulating syltherm spends less time in the cold bath, lessening the heat transfer.

Once the dewar was set up with the coil we covered the top with a tight cap made of aluminum foil. We placed a thermocouple into the bath and turned the source and chiller on. While we could see that the temperature of the bath was quickly dropping to  $-30^{\circ}\text{C}$  (much colder than the  $\sim 20^{\circ}\text{C}$  without the chiller), there wasn't really any indication that anything had improved inside the source based on the current from some test targets. We needed to know what the temperature was inside the flow so we added to the loop a tee-segment just before entering the source and fed a thermocouple into the flow of the syltherm through the 't'. The thermocouple was on the end of a thin steel rod that happened to be the right size for a Swagelock fitting and we fastened it to the tee in the usual way. Swagelock works by biting into the material of the pipe (or rod in this case) to form a permanent leak-proof seal. The rod of the thermocouple was fed through the tee and into the middle of the flow of the syltherm, making sure not to touch the metal of the far wall of the pipe, which we checked visually before attaching the tee segment to the syltherm hoses. This way we get accurate temperature measurements of the syltherm in the flow and of the syltherm in the bath and can gauge how well the heat transfer is occurring. Figure 4 and 5 show photos of the cooling setup we built and temperature readings that characterize how well our system is cooling the source.

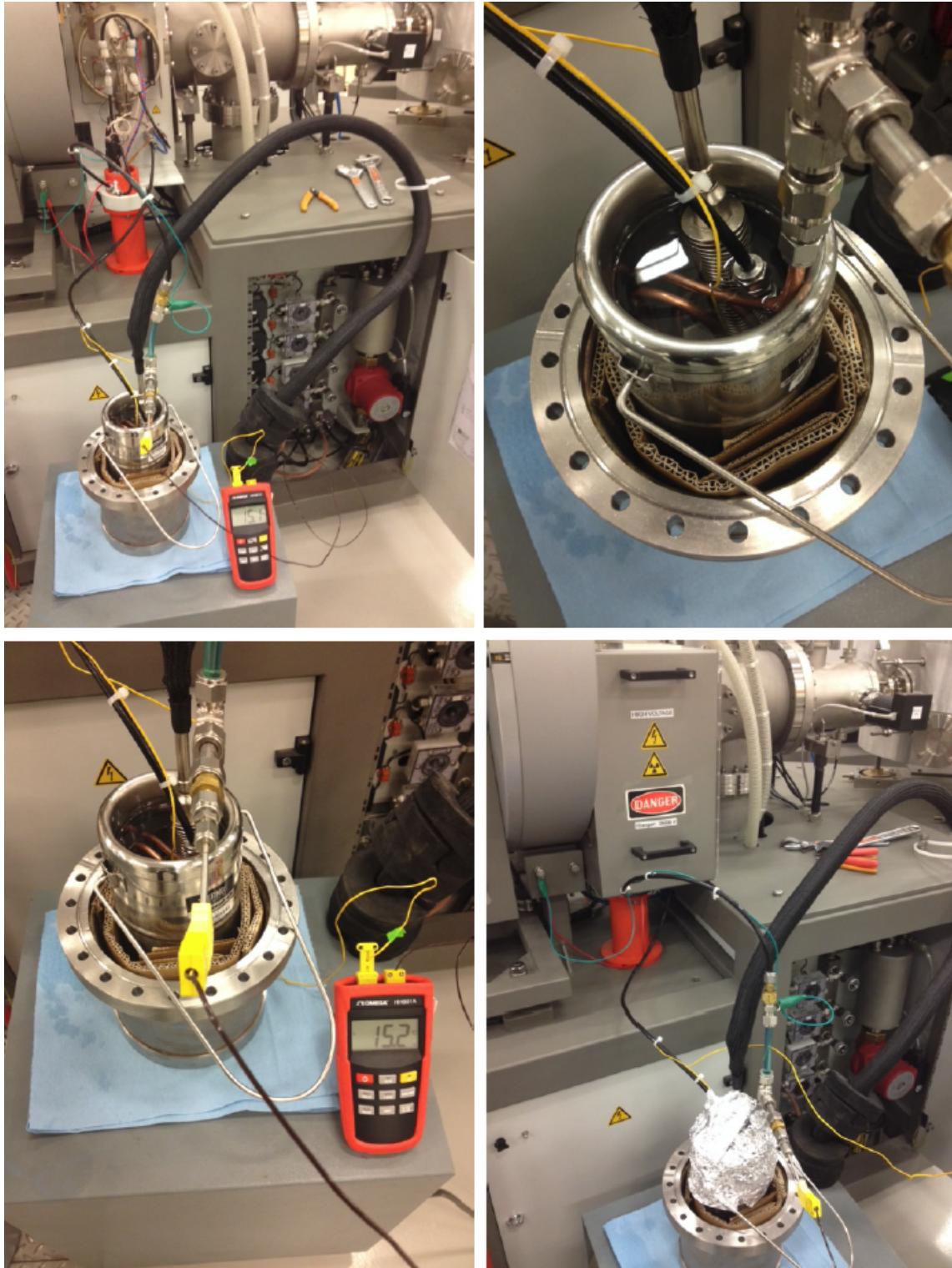


Figure 4: Photos of the chiller set up. Note the coil in the bath shown in the upper right image. Also note the thermocouple jutting out of the tee in the lower left image. The lower right image shows the setup in operation. Photos taken by Dr. Chris Charles.

