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Nuclear Structure Measurements of Fermium-254 and Advances in Target Production Methodologies

By
Oliver Ralf Gothe

A dissertation submitted in partial satisfaction of the requirements for the degree of Doctor of Philosophy in Chemistry in the Graduate Division of the University of California, Berkeley

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Abstract

Nuclear Structure Measurements of Fermium-254 and Advances in Target Production Methodologies

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Oliver Ralf Gothe

Doctor of Philosophy in Chemistry

University of California, Berkeley

Professor Joseph Cerny, Chair

The Berkeley Gas-filled Separator (BGS) has been upgraded with a new gas control system. It allows for accurate control of hydrogen and helium gas mixtures. This greatly increases the capabilities of the separator by reducing background signals in the focal plane detector for asymmetric nuclear reactions. It has also been shown that gas mixtures can be used to focus the desired reaction products into a smaller area, thereby increasing the experimental efficiency.

A new electrodeposition cell has been developed to produce metal oxide targets for experiments at the BGS. The new cell has been characterized and was used to produce americium targets for the production of element 115 in the reaction $^{243}$Am($^{48}$Ca, 3$n$)$^{288}$115. Additionally, a new method of producing targets for nuclear reactions was explored. A procedure for producing targets via Polymer Assisted Deposition (PAD) was developed and targets produced via this method were tested using the nuclear reaction $^{208}$Pb($^{40}$Ar, 4$n$)$^{244}$Fm to determine their in-beam performance. It was determined that the silicon nitride backings used in this procedure are not feasible due to their crystal structures, and alternative backing materials have been tested and proposed.

A previously unknown level in $^{254}$Fm has been identified at 985.7 keV utilizing a newly developed low background coincident apparatus. $^{254}$Fm was produced in the reaction $^{208}$Pb($^{48}$Ca, 2$n$)$^{254}$No. Reaction products were guided to the two-clover low background detector setup via a recoil transfer chamber. The new level has been assigned a spin of $2^-$ and has tentatively been identified as the octupole vibration in $^{254}$Fm. Transporting evaporation residues to a two-clover, low background detector setup can effectively be used to perform $\gamma$-spectroscopy measurements of nuclei that are not accessible by current common methodologies. This technique provides an excellent addition to previously available tools such as in-beam spectroscopy and $\gamma$-ray tracking arrays.
This thesis is dedicated to Professor Heino Nitsche who was my teacher, friend, and guide through graduate school. You will be missed.
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List of Acronyms

A  Atomic Mass Number
ADAM  Advantech Data Acquisition Module
ADC  Analog to Digital Converter
BASE  Berkeley Accelerator Space Effects
$B_n$  Neutron Binding Energy
$B_f$  Fission Barrier
BGO  Bismuth germanate
BGS  Berkeley Gas-filled Separator
$B\rho$  Magnetic Rigidity
C3  Clover Corner Cube Detector
CN  Compound Nucleus
DOE  Department of Energy
DSSD  Double-Sided Silicon Strip Detector
ECR  Electron Cyclotron Resonance
EVR  Evaporation Residue
FPD  Focal Plane Detector
FWHM  Full Width Half Maximum
GRC  Graduate Research Conference
GRETINA  Gamma Ray and Energy Tracking Array
GSI  Gesellschaft für Schwerionenforschung
JINR  Joint Institute for Nuclear Research
LBNL  Lawrence Berkeley National Laboratory
M1  Gradient-Field Dipole Magnet
M2  Flat-Field Dipole Magnet
MBS  Multi-Branch System

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TOF  Time-of-Flight
PID  Proportional-Integral-Derivative
PIN  Positive-Intrinsic-Negative
PTFE  Polytetrafluoroethylene
Q1  Quadrupole Magnet
RGA  Residual Gas Analyzer
RIA  Rare Isotope Accelerator
RTC  Recoil Transfer Chamber
SRIM  Stopping and Range of Ions in Matter
\( \sigma \)  Cross Section
VENUS  Versatile ECR ion source for Nuclear Science
Z  Atomic Number
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Chapter 1

Introduction

In the past fifteen years, the study of the nuclear structure of the heaviest elements has advanced greatly. Many highly sophisticated detection setups coupled with $\gamma$-ray detector arrays have been developed and constructed to explore the rotational structure, K-isomerism, and single particle excited states of the heaviest elements. In recent years, increasing emphasis has been placed on studying the extremes of nuclear stability. Neutron deficient actinides are such an extreme region. However, the study of many neutron deficient actinides is not only limited by particle stability along the proton drip line, but also short half-lives, very small production cross sections, and their decay signatures. These limitations do not necessarily apply to all the neutron deficient actinides but many of them remain unstudied due to the fact that they often decay via electron capture, a particularly difficult decay to detect. High $\gamma$-ray background rates at heavy element production facilities prohibit the detection of the characteristic x-ray in this type of decay.

Combining ever more efficient separators with higher beam intensities has opened up many new isotopes for study. However, the study of their nuclear properties is still difficult. By moving reaction products from the high $\gamma$-ray and neutron background at their production site, the study of many of these isotopes with ever lower production rates has been made possible. Exploring and understanding nuclei at these extremes of the nuclear landscape has always been significant in advancing and constraining nuclear models. The heaviest elements are particularly interesting as they may provide clues pointing towards the ”fabled” Island of Stability, a region believed to contain spherical nuclei with long half-lives. Theoretical predictions currently disagree on the details of where this region of stability should be centered. Some of the different predictions are described in more detail by Minaya Ramirez et al. [MAB$^+$12].

