

METALLIC BEAM DEVELOPMENT WITH AN ECR ION SOURCE AT MICHIGAN STATE UNIVERSITY (MSU)*

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Abstract

Electron Cyclotron Resonance (ECR) ion sources have been used at MSU to provide metal ion beams to the coupled cyclotron facility (CCF), and in the future, for The Facility for Rare Isotope Beams (FRIB). The challenges of metallic beam production with ECR are in production, efficiency, stability and contamination. Future facilities such as FRIB will add the challenge of intensity. We report development of two rare earth metals and the conversion from the oxidized state into metal. The enriched isotopes of ^{144}Sm , and ^{176}Yb are commonly available in the sesquioxide form which is unsuitable for use in our standard ovens. We report here results from the off-line chemical reduction of samarium, and ytterbium oxides into metal. We were able to demonstrate efficiencies of up to 90% throughout the conversion process. The samples were then run on our ECR ion sources to confirm the products of the reduction. In addition we report the development of cadmium metal by passing vapor through over 3/4 m of heated stainless steel tubing and observed $4.3\ \mu\text{A}$ of Cd^{20+} with an average consumption of 1 mg/hr.

INTRODUCTION

The Coupled Cyclotron Facility (CCF) at Michigan State University provides the nuclear science community with beams of rare isotopes produced by fast fragmentation. All rare isotopes produced through CCF operation have been obtained from about 30 primary beams (from Oxygen to Uranium) accelerated to an energy range of 80 to 160 MeV/u. To enhance the yield of rare isotopes, the primary beam is generally a separated isotope with either the largest or the smallest neutron excess available. So far the primary beam list has never included rare earth elements. Specifically of interest for the nuclear science community are ^{144}Sm , and ^{176}Yb , for good production of proton rich and neutron rich elements respectively. Initial production of the ion beam is done using an ECR ion source. Normal operation of an ECR relies on feeding the plasma with the vapor of the element to be ionized. A practical problem with rare earth elements is that they are naturally found in an oxidized form that would require extremely high temperature to reach a decent vapor pressure (range of 10^{-2} to 10^{-3} mbar) for operation with an ECR. Therefore, we need to efficiently convert the available rare earth oxide into metal before using them with an ECR ion source.

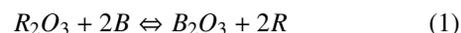
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Also of interest are primary beams of ^{82}Se , ^{106}Cd , and ^{204}Hg . Due to the relatively high volatility of these metals [1], it may be possible to control the flow of vapor into our plasma chamber with a simple setup that positions the sample outside the ion source and uses a variable leak valve and a heated transfer line to transmit the vapor to the plasma chamber. This was demonstrated recently in the case of mercury [2] where transfer of the vapor to the chamber and ionization by the ECR plasma lasted for about 150 hours.

CHEMICAL REDUCTION OF RARE EARTH SESQUIOXIDES

The chemical separation of metal oxides were investigated for the purpose of reducing calcium carbonate and calcium oxide (quicklime) into metal, and our procedure for the chemical conversion of the rare earth oxides is largely based on techniques developed for the benefit of the experimental program at NSCL. The rare earth metals (including scandium and yttrium) will readily oxidize into the sesquioxide, R_2O_3 [3], wherein R represents a rare earth element. The process to convert from the sesquioxide of samarium and ytterbium is known [4], and is reversible.



Metal B is the reagent to the reaction, and R is a rare earth metal. The chemical conversion will proceed in both directions if the temperature is sustained and the supply of materials allows. Extraction of a metal from the mixture occurs by diffusion with one metal leaving the mixture at a larger rate than the other. The reagent B is chosen to have a negligible vapor pressure in both elemental and oxide forms at the reaction temperature, to ensure minimal evaporation. According to [4] the oxides of ytterbium and samarium will undergo reactions with lanthanum reagent at $1350\ ^\circ\text{C}$ and $1200\ ^\circ\text{C}$ respectively. In the case of ytterbium sesquioxide the reduction-distillation will proceed at a temperature far below the oxide melting point. Furthermore, samarium and ytterbium metals are relatively stable in atmosphere unless heated in excess of $200\ ^\circ\text{C}$ [5] and allowed for handling of pure metal samples in atmospheric conditions.