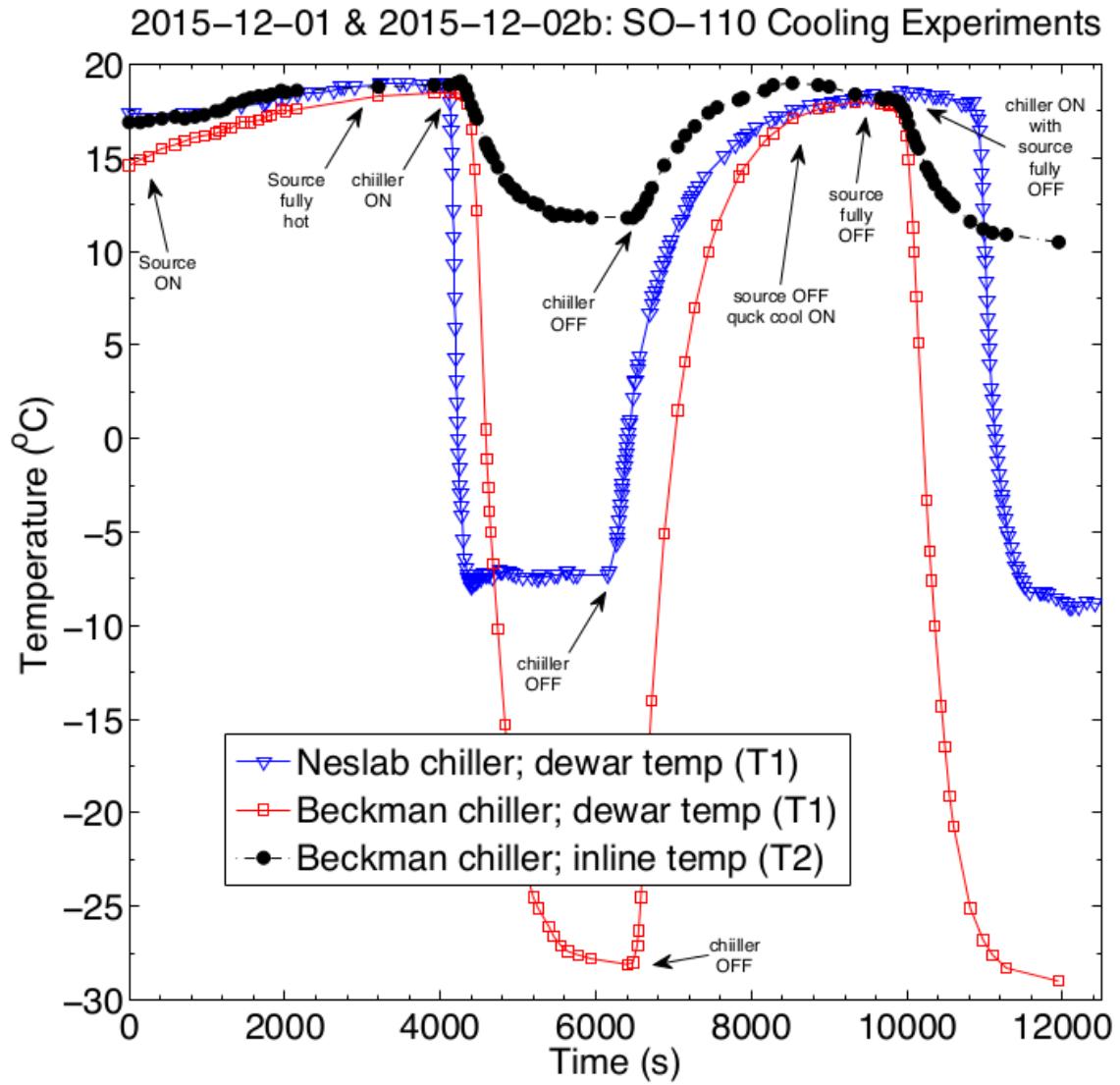


Figure 5: Temperatures during chiller test while the source is on. The Neslab chiller refers to the model of our older, first attempt chiller which reaches  $-40^{\circ}\text{C}$  optimally. The Beckman chiller is the better chiller that reaches  $-100^{\circ}\text{C}$ . The goal is to make the inline temperature, denoted by the black circles, dip as low as possible. Figure by Dr. Chris Charles.

As can be seen in Figure 5 the temperature of the syltherm in the flow just before entering the source with the good chiller on is around  $12^{\circ}\text{C}$ , while we also know that the cold finger has cooled the syltherm in the bath to  $-28^{\circ}\text{C}$ . We can also compare the temperatures from the graph before and after the source is turned on to see that the heat load from the source and after the main heat exchanger (since the syltherm is heated and then cooled before we measure it again in our setup) leaves the syltherm around  $2^{\circ}$

hotter. This value doesn't seem to change whether the chiller is on or off, also based on the graph. The chiller when turned on in our setup drops the temperature of the inline syltherm by about  $6^{\circ}\text{C}$ , which is a little disappointing considering the chiller gets much colder, as shown by the temperature of the bath.

Improving on this is difficult: we know that we can easily increase the temperature drop by just adding more coil into the bath (this setup has maybe two turns inside the bath) but we know that the syltherm pump will trip if we do that. So we seem to be stuck in having to get a more powerful pump or maybe a second pump placed at the coil. The situation inside the source is also unclear. Ideally we would place a thermocouple inside right in the vicinity of the target, but this has its own problems. The ionizer we know can heat up to  $1200^{\circ}\text{C}$  so this thermocouple could just melt (unless it's very expensive). A thermocouple inside would also need to be placed so as to not interfere with the electric fields and so as to not erode with time due to sputtering.

Ultimately the story of cooling has to do with increasing cesium and with limiting the violent ejection of sample material by overheating the target. Failing a thermocouple, the clear indication of an improvement in source cooling will be given by comparing the analysis of a sample that is overheating with one that is well cooled: current should be expected to be more stable, the voltages on the source components will fluctuate less, background signals and counts will be lessened, and less current caused by contamination will be seen.

### **Change the Target Base (again)**

Another way to improve the situation in the source is to once again redesign the target base. The last design of a base we created had a copper jacket to improve cooling. But now since we also expect that the cesium focus is behind the current position of the target, a target base that moves the target back could be useful. In order to reduce the amount of machining and re-engineering required, we can embed the target inside the base so that this new base interfaces with the cooling in the HVEE source in the same way. Figure 6 shows a drawing of such a concept.