Figure 1.1 below shows the region of the table of isotopes that has recently become accessible for nuclear structure studies to the Heavy Element Group at the 88-inch cyclotron. This chapter discusses the 88-inch cyclotron, the Berkeley Gas-filled Sep-
Figure 1.1: Table of isotopes highlighting the neutron deficient actinides that have recently become accessible for nuclear structure measurements. Isotopes shaded in blue primarily decay by electron capture, isotopes shaded in yellow primarily decay by α-decay, isotopes shaded in red decay by β−, and isotopes shaded in green decay by spontaneous fission. The decay mode of those shaded in grey is unknown.

The 88-inch cyclotron is a sector-focused cyclotron that can accelerate both light and heavy ions required for the production of trans-fermium nuclei. Figure 1.2 shows a schematic of a standard cyclotron. Ions are accelerated in spiral paths from the center outwards. This is accomplished by applying rapidly varying electric fields, synchronized with the ions orbital frequency, to the D-shaped electrodes between...
the north and south pole of the electromagnet. A bunched beam is injected near the center along an inner circular orbital and is accelerated in the gaps between the electrodes on each pass.

Figure 1.2: Schematic of a cyclotron showing the electromagnet and D-shaped electrodes. Alternating current is applied to the electrodes to accelerate the bunched ions between the electrode gaps.

The frequency, \( f \), of the voltage applied to the D-shaped electrodes is determined by the cyclotron frequency:

\[
  f = \frac{q_i B}{2\pi m_i},
\]

where \( B \) is the static magnetic field between the two poles of the electromagnet shown in figure 1.2, and \( q_i \) and \( m_i \) are the charge and mass of the ions which is to be accelerated. This equation is easily derived by equating the centripetal and magnetic Lorentz forces. The beam is accelerated only when it passes through the gap between the two D-electrodes. In some cases a relativistic correction factor shown below in equation 1.2 has to be applied to the cyclotron frequency:

\[
  f = \frac{qB}{2\pi m_i} \sqrt{1 - \left(\frac{v_i}{c}\right)^2}
\]

Here, \( m_i \) is the mass of the ion, \( v_i \) is its velocity and \( c \) is the speed of light.

Current studies at the 88-inch cyclotron are focused on heavy element research, \( \gamma \)-spectroscopy, nuclear structure, astrophysics, fundamental interactions, symmetries, and technology research and development. Currently, the 88-inch cyclotron is most known for housing the Berkeley Gas-filled Separator (BGS), the Berkeley Accelerator Space Effects (BASE) facility, and the versatile Electron Cyclotron Resonance (ECR) ion source for Nuclear Science (VENUS), a model source for the Rare Isotope Accel-
1.1.1 Electron Cyclotron Resonance Ion Sources

Most high intensity ion sources, and all the ones currently in use at the 88-inch cyclotron, are based on a phenomenon called electron cyclotron resonance. In order to provide a beam of the required energy, a cyclotron requires an incoming stream of charged ions. These ions are most commonly provided by sources based on this resonance. In an ECR source, the desired atoms are first brought into their gas phase and then confined in a specially designed magnetic field. They are ionized through collisions with electrons which have been accelerated via microwaves in this magnetic bottle. In a static magnetic field, energetic electrons gyrate around the field lines due to the Lorentz force as described by:

\[ F = -q_e (v_e \times B) \]  

where \( F \) is the force experienced by a particle with velocity \( v_e \) and charge \( q_e \) in a magnetic field \( B \). The cyclotron frequency, \( \omega \), is the angular frequency of the charged particles around the field lines and is defined as:

\[ \omega = \frac{eB}{m_e} \]  

where \( e \) and \( m_e \) are the charge and mass of an electron respectively and \( B \) is the magnetic field. For high energy cases, this formula has to be adjusted for the effects predicted by the special theory of relativity to the following:

\[ \omega = \frac{eB}{\gamma_r m_e} \]

Where \( \gamma_r \) is the relativistic mass correction factor, \( \sqrt{1 - \frac{v^2}{c^2}} \), and \( m_e \) is the electron mass. When microwaves are injected, energy is added to the electrons in regions where the \( B \)-field is in resonance with the cyclotron frequency. Inside the plasma, the heated electrons collide with the gas molecules and successively ionize them. The ions are then extracted and the desired charge state is selected by means of a bending magnet. Lighter elements such as oxygen may be extracted in a 3+ charge state, whereas heavier elements such as uranium might have a charge of 32+. A general schematic of ECR ion sources illustrating their operating principle is shown in figure 1.3.

Ion sources generally have a set frequency and this number is quoted when describing the source. The 88-inch cyclotron is equipped with two ECR high-charge-ion sources \([Wu00]\); the double-frequency (14 GHz and 10-13 GHz) AECR-U and the 6.4 GHz Lawrence Berkeley National Laboratory (LBNL) ECR. Other institutions
have their own ion sources run at different frequencies based on their magnet setup. Higher frequencies allow for higher plasma densities, leading to higher charge states and beam intensities, and thus over the last decades the cyclotron frequency of new sources has steadily been increasing, requiring bigger and stronger magnets for plasma containment.

