ADDRESSING CONTAMINATION IN ECR CHARGE BREEDERS

R.C. Vondrasek†, Physics Division, Argonne National Laboratory, Lemont, IL, USA 60439

Abstract

The Electron Cyclotron Resonance (ECR) ion source was first utilized for charge breeding in 1995 [1]. Since that time the charge breeding technique has been refined. Single charge state efficiency has improved by a factor of ten, the efficiency discrepancy between solid and gaseous species has narrowed, and low-mass species efficiency has improved. But the limiting characteristic of the ECR charge breeder continues to be a high level of contamination which often obscures the beam of interest [2]. Multiple techniques have been employed to reduce this contamination with varying levels of success, and attempts are currently underway to improve upon the successes achieved to date. This paper will review those past techniques, current attempts, and possible future paths for reducing the contamination level in ECR charge breeders.

INTRODUCTION

Caribu Facility

The CAlifornium Rare Isotope Breeder Upgrade (CARIBU) [3] provides radioactive beams to the Argonne Tandem Linac Accelerator System (ATLAS). Fission fragments are produced by a 200 mCi 252Cf fission source located inside a large-volume helium gas catcher. The fragments are thermalized and rapidly extracted at up to 50 kV forming a low-energy beam of 1+ or 2+ ions. The isotope of interest is selected via a high-resolution (1:20,000) magnetic separator. The beam is then transported to either an in-room experimental area, a remote stopped beam experimental area, or an ion source where the beam is charge bred for subsequent acceleration in the ATLAS linac. Originally, an ECR source charge bred the CARIBU beams, but due to the inability to adequately reduce the stable beam contamination that source was replaced by an EBIS in 2016.

ANL ECR CHARGE BREEDER

The ANL ECR breeder [4] (Fig. 1) is a room temperature source with the plasma excited by two RF frequencies – a 10.44 GHz klystron and an 11-13 GHz traveling wave tube amplifier (TWTA). It has an open hexapole structure providing good pumping to the plasma chamber region and allowing the RF and support gas to be introduced radially. This scheme eliminates the need for cut-outs in the field shaping iron to accept the RF waveguides. The 1+ ions were introduced into the plasma through a grounded high-purity aluminum tube mounted on a linear motion stage with a 30 mm range of travel. The

† vondrasek@anl.gov

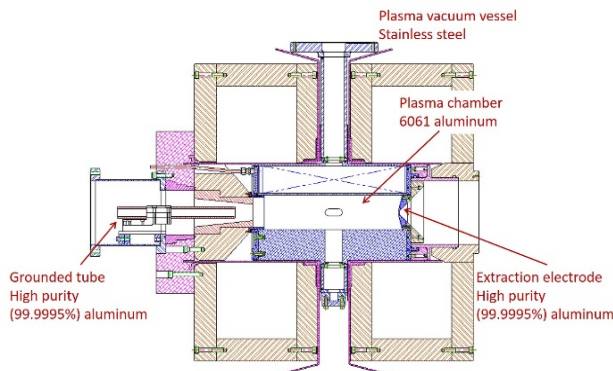source is designed to operate at a 50 kV potential although it typically operated at 36 kV.

Figure 1: The Argonne National Laboratory ECRCB.

BACKGROUND CONTAMINATION

The background contamination which plagues ECR charge breeders has been well documented [5, 6, 7]. A mass scan of an oxygen plasma in the A/q region in which the ANL ECR breeder operated showed numerous peaks for nitrogen, argon, aluminum, fluorine, and chlorine on the >1 epA scale. Even in A/q regions within the spectrum that showed no background as measured with a picoammeter, a more sensitive examination with a silicon barrier energy detector revealed significant levels of background which dominated the radioactive ion beam of interest, as shown in Fig. 2. In the case of 146Ba28+, the radioactive beam accounted for only 3% of the total rate into the detector. The stable contaminants (Ti, Zn, Zr, Mo, Sn, Xe, Ir, Hg) were all within the LEBT resolution window and could only be eliminated by addressing the contamination at the source. In fact, even with a 1:1000 spectrometer after the charge breeder such as the one employed at SPES [8], only the Zn and Xe would be eliminated from the 146Ba spectrum.
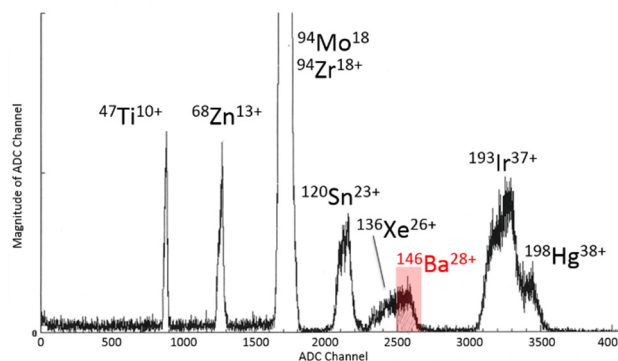

Figure 2: Spectrum is the radioactive beam 146Ba (highlighted in red) and its contaminants from the ECRCB.