Procedure

Our experimental set-up is constructed of two basic parts, the oven which heats the oxide-reagent mixture and collector plates which captures metal vapor diffusing from the oven. The chemical conversions were performed in a vacuum chamber that could reach pressures of about 10^{-7} mbar.

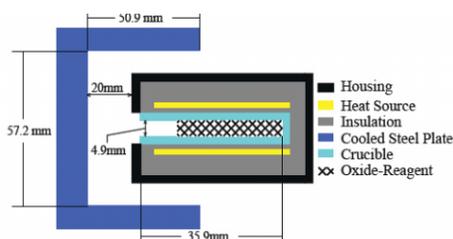


Figure 1: Schematic of reduction-distillation apparatus (not to scale) with a resistive oven, the oxide-reagent mixture may in general be heated by a resistive or inductive vaporization oven, and the more volatile rare earth metals distill out of the crucible and deposit on the steel plate.

In order to maximize the reduction efficiency an excess of lanthanum was added to samarium and ytterbium sesquioxides. We used a molar ratio of lanthanum to rare earth metal between 8:7 and 3:2 and for comparison R. E. Reed and W. B. Grisham used an 11:10 ratio [6]. The lanthanum was stored in an anaerobic environment and was exposed to atmospheric conditions only when mixing and weighing. The oxide and reagent were then mixed and placed into a cylinder open at one end and is used as a crucible. The oxide-reagent mixture was pressed by hand to the rear of the crucible with a metal rod and then loaded into our oven. Compression of the mixture ensures good contact between rare earth oxide and lanthanum reagent, producing the largest yield [4]. The setup is summarized in Figure 1.

Although, typically conversions are performed with a resistive oven, sometimes an inductively heated oven capable of reaching a higher temperature was also used. The temperature of our oxide-reagent mixture was calculated from calibration curves taken with a thermocouple placed inside the crucible, and are listed in table 1. Reductions were typically maintained at temperature 12-24 hrs to ensure a complete distillation of the metal sample onto the water cooled plates.

Results and Discussion

The distilled rare earth vapor was deposited onto 5 stainless steel plates (3 are shown in Fig. 1). The collection plates are screwed into water cooled blocks to prevent heating of the plates and subsequent evaporation of the rare earth sample. By using separate plates the deposited metal is easier to remove from the surface. The rare earth sample was scraped from the stainless steel plate with a razor. Table 1 exhibits efficiencies of rare earth sample collection by weight as a percent of the ideal yield. Both of our ytterbium runs were performed with the resistive oven and the oxide-reagent mixture was compressed by hand. An image of ytterbium metal as deposited on our plate assembly is shown in Fig. 2.

Our lowest efficiency, at 39 % for the element Sm, was performed on our inductive oven and is probably due to poor compression of the oxide-reagent mixture and incomplete reduction of the samarium sesquioxide. In this case the ge-



Figure 2: Ytterbium metal deposited on the collector plates.

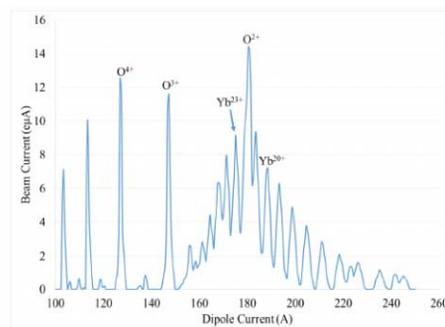


Figure 3: Natural ytterbium metal, reduced from the sesquioxide, as observed in ARTEMIS.

ometry of our inductive oven did not allow for effective manual compression of the oxide-reagent mixture. Reduction efficiencies up to 90 % were achieved in the case of samarium using the resistive oven with good compression of the oxide-reagent mixture. Collected samples were placed into the ARTEMIS plasma chamber and heated.

ARTEMIS is a 14 GHz Electron Cyclotron Resonance (ECR) ion source based on the Advanced ECR-Upgrade (AECR-U) at Berkley [7]. The plasma chamber has radial openings allowing for access ports and vacuum pumping. Microwave powers of up to 2 kW are attainable however, a maximum of about 800 W was employed to produce highly charged ions of samarium and ytterbium. For these measurements the axial magnetic field in ARTEMIS was about 1.8 T at injection and 0.8 T at extraction. A charge state distribution for Yb and Sm metal samples are shown in Fig. 3 and Fig. 4 respectively.