Currently, the VENUS ECR at 28 GHz is one of the highest frequency ECRs. This higher frequency was made possible by utilizing superconducting magnets and allows the production of ion beams at intensities up to an order of magnitude larger than from the AECR-U and LBNL ECR. Once accelerated to the correct energy, these ions can now be used in nuclear reactions.

1.2 Nuclear Reactions

A nuclear reaction is a process in which two nuclei interact to produce products different from the original nuclei. In the reactions treated in this dissertation this is accomplished by accelerating beam nuclei at the 88-inch Cyclotron at LBNL, guiding them with bending magnets to the target chamber of the BGS where they hit targets placed on either a stationary or a rotating target wheel. An example of such a reaction is: $^{208}\text{Pb}(^{48}\text{Ca}, 2n)^{254}\text{No}$ where a $^{48}\text{Ca}$ beam impinges on a $^{208}\text{Pb}$ target to produce the nucleus $^{254}\text{No}$ and two neutrons. The products of the nuclear reaction are then separated from the beam nuclei and various other reaction products in the BGS as discussed in section 1.6. They are then analyzed in silicon strip detectors, or, alternatively, transported to a chemistry setup or to a nuclear structure apparatus via a recoil transfer chamber (RTC).

Production of exotic isotopes in nuclear reactions makes it possible to probe regions of the chart of isotopes that are very unstable. Advancing our knowledge of
nuclear reactions is of particular interest as it will help to constrain nuclear models through additional data and may, for example, eventually lead to a better understanding of how elements were produced during nucleosynthesis [SL90].

1.2.1 Beam Energies Required for Nuclear Reactions

A nuclear reaction can generally only be successful if the beam projectiles have sufficient energy to overcome the Coulomb barrier. The amount of energy necessary to form the reaction products due to differences in binding energies is known as the $Q$-value. Any additional energy imparted by the beam populates high lying excitation states in the compound nucleus. These states rapidly de-excite by emitting particles and $\gamma$-rays via one of many pathways known as exit channels. In fusion evaporation reactions the neutron exit channels are generally of most interest. The number of neutrons evaporated by the compound nucleus can be controlled via the amount of excitation energy supplied in addition to the $Q$-value. The energy required for the formation of the compound nucleus at the center of the target is determined through a simple mass defect calculation. For example, when considering the reaction $^{208}\text{Pb}(^{48}\text{Ca},2n)^{254}\text{No}$ the energy required for the formation of the $^{256}\text{No}$ compound nucleus is calculated from the mass defects ($\Delta$) of all involved isotopes as follows:

$$Q = \Delta(^{208}\text{Pb}) + \Delta(^{48}\text{Ca}) - \Delta(^{256}\text{No})$$

(1.6)

For the above reaction the $Q$-value is 153.8 MeV. Next, the appropriate amount of energy, generally experimentally determined, must be added to obtain the desired exit channel, yielding the center of mass reaction energy, $E_{cm}$. If such data are not available, it is generally accepted to add the binding energy of the neutron ($B_n$) and 2 MeV of kinetic energy per evaporated neutron. 22 MeV have to be added to obtain a total energy of 175.8 MeV to evaporate two neutrons from $^{256}\text{No}$. This energy is calculated in the center-of-mass frame and has to be boosted into the laboratory frame. The beam reaction energy in the laboratory frame, $E_{lab}$, becomes:

$$E_{lab} = E_{cm} \frac{m_{cn}}{m_{target}}$$

(1.7)

where $E_{cm}$ is the energy supplied to the nuclear reaction in the center-of-mass frame, and $m_{cn}$ and $m_{target}$ are the masses of the beam and compound nucleus. This relation can easily be derived from momentum and energy conservation. For the calcium plus lead reaction above this laboratory frame energy is 216.4 MeV. A more detailed derivation of the total beam energy can be found in Modern Nuclear Chemistry [LMS05].

Thus far the center of target energy has been calculated. After this energy has been determined, it is now necessary to correct it for the energy that is lost by the beam particles as they pass through materials on the way to the target. Such materials
may include, the target backing, windows separating the beam line and the target chamber, and half the target material. At first glance such calculations may appear complicated but there is a simple trick that allows the extrapolation of the additional energy required due to the energy loss in these materials. For this extrapolation, a program called Stopping and Range of Ions in Matter (SRIM) [Zie04] is used to determine the average range of the beam particles in the material at the energy of interest. SRIM is a set of programs which calculate the stopping power and range of ions in matter using a quantum mechanical treatment of ion-atom collisions. Fitting the obtained ranges with a linear fit, as seen in figure 1.4, yields a linear equation that can be used to extrapolate the energy needed before passing through the interfering material, in order to correct for the energy lost in the material.

Figure 1.4: Range of $^{48}$Ca in PbO$_2$ as a function of energy. These data were obtained from SRIM and were used to estimate the energy loss of a calcium beam in the lead target material.