Sources of Contamination and Reduction Techniques

There were three identified sources of contamination – gases entering the system via leaks or the support gas, loose particulates on the ion source and surrounding vacuum chamber surfaces, and the bulk surfaces exposed to the plasma such as the plasma chamber, the extraction electrode, or the RF waveguides.

Investigation with a residual gas analyzer (RGA) showed that nitrogen and argon contaminants were not constituents of the oxygen support gas but were mainly due to o-ring permeation and to a smaller extent desorption from the plasma chamber and beamline surfaces. The o-rings were integral to the source construction, and their presence resulted in the equivalent of a 10-5 Torr-l/sec leak limiting the source to an ultimate pressure of 2x10-8 Torr. A method was developed to replace several of the o-rings with indium seals, but the gaseous contaminants were not the dominant problem so this course of action was not pursued.

Wall desorption is normally addressed with a standard thermal bake-out technique, but the presence of the permanent magnet hexapole and the high voltage isolation materials (which are all susceptible to heat damage) precluded this. The vacuum chamber was instead baked out with UV lamps which impart enough energy to desorb the water molecules but do not produce sufficient radiative heat to cause magnet damage. Using this technique, the base pressure was reduced by a factor of two mainly due to decreased water desorption, but there was only a small effect on the overall contaminant load as observed with the silicon barrier detectors.

Particulate contamination was previously addressed by the KEK charge breeding group with sand blasting and high pressure rinsing of the plasma chamber [5]. In the case of the ANL charge breeder, it was not practical to disassemble the ion source so alternative cleaning methods were investigated.

To remove the surface contaminants, CO_2 snow cleaning [9] of the plasma chamber surfaces as well as the injection and extraction hardware was employed. The method is nondestructive, nonabrasive, and residue-free. It is based upon the expansion of either liquid or gaseous carbon dioxide through an orifice leading to the nucleation of small dry ice particles in a high velocity gas carrier stream. The CO_2 pellets remove micron and submicron particulates by momentum transfer and hydrocarbons via a freeze-fracture mechanism. The high-velocity carrier gas propels the contaminants out of the system thus eliminating the need for high pressure rinsing and allowing the entire process to be done in situ.

The last source of contamination was sputtering of the plasma chamber and extraction electrodes, constructed of 6061 aluminum alloy, and the copper RF waveguides. The 6061 alloy has components of magnesium, silicon, titanium, chromium, manganese, iron, copper, and zinc - many of which were observed background contaminants.

Figure 3: The ANL ECRCB plasma chamber after coating with ultra-high purity aluminum. Note the shadow at the top of the chamber caused by the high current lead which shows the uncoated surface. The aluminum disk at the injection side also still shows the plasma loss lines demonstrating the limitations of the evaporation coating technique.

New versions of the extraction electrode and the grounded tube were constructed from ultra-high purity aluminum (99.9995%), cleaned with the CO_2 system, and installed in a clean environment. It was not practical to construct a new plasma chamber from ultra-high purity aluminum. Instead the chamber was CO_2 cleaned and vacuum coated with ultra-high purity aluminum (99.9995%). A tungsten coil which had been saturated with the aluminum was suspended in the middle of the plasma chamber. The source was evacuated to 10-7 Torr and the coil heated resulting in an average surface deposition of 1 micron. While the central portions of the plasma chamber were coated, an injection side disk, the radial ports, and an area shielded by one of the high current leads did not receive an adequate amount of flux, as shown in Fig. 3

Contamination Reduction Results

Before the CO_2 cleaning, a mass scan of the source output was performed with analyzing slits set at +/-0.1 mm recording all peaks with an intensity >1 epA. After the CO_2 cleaning, the scan was repeated with the same source conditions. Reductions in three major contaminants were observed – a factor of 20 reduction for fluorine, a factor of 4 for chlorine, and a factor of 50 for iron.

After the aluminum coating, the mass scan was repeated with the same source settings. The three main contaminants were further reduced – a factor of 160 reduction for fluorine, a factor of 17 for chlorine, and iron was no longer detectable.

While these three components were reduced at the source, the key metric is what is accelerated in the linac. A 98Zr beam had been produced shortly before aluminum coating the source. After coating, a 98Y beam was produced utilizing the same accelerator tune. Energy spectra were obtained for both beams with the silicon barrier detector. As a result of the coating, several of the stable contaminants observed in the silicon barrier spectrum had either been eliminated (iron, cadmium, cerium) or had come down significantly (titanium). A significant 98Mo peak remained and two new contaminants 181Ta and 186W were introduced, all presumably due to the tungsten heating coil used for the evaporation which had a 10 ppm component of molybdenum and a 20 ppm component of tantalum.

Other Reduction Techniques

While the above techniques demonstrated significant reductions in the level of background, especially that due to surface contamination, there are several improvements which can be made.

The o-rings need to be eliminated from the charge breeder design to establish a truly UHV system. This advancement has been incorporated into the construction of the SPIRAL PHOENIX ECR charge breeder [10]. This will eliminate the contribution from any o-ring permeation.