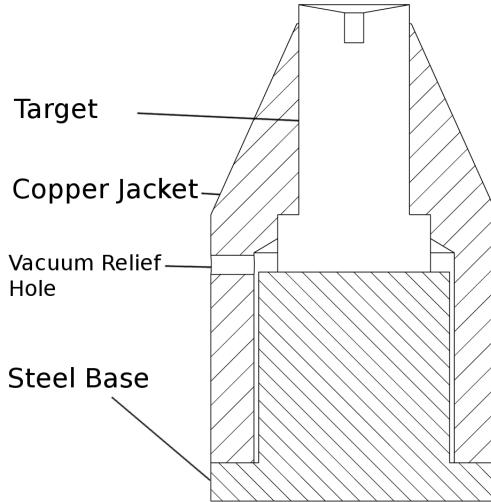


Figure 6: Target Base Concept

The target in this concept is clamped down onto the steel base by a large copper jacket, which is screwed onto the base. On the regular bases, the bottom of the target is screwed onto the top of the base, which in this figure would correspond to about the top of the target, so the target here has been pulled back about 5mm compared to the regular base. The base needs to be magnetic to be held by the source's push rod. To allow for dimensions with generous tolerances (to reduce machining costs), we need to assume that there might be some gap between the target and the copper jacket. This gap could trap air and act as a virtual vacuum leak in the system because it could take a long time for air to escape from this gap, so a hole is drilled into the side that should allow air to escape more easily and not interfere with the vacuum.

An advantage of this concept in addition to better positioning the target in the cesium beam is that the contact between the copper jacket and the target covers a much larger surface area. In the previous design only the bottom surface of the target touched the copper and that surface was a thin ring, to allow room for a threaded hole in the bottom. Here almost the entirety of the surface of the target makes contact with copper, which then makes contact with the cooling system, so we can expect this kind of base to be much better cooled.

A disadvantage with this base concept is the complexity in sample loading that it adds. Multiple parts need to be kept track of and cleaned, and the targets (which are

mass produced and disposable) now have a more complicated shape to machine. This racks up the cost drastically. It could be that these bases would only be useful for special or delicate samples, so that a few of them might be used alongside the regular bases.

## Conclusion

In this report I've given an overview of ion sources and focused on the cesium sputter negative ion source used at the uOttawa AMS Lab. I talked about some of the problems encountered with the source and then described ways to solve those problems and improve the efficiency of the source and reduce the measurement time with it. These included modeling source to understand the cesium beam, an experiment to find where the target needs to be to be in the focus of the cesium beam, a system to chill the syltherm cooling fluid, and a target base concept that improves cooling and brings back the target into the cesium focus.

Improving the ion source is important for accelerator mass spectrometry because it reduces the measurement time of samples, allowing labs to process more samples, and increases the precision with which samples can be measured by reducing contamination.

I feel grateful towards Dr. Xiaolei Zhao for the discussions about the target base concept, the cesium beam focus, and for the problems he's encountered with the source. I'm also grateful towards Dr. Chris Charles and Normand St-Jean, Collin Tiessen and Vince Morin for helping setup and test the syltherm chiller and for the general day to day work of the lab. I'm grateful to Dr. Liam Kieser for explanations of the tandem and for introducing me to Solidworks and machining. I feel that I've learned a lot during my time at this lab and that I've been able to contribute meaningfully to the work being done here.

# Bibliography

- [1] P. Williams, "Historical Development of Ion Sources for SIMS". Arizona State University. [www.simsworkshop.org/annualworkshops/workshop2013/Historyof\\_ion\\_sourcesfor\\_SIMS.pdf](http://www.simsworkshop.org/annualworkshops/workshop2013/Historyof_ion_sourcesfor_SIMS.pdf). Retrieved January 8 2016.
- [2] R. Middleton, "A Negative-Ion Cookbook", University of Pennsylvania. October 1989. <http://www.pelletron.com/cookbook.pdf>. Retrieved January 4 2016.
- [3] A. Asi, "Ion Source Modeling with LORENTZ-2D", Integrated Engineering Software (2001), [www.integratedsoft.com/papers/research/ICIS2001.pdf](http://www.integratedsoft.com/papers/research/ICIS2001.pdf). Retrieved January 5 2016.