This is accomplished by rearranging the best fit line to calculate the energy of the beam particles as a function of range after passing through the material, then adding the thickness of the material, and calculating the original energy. Equation 1.8 below is the result of such a calculation for the reaction discussed above.

$$E_{\text{corrected}} = R^{-1}(R(E_{\text{lab}}) + \text{Thickness}_{\text{material}})$$  \hspace{1cm} (1.8)

where $R(E)$ is the best fit equation determined in Figure 1.4 that gives the range as a function of Energy, $R^{-1}(E)$ is the inverse of that function. Thus considering the calcium beam passing through 0.25 $\mu$m of lead oxide target material (half of the total target material) yields a corrected energy of 218.6 MeV. Table 1.1 summarizes all the calculations taken to arrive at the final energy. Thus a projectile energy of approximately 219 MeV should be used in order to produce $^{254}$No in this reaction.
<table>
<thead>
<tr>
<th>Quantity</th>
<th>Energy [MeV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Q$-value</td>
<td>153.8</td>
</tr>
<tr>
<td>$E_{cm}$ ($Q$-value + 2 neutron evaporation energy)</td>
<td>175.8</td>
</tr>
<tr>
<td>$E_{lab}$</td>
<td>216.4</td>
</tr>
<tr>
<td>$E_{lab}$ corrected for energy loss in the target material</td>
<td>218.6</td>
</tr>
<tr>
<td>$E_{lab}$ corrected for energy loss in the target backing</td>
<td>219.3</td>
</tr>
</tbody>
</table>

Table 1.1: Summary of calculations necessary to determine the correct beam energy for the nuclear reaction $^{208}\text{Pb}(^{48}\text{Ca},2n)^{254}\text{No}$.

### 1.2.2 Nuclear Cross Sections

Even given the ideal reaction energy, nuclear reactions occur at vastly different rates. The rate at which nuclear reactions occur is represented by the cross section, $\sigma$. Cross sections are measured in units of barns ($10^{-24} \text{cm}^2$) and they represent a geometrical probability of the beam overlapping and interacting with the target. The cross section can be used to calculate the expected production rates of nuclear reactions given a particular beam energy and intensity and target thickness.

Nuclear reactions are performed under varying conditions; however, the cross section is absolute and can be used to compare experiments performed at different institutions or at different times. For example, a reaction run at LBNL might be using targets that are of a different thickness than those of an experiment being run at Gesellschaft für Schwerionenforschung (GSI). Essentially the cross section is the rate of the reaction, scaled by the product of target thickness and beam intensity:

$$\sigma = \frac{R}{\Phi \ast N}$$  \hspace{1cm} (1.9)

where $R$ is the reaction rate, $\Phi$ the beam current in units of Hz, and $N$ the target thickness in units of $\frac{\text{atoms}}{\text{cm}^2}$ [Kra88]. In the BGS the product of beam intensity and target thickness are measured via Rutherford scattering, described in section 3.23, in the target. There are two Rutherford detectors at an angle of $\pm 27^\circ$ with respect to the beam direction, Rutherford West and Rutherford East. Before reaching the Positive-Intrinsic-Negative (PIN) diode detectors, the Rutherford scattered beam particles pass through a 4.78 mm diameter collimator as well as attenuating foils with regularly spaced holes that reduce the intensity by a factor of 1348 $\pm$ 20 [Fol04]. This attenuation is necessary to avoid radiation damage to the Rutherford detectors. The attenuation coefficients were remeasured in an experiment described in chapter 3. The distance between the target and the collimator is 292 mm. The detectors, therefore, subtend a solid angle, $\Omega$, $2.1 \ast 10^{-4}$ steradians based on the following calculation in the limit that $Col_r << d$:  

8
\[ \Omega = 4\pi \frac{\pi C_{r}^2}{4\pi d^2} = \frac{\pi C_{r}^2}{d^2} \] (1.10)

where \( C_{r} \) is the radius of the collimator and \( d \) is the distance between the target and the collimator. This quantity represents the total solid angle observed by the Rutherford detector and can be used to measure the product of beam intensity and target thickness based on the Rutherford cross section equations 1.11 and 1.12 taken from [Seg77]:

\[
d\sigma = \left( \frac{e^2 Z_b Z_t}{2E_{lab}} \right)^2 \sin^2(\theta) \frac{\left( \cos(\theta) + \sqrt{1 + \frac{A_b^2}{A_t^2} \sin^2(\theta)} \right)^2}{\sqrt{1 - \frac{A_b^2}{A_t^2} \sin^2(\theta)}}
\] (1.11)

where the variables are defined in table 1.2.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>( e )</td>
<td>Elementary charge</td>
</tr>
<tr>
<td>( Z_b )</td>
<td>Proton number of the beam</td>
</tr>
<tr>
<td>( Z_t )</td>
<td>Proton number of the target</td>
</tr>
<tr>
<td>( E_{lab} )</td>
<td>Beam energy in the laboratory frame</td>
</tr>
<tr>
<td>( \theta )</td>
<td>Angle between beam and Rutherford detectors</td>
</tr>
<tr>
<td>( A_b )</td>
<td>Mass number of the beam</td>
</tr>
<tr>
<td>( A_t )</td>
<td>Mass number of the target</td>
</tr>
</tbody>
</table>

Table 1.2: Definition of variables used in equation 1.11.