A method of depositing aluminum onto the chamber surfaces without introducing any new contaminants is required. It is possible that a greater reduction in the Mo, Ta, and W components could have been realized if a different heating element was used. Off-line tests with various carbon-based heating elements indicated pyrolytic graphite is the best candidate due to its high temperature performance, high purity, and low reaction rate with the aluminum.

A previously untested technique of introducing a pure aluminum coating is atomic layer deposition (ALD) [11]. This technique utilizes gaseous chemical compounds which bind to exposed surfaces producing a thin film comprised of successfully deposited monoatomic layers. The ECR chamber could be processed in situ using the compound trimethyl aluminum (TMA) [12].

The source chamber would be evacuated to 10-5 Torr and back-filled with the TMA. The TMA chemisorbs to the water monolayer present on the unbaked vacuum surfaces. Once all reactive sites are occupied, the process self-terminates and the remaining TMA is pumped from the chamber leaving a surface of AlCH3. The system is then back-filled with a carrier gas containing a small amount of water which reacts with the –CH3 forming CH4 and resulting in a hydroxylated Al2O3 surface thus enabling the cycle to be repeated. The cycle time can be as short as 30 seconds and results in the deposition of a 1.29 Å layer. Assuming an automated system operating on a 30 second cycle time, a one micron layer of aluminum can be deposited in <3 days. Since the process is conformal, all exposed surfaces are coated regardless of geometry.

FUTURE PLANS

The ALD technique will be tested with a new permanent magnet ECR source [13] being installed at ATLAS (Fig. 4). Since all surfaces of the source will be coated – chamber walls, vacuum chamber, RF waveguides, insulators, o-rings – this should result in an absolutely clean surface exposed to the plasma. Tests will be performed regarding source function before and after coating with a focus on source contamination levels. The longevity of the coating will also be an important metric.
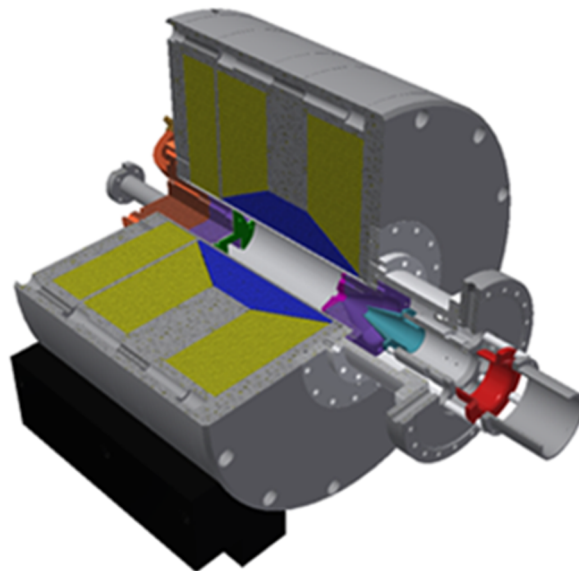

Figure 4: The new all permanent magnet ECR source which is being installed at ATLAS and will be used as test bench for the atomic layer deposition technique.

ACKNOWLEDGMENTS

REFERENCES

[1] R. Geller, C. Tamburella, and J. L. Belmont, R*eview of Scientific Instruments*, vol. 67, p. 1281 (1996)

[2] R. Vondrasek, J. Clark, A. Levand, T. Palchan, R. Pardo, G. Savard, and R. Scott, *Review of Scientific Instruments*, vol. 85, p. 02B903 (2014)

[3] R. Pardo, G. Savard, S. Baker, C. Davids, E. F. Moore, R. Vondrasek, G. Zinkann, *NIM-B,* vol. 261, Issues 1-2, p.965, (2007)

[4] R. C. Vondrasek, R. Scott, J. Carr and R. C. Pardo, *Rev. Sci. Instrum*., vol. 79, p. 02A901 (2008)

[5] N. Imai, S. C. Jeong, M. Oyaizu, S. Arai, Y. Fuchi, Hirayama, H. Ishiyama, H. Miyatake, M. H. Tanaka, M. Okada, *et al*., *Rev. Sci. Instrum*., vol. 79, p. 02A906, (2008)

[6] F. Ames, R. Baartman, P. Bricault, K. Jayamanna, *Hyperfine Interactions*, January 2014, vol. 225, Issue 1-3, pp. 63-67 (2013)

[7] R. Vondrasek, J. Clark, A. Levand, T. Palchan, R. Pardo, G. Savard, R. Scott, Rev. Sci. Instrum. 85, 02B903 (2014)

[8] A. Galata; IOP Conf. Series: Journal of Physics: Conf. Series 874 (2017) 012052

[9] www.co2clean.com

[10] P. Delahaye, L. Maunoury, R. Vondrasek, Nucl. Instrum. Meth. A 693(2012) p. 104-108

[11] J. Lu, J. W. Elam, and P. C. Stair, Surface Science Reports, 71, 410-472, (2016)

[12] R. Puurunen, Journal of Applied Physics 97, 121301 (2005)

[13] D. Xie, Review of Scientific Instruments 73, 531 (2002)