Contamination due to evaporation of lanthanum reagent with the rare earth metal could not be evaluated because our resistive ovens do not reach high enough temperatures on-line to vaporize lanthanum. However, deposition of the lanthanum reagent onto the collector plate is likely a small effect due to the fact 1760 °C is required to reach a vapor pressure of about 10^{-2} mbar [1] and the reduction of both Sm and Yb was maintained below 1500 °C as shown in Table 1. Trace analysis of a reduced sample by a technique such as mass spectroscopy in an inductively coupled plasma is an avenue of further investigation.

Oxidation of the rare earth metals while exposed to atmospheric levels of oxygen for approximately 1-5 hr while

Table 1: Table of chemical conversions by weight.

| Element | $R_2O_3(m)$ | La (mg) | R (mg) | Residues (mg) | Temperature (°C) | Efficiency % |
|---------|-------------|---------|--------|---------------|------------------|--------------|
| Yb | 353 | 353 | 208 | 433 | 1200 | 66 |
| Yb | 251 | 252 | 196 | 302 | 1300 | 87 |
| Sm | 214 | 270 | 167 | 290 | 1500 | 90 |
| Sm | 203 | 277 | 68 | 260 | 1340 | 39 |

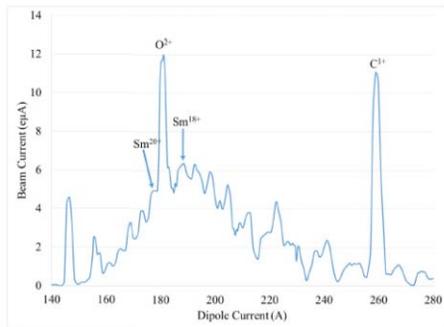


Figure 4: Natural samarium metal as obtained from reduction of the sesquioxide, as observed in ARTEMIS. The ragged peaks are from the mass spread of 10 amu in the natural abundance of samarium metal, with the most abundant isotope is ^{152}Sm at 26.75 % [8].

fabricating the alumina crucible for use in ARTEMIS is unclear. It is assumed some oxidation took place when exposing the samples to atmosphere and the photograph in Fig. 2 is representative of rare earth metal immediately after removal from vacuum. The rainbow of colors may be indicative of a thin oxide layer as is the case with bismuth [9].

Our experience in chemical reduction may be summarized in that larger temperatures and good compression of the oxide-reagent mixture produce the largest efficiencies. Furthermore, our results from ytterbium and samarium reduction are consistent with R. Reed and W. Girsham who reduced samarium oxide to metal over 6 hr with an efficiency of 94.9 % [6].

CADMIUM DEVELOPMENT

A technique was developed for the production of mercury ion beams on ARTEMIS. Mercury metal vapor was introduced into the plasma chamber, like a gas, though a variable leak valve and a heated transfer line. The ion source was operated using this setup for over 150 hours and we were able to achieve 4 eµA of $Q=30+$ with heating of about 60 °C. In addition the charge state distribution of mercury could be easily shifted toward higher or lower charge states by adjustment of the leak valve set-point [2]. With the aim to expand this technique, we sought to produce cadmium ions. A new heating scheme was developed to reach a temperature of up to 260 °C corresponding to a vapor pressure of 10^{-2} mbar in cadmium.

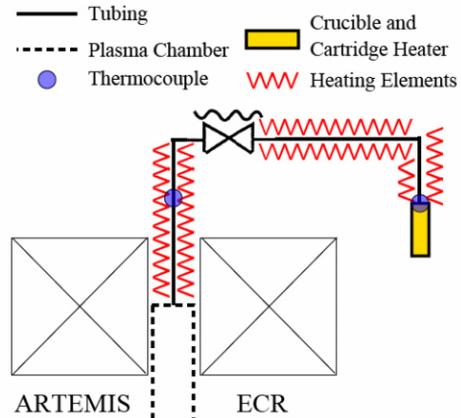


Figure 5: A schematic of the apparatus to produce cadmium vapor in reference to our ECR ion source. The cadmium sample is heated by a cartridge heater (yellow rectangle).

Oven Design

A schematic of the experimental set-up is shown in Fig. 5 in relation to our ion source. The vapor diffuses from the oven crucible to the plasma chamber through heated tubing and a bellows valve. The transfer line allows for vapor to enter the plasma chamber through the injection assembly on ARTEMIS. Connected to the transfer line is the sample reservoir which contains and heats the metal sample. The cadmium is loaded into a small stainless steel cylinder that is then sealed using a compression fitting onto the sample reservoir. The cadmium crucible and sample reservoir are visible in Fig. 6 without the cartridge heater installed.