The integrated product of beam intensity and target thickness, is related to this equation by:

\[ N\Phi t = N_{Robs} \frac{ScaleDown}{d\sigma \Omega} \] (1.12)

where \( N \) is the number of scattering centers per unit area and \( \Phi \) is the number of beam particles per unit time. During a typical BGS run the Rutherford rate has to be reduced with a \( ScaleDown \) in order not to damage the PIN diodes. The Rutherford rate gives a direct measure of the product of beam intensity and target thickness, which can then be substituted into equation 1.9.

\[ \sigma = \frac{N_{obs}}{i\epsilon_{BGS}\epsilon_{det}} \frac{1}{N\Phi} = \frac{N_{obs}}{\epsilon_{BGS}\epsilon_{det}} \frac{d\sigma \Omega}{N_{Robs}ScaleDown} \] (1.13)

Note that \( \sigma \), the cross section given \( N_{obs} \) observed events, here represents the cross section of the nuclear reaction, whereas \( d\sigma \) represents the cross section of Rutherford scattering of the beam in the target material. \( \epsilon_{BGS} \) and \( \epsilon_{det} \) are the efficiencies of the
BGS and detector setup. The following errors have been measured and estimated in determining nuclear cross sections at the BGS.

**Systematic Errors in Cross Section Measurements**

The systematic errors within the BGS have contributions from five different aspects [GLP+05][Gat08]:

1. The uncertainty in the beam energy provided by the 88-inch cyclotron is estimated to be 1% which results in approximately 2% uncertainty in the Rutherford scattering cross sections.

2. A systematic uncertainty of 10% in the efficiency of the BGS has been estimated by Gregorich et al. [GLP+05] for the transport of EVRs from the target to the detector.

3. The error in the angle of the Rutherford detectors is 0.2° with respect to the beam axis. This translates to a systematic uncertainty of 3%.

4. The systematic uncertainty in the collimator area is estimated to be 4% based on the uncertainty in the measurement of the radius.

5. The error caused by the attenuation screens for the Rutherford detectors is estimated to be 5%.

Standard error propagation sums these contributions in quadrature into a total systematic error of 12%.

### 1.3 The Berkeley Gas-filled Separator (BGS)

The BGS is a system designed to selectively separate the desired products of nuclear reactions based on magnetic rigidity. Magnetic rigidity is the momentum per unit charge of a particle; a measure of the particles resistance to having its flight path bent in a magnetic field. Magnetic rigidity is defined as:

\[
B\rho = \frac{mv}{\bar{q}}
\]  

(1.14)

where \(B\) is the magnetic field, \(\rho\) is the bending radius due to the magnetic field, and \(m, v, \bar{q}\) are the mass, velocity and average charge of the particle. The bending radius to the detector system and the magnetic field are both fixed in the BGS. The average charge is a function of the atomic numbers of the ion and the fill gas, as well as the ion velocity and fill gas pressure as described in more detail by [GYL+88]. The nuclear
products which are produced in several different charge states collapse to the average charge state, $\bar{q}$, through multiple charge-changing collisions with the fill gas.

![Figure 1.5: Illustration of the ion path. Left: a vacuum separator; Right: Charge averaging due to collisions with the fill gas. These paths are for illustrative purposes only and are not based on simulation or calculation.](image)

The average charge is estimated via a semi-empirical formula as described previously by Gregorich et al. [GLP+05]. In general, faster moving ions attain a higher charge state. The charge state of unreacted beam, reaction transfer products, and elastically scattered target atoms is, therefore, higher than that of evaporation residues because they are moving at significantly higher velocity. Therefore, they have a lower $B\rho$ and by altering the field strength of the bending magnets, the $B\rho$ of the evaporation residue can be guided to the detector assembly and efficiently separated from unwanted byproducts. The Evaporation Residues (EVRs) are thus focused as seen in figure 1.5. This operational principle is illustrated in figure 1.6.

As previously described, the 88-inch cyclotron provides the required beam. The projectiles are guided towards the target area through a beam line held at approximately $10^{-6}$ torr. The beam line is separated from the target chamber by a differential pumping system that steps the pressure up to 0.2 - 0.9 torr of helium and/or hydrogen in the target chamber and the BGS. In most cases a rotating target wheel is mounted in the target chamber. Four banana shaped target segments that are most often produced via electrodeposition, vapordeposition, or sputtering are mounted to the target wheel. Target production methods will be treated in great detail in chapter 2. After passing through the target, the products recoiling from the target, and unreacted beam reach the BGS magnets.

### 1.3.1 BGS Magnets

The force acting on moving ions is known as the Lorentz force. This force is, in general, described by the equation:

$$\vec{F} = q[\vec{E} + (\vec{v} \times \vec{B})]$$

(1.15)
where $\vec{F}$ is the force felt by the moving ion, $q$ and $\vec{v}$ are its charge and velocity, and $\vec{E}$ and $\vec{B}$ are the electric and magnetic fields. Since the BGS is purely a magnetic separator the electric component can be ignored and the motion of the ions is purely governed by the B-field within the BGS. Furthermore, since the magnetic field is perpendicular to the velocity of the reaction products equation 1.15 simplifies to:

$$F = qvB$$  \hspace{1cm} (1.16)

Note that by balancing the Lorentz force to the centripetal force of the ions one easily obtains the relation for magnetic rigidity shown in equation 1.14

In the BGS ions are guided and focused by three magnets. The first magnet directly after the target box is a quadrupole magnet (Q1) that focuses the EVRs vertically. This vertical focusing also results in horizontal defocusing. The quadrupole magnet is placed directly after the target area in order to improve the angular acceptance of the BGS. Products can be accepted in a cone formed $\pm 9\%$ in the vertical and $\pm 4.5\%$ in the horizontal direction. This corresponds to approximately $4.5 * 10^{-2}$ steradians. Reaction products are guided towards the focal plane detectors by two dipole magnets, D2 and D3. The magnetic field strength in all three electromagnets can be set by controlling the current they are supplied with. Figure 1.7 shows the relative geometric arrangement of the three magnets. The separation of reaction products is accomplished mainly in the first dipole magnet, D2. The BGS has a very large bending angle of $70^\circ$ which leads to large dispersion, resulting in an horizontal displacement of 1.81 cm [Gre13] for a 1% change in magnetic rigidity.
1.3.2 Detection of Nuclear Reaction Products

Once a desired nuclear reaction has occurred, the excited compound nucleus rapidly evaporates neutrons before it recoils into the BGS which guides it towards the focal plane of the detector box. In the detector box, recoils first pass through a MultiWire proportional counter before implanting in the Clover Corner Cube Detector (C$^3$) detector array. The MultiWire proportional counter can be used to veto implantation-like signals that passed through it within a given time window of the event. The C$^3$ detector array consists of three 64 mm by 64 mm silicon strip detectors combined with a silicon detector crown. The detector without the crown is shown in figure 1.8. The front face of a clover detector can be placed behind the silicon chips. The C$^3$ was designed to optimize the efficiency for detecting $\gamma$ and x-rays in coincident with $\alpha$-decays. For the study of neutron deficient actinides a RTC replaces the C$^3$ detector array. It separates the BGS from a small chamber held between one and two bars of helium in order to thermalize and then transfer reaction products via a KCl gas jet. The RTC is described by Düllman et al. in great detail [DFG+05] and will be covered in section 3.4.

Even though the BGS is ideal for separating EVRs from many reactions, there are some classes of reactions, such as highly asymmetric reactions, for which it has previously not been a very effective separator. In such reactions their was often very
Figure 1.8: Image of the BGS focal plane detector showing the relative geometrical arrangement of the silicon chips.

high background in the silicon detector due to elastically scattered target material prohibiting the study of such reactions. In order to overcome these problems the BGS had to be upgraded.

1.4 Upgrades to the BGS

As heavy element science, and particularly production of new isotopes is moving towards ever smaller, sub pico-barn cross sections, it is becoming ever more important to achieve high separation efficiency in order to accurately identify rare events with short half-lives. In order to achieve increased separation as well as lower background the BGS fill gas was upgraded. During this process a new target wheel speed control system was implemented into the same hardware components necessary for the upgrade of the gas system.

1.4.1 BGS Gas Control System Upgrade

The BGS has very good transmission for nuclear reaction products for more symmetric reactions such as $^{242}\text{Pu}^{(48\text{Ca},3n)^{287}\text{Cn}}$. However, for highly asymmetrical re-
actions, reactions in which the target and beam atoms differ greatly in mass, such as $^{242}\text{Pu}(^{22}\text{Ne},3n)^{261}\text{Rf}$ problems often arise from the fact that the magnetic rigidities for elastically scattered target atoms and EVRs are similar. Figures 1.9 illustrates an example of this problem: the red curve shows the magnetic rigidity of scattered plutonium atoms from the target, the green curve shows the magnetic rigidity of the compound nucleus evaporation residue product of the asymmetric reaction neon + plutonium, whereas the blue curve shows the magnetic rigidity of the EVRs from a calcium + plutonium reaction. The velocity of scattered target atoms and EVRs of

![Graph showing magnetic rigidities](image)

Figure 1.9: Helium fill gas: Comparison of the magnetic rigidity of the reaction products of an asymmetric (green), a symmetric (blue), and of elastically scattered (ES) target material (red). Arrows show the actual velocities and magnetic rigidities for calcium + plutonium and neon + plutonium reactions, respectively.

the different reactions is calculated through reaction kinematics [Gre13] and shown on the graph with arrows. Unlike the case of the calcium + plutonium reaction, the magnetic rigidity for elastically scattered target atoms and EVRs in the neon + plutonium reaction are very similar even though they have vastly different velocities. By comparison the same reactions would have the magnetic rigidities shown in figure 1.10 if pure hydrogen were used as a fill gas. Since higher concentrations of hydrogen require stronger magnetic fields, mixtures with a hydrogen content of higher than 30% were not possible. The required magnetic rigidity settings would require magnetic currents that exceed the capabilities of the BGS magnets.

The drop of magnetic rigidity in helium gas below 1.5 $v_0$ makes studying asymmet-
Figure 1.10: Hydrogen fill gas: Comparison of the magnetic rigidity of the reaction products of an asymmetric (green), a symmetric (blue), and of elastically scattered (ES) target material (red). Arrows show the actual velocities and magnetic rigidities for calcium + plutonium and neon + plutonium reactions, respectively.

Reactions very difficult because frequently the magnetic rigidity of the elastically scattered target atoms is similar to that of the EVRs and they overwhelm the detectors with background signals. A combination of these two gases should correct the drop of magnetic rigidity at low EVR velocities. Using a mixture of hydrogen and helium at the appropriate ratio greatly reduces the background and makes the study of these reactions possible with almost no elastically scattered target atoms hitting the detector. In this experiment gas mixtures as outlined in Table 1.3 were compared to illustrate this improved separation.