The sample crucible was heated with a carbon cartridge filament and steel sheath screwed onto the bottom of the crucible. A K type thermocouple was wrapped around the cartridge heater sheath to provide an approximate sample temperature. The sample reservoir was wrapped in two heater tapes, as shown in Fig. 6, and then was wrapped in aluminum foil. The transfer line was heated with sections of threaded alumina tubing wrapped in filament wire. A second K type thermocouple was placed midway between two heaters to measure temperature as highlighted in Fig. 7. The transfer line is 79 cm in length and has an inner diameter of 4.8 mm.

Results and Discussion

Power to the transfer line heaters, heating tapes, and crucible cartridge heater could be independently controlled, and we maintained transfer line heaters and heater tapes

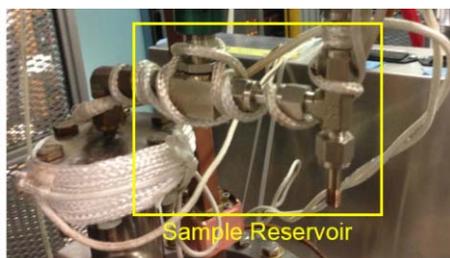


Figure 6: The sample reservoir as seen, without aluminum foil, exhibiting the heating tapes. In addition the bellows valves (green) and the sample crucible (on bottom) are shown. The crucible cartridge heater was removed for this photograph.



Figure 7: A photograph of the transfer line to transmit cadmium vapor through the ARTEMIS injection assembly with minimal losses. Heating elements are visible (white boxes) and an arrow points to our thermocouple (on right).

at a fixed setting while adjusting Cd vapor production by increasing the temperature of the crucible cartridge heater. The bellows valve isolating the transfer line and sample reservoir was fully opened for operation, and we used the ion source vacuum system to evacuate the sample reservoir.

A sample of natural cadmium was developed for the $Q=20+$ charge state. The most abundant isotope of cadmium, at 28.73 % [8], is atomic weight 114 and this isotope dominated our spectrum and was used to identify charge states. Cadmium was developed for 13 hrs with an average consumption of 1 mg/hr. The charge state distribution in Fig. 8 was obtained with a transfer line thermocouple reading of 140°C and a sample temperature of 260°C at a power consumption of 336 watts across all heating elements, with 65 % drawn by the heater tapes. Microwave power from the klystron was 500 W with a drain current of 1.2 mA at 23kV.

Initially an attempt was made to develop elemental cadmium using a variable leak valve similar to the one used

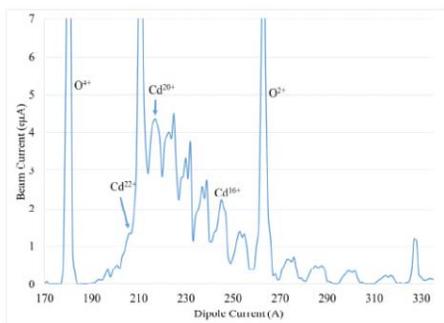


Figure 8: Cadmium as observed in ARTEMIS using the exterior oven technique.

previously with mercury. However, with cadmium we were unable to pass vapor through the leak valve, possibly due to cadmium vapor depositing inside the valve. A similar situation occurred when we tried to develop a selenium beam with the same setup using a variable leak valve, we observed excellent volatility of the selenium metal and residues were present throughout the sample reservoir but no metal was transmitted into the plasma. It is possible selenium vapor was stopped at the sapphire-copper interface inside the variable leak valve due to selenium-copper reactivity [5]. The variable leak valve was then replaced with a bellows valve as described above.

CONCLUSION

We demonstrated the efficient chemical conversion of ytterbium and samarium from the sesquioxide into elemental form with efficiencies up to 90 % without modification to our standard resistive oven. We were able to positively identify Yb and Sm samples by vaporizing these metals into ARTEMIS and observing the species distribution of the plasma. In addition we proved the concept of an exterior oven design previously used to provide mercury vapor is applicable to a metal such as cadmium. A beam current of $4.3 \mu\text{A}$ of Cd^{20+} was measured for a sample temperature of 260°C and, the exterior oven may be useful for developing selenium beams from the metal or dioxide.

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