<table>
<thead>
<tr>
<th>Gas Mixture</th>
<th>Helium</th>
<th>Hydrogen</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>87%</td>
<td>13%</td>
</tr>
<tr>
<td>2</td>
<td>79%</td>
<td>21%</td>
</tr>
<tr>
<td>3</td>
<td>70%</td>
<td>30%</td>
</tr>
</tbody>
</table>

Table 1.3: List of the relative gas concentrations for the three tested helium/hydrogen gas mixtures.
The magnetic rigidity of the studied mixture can be estimated by taking a linear combination of two magnetic rigidity curves weighted by their probability of charge exchange in the gas mixture. Applying this procedure to hydrogen and helium yields the dependencies of magnetic rigidity on velocity shown in 1.11.

Figure 1.11: Linear combinations of the H\textsubscript{2} and He equations for B\rho. Velocity and magnetic rigidity values are shown for the asymmetric reaction that cannot be run with pure helium showing that the gas mixture can be chosen such that elastically scattered (ES) material can easily be separated from the EVRs.

Figure 1.11 shows that much lower EVR velocities can be studied before the magnetic rigidities trend towards zero. Additionally, at velocities between 1v\textsubscript{0} and 1.6v\textsubscript{0} the ratio of the gases can be chosen in such a way that the magnetic rigidity stays mostly constant. This effectively focuses the reaction products into a smaller area on the detector and thus increases efficiency.

Technical Aspects of the Gas Mixture Control System

The gas mixture was controlled with a new gas control system based on the RS-485 communication protocol. Two mass flow controllers, type MKS-1179, were used to control the hydrogen and helium flow into the BGS. The controllers are continuously monitored from the BGS counting shack via a LabVIEW interface and the composition of the fill gas can be verified with a Residual Gas Analyzer (RGA). The control box for the mass flow controllers was designed to easily be expandable for
additional controllers as well as relays, voltage in and outputs. The gas system is capable of gas flow rates ranging from 0 and 200 sccm. The RS-485 signal is also sent to two Advantech Data Acquisition Module (ADAM) 4060 units for relay control, one ADAM 4017 unit for voltage input, and one ADAM 4024 unit for voltage output. An additional port on the front of the unit allows for daisy-chaining additional RS-485 components to the current control system. The front panel of the gas control box is shown in figure 1.12.

![Front panel of the gas control box showing the connectors for gas controllers as well as the additional output and input signals that can be controlled from the LabVIEW interface.](image)

The current version of the LabVIEW interface is capable of controlling the ratios of the gas mixture as well as the flow rate of gas into the BGS. The hardware that the LabVIEW program interfaces with is designed to easily be adaptable to new requirements that come up during new experiments. Currently there are eight relay outputs, 2 voltage outputs ranging from 0 to 10 Volts and 2 voltage inputs available. The LabVIEW program can easily be adjusted to control these outputs in order to control equipment in the BGS from the counting shack. For example, one voltage input and output is currently utilized to control the rotation speed of the target wheel.

Figure 1.13 shows the three power supplies providing 24V, +15V and -15V to the control units as well as the power distribution board. The 24V supply is necessary for powering the ADAM units, whereas the +/- 15V supply powers the Mass Flow Controllers. Figure 1.14 shows the ADAM units within the box. Each ADAM unit is assigned a unique address between 0 and 99 in order to address RS-485 commands to specific units. Table 1.4 shows the addresses currently assigned to the units.

### 1.4.2 Production of $^{258}\text{Rf}$ via the $^{22}\text{Ne} + ^{242}\text{Pu}$ reaction

The $^{242}\text{Pu}^{(22}\text{Ne}, 6n)^{258}\text{Rf}$ reaction was chosen to test this newly developed gas mixture system for the following reasons:

1. Both the recoiling target atoms and the EVRs have a similar magnetic rigidity
in helium. $^{258}\text{Rf}$ has a $v/v_0$ of approximately 1.3 and the elastically scattered target atoms have a $v/v_0$ 2.8. As seen in figure 1.9, this gives them magnetic
<table>
<thead>
<tr>
<th>Unit</th>
<th>Address</th>
<th>Inputs/Outputs</th>
</tr>
</thead>
<tbody>
<tr>
<td>ADAM-4060</td>
<td>02</td>
<td>4 Relays</td>
</tr>
<tr>
<td>ADAM-4060</td>
<td>03</td>
<td>4 Relays</td>
</tr>
<tr>
<td>ADAM-4017</td>
<td>04</td>
<td>4 Voltage Outputs</td>
</tr>
<tr>
<td>ADAM-4024</td>
<td>06</td>
<td>8 Voltage Inputs</td>
</tr>
</tbody>
</table>

Table 1.4: ADAM-4000 control unit RS-485 addresses in the gas control system.

rigidities of 2.0 and 1.9 tesla meters respectively.

2. The production rate is comparatively high.

3. The reaction product has a easily detectable recoil-fission decay signature. $^{258}$Rf largely decays via spontaneous fission (87%) with 12 ms half life.

A 130-MeV beam in the laboratory frame of $^{22}$Ne$^{6+}$ particles was accelerated in the 88-inch Cyclotron for the production of $^{258}$Rf in the reaction $^{242}$Pu($^{22}$Ne, 6$n$)$^{258}$Rf ($\sigma$ = 1.7 nb[LL0+00]). This corresponds to a lab frame energy of 114 MeV at the center of the target. The beam passed through a 42.5 ± 2.5 $\mu$m carbon window and a negligible amount of gas before reaching the rotating target wheel. The target wheel had a radius of 4.25 cm and four banana shaped target segments of $^{242}$Pu (>99% pure) with 440, 340, 320, and 270 $\mu$g/cm$^2$ on a 2.4 $\mu$m titanium backing foil. The target wheel was rotated at a frequency of approximately 11 Hz in order to distribute heat deposited by the beam over all four target segments. Energy losses were calculated using SRIM [Zie04] as described in more detail in section 1.2.1.

The reaction products, beam, elastically scattered target atoms, and transfer reaction products then passed through the BGS which was filled with different fill gas mixtures during separate stages of the experiment. The gas mixture was controlled via two mass flow controllers. This ratio was confirmed with a RGA that was sampling the gas mixture during the entire experiment.

Rutherford scattered particles were detected in a p-i-n diode detector at an angle of 27° to the beam. This detector was used to measure the product of target thickness and beam intensity. The EVRs were detected in an older BGS focal plane detector array in use before the C$^3$ detector. It consisted of three 300-µm thick 60 × 60 mm 16-channel strip detectors. Vertical position was determined by resistive charge division while horizontal position was determined by the strip number. In addition, eight silicon cards were mounted perpendicular to the focal plane detector. These non position sensitive upstream detectors formed a box open only on the side at which the EVRs enter. Detection of decay chains in this experiment was accomplished searching for fission events within 120 ms (ten half-lives) of a recoil event in the same x-y location in the detector.
1.4.3 Reduction of Elastically Scattered Target Atoms in the Detector

The introduction of hydrogen into the fill gas mixture had the expected effects. Previously, with a 100% helium and 0% hydrogen fill gas mixture the rate of elastically scattered target atoms was so high that measurements of the decay of the EVRs was impossible. By adding hydrogen to the BGS gas mixture the peak due to elastically scattered target atoms was shifted out of the detectors at almost all gas ratios. In the measurement with a ratio of 87% helium and 13% hydrogen the tail end of the elastically scattered target peak could still be observed. At higher hydrogen admixtures the elastically scattered target peak was completely absent. Figure 1.15 shows the event rate in the focal plane detector in units of counts per minute per 10 keV for the three measured ratios.

![Elastic Scatter Peak](image)

Figure 1.15: Peaks in the focal plane detector due to the elastically scattered target atoms. For hydrogen concentrations > 13% the peak is completely removed.

In previous experiments the beam intensity had to be reduced and a 50% duty factor had to be employed in order to lower the background to a point where the acquisition system could record data. It is essential to bend the elastic scattering peak out of the detector, which was achieved with hydrogen/helium mixtures. This can further be verified by analyzing the events per strip on the detector. Figure 1.16 shows that some scattered target atoms are still reaching the west side of the detector using the 13%H$_2$/87%He gas mixture.

As figure 1.15 shows this peak is completely gone for the hydrogen concentrations
Figure 1.16: Scatter peak on the edge of the detector for the 13% hydrogen fill gas mixture. Elastically scattered material has been almost completely moved away from the detector and can now only be seen in the high-numbered strips at the low magnetic rigidity side of the detector.

greater than 13% tested in this experiment. The successful removal of elastically scattered target atoms makes it possible to run at higher intensities without having to chop up the beam. In addition, having removed this background, correlations can be identified with higher certainty since they are less likely to be due to random correlations.

1.4.4 Focusing of Reaction Products

By analyzing the position on the detector at which an EVR of a given energy was deposited, the horizontal spread due to velocity differences can be plotted. Since horizontal position is directly proportional to magnetic rigidity even small changes in the average charge can lead to horizontal defocusing. If the gas ratio is set in such a manner that the magnetic rigidity is constant over the relevant velocities, the EVRs will be more focused. For the reaction discussed above this would occur at a hydrogen mixture of 40% according to figure 1.11. Figure 1.17 shows that as the hydrogen content increases spread of the EVRs collapses into a smaller spread of
magnetic rigidities. The lines drawn on this graph are meant to guide the eye; they are not best fit lines.

Figure 1.17: Beam focusing effect due to optimizing the gas ratios. Increasing the hydrogen percentage tightened up the magnetic rigidity of the EVRs and thereby improving their focusing in the focal plane.

Gas Mixture Conclusions

While no mixture of gases is ideal for every possible nuclear reaction, it has been shown that having the ability to alter the gas mixture in a gas-filled separator greatly increases the capabilities of the separator by reducing the detector background for those reactions where magnetic rigidities of transfer products, elastically scattered atoms, or the beam are similar to that of the desired reaction products. Additionally, gas mixtures can be used to focus the wanted reaction products into a smaller area, thereby increasing the experimental efficiency.
1.4.5 Target Rotation Speed Control

In addition to being able to modify the gas mixtures, the LabVIEW program reads a voltage from a frequency counter measuring the frequency of a laser pulsing through the target wheel once per revolution. This frequency is then compared to the desired rotational frequency of the target wheel and a Proportional-Integral-Derivative (PID) code can be used to adjust the output voltage to either increase or decrease wheel speed. The rotation speed control box for the large target-wheel has been modified by inserting a 50% voltage divider to cut the control signal to 0 - 5 Volts. Therefore there is no need to make any changes within the LabVIEW program depending on which type of wheel is being used. Digitally controlling the rotation speed of the wheel is a vast improvement because a stop beam signal can now automatically be sent from the control unit to the control room in the event of a target motor failure. This will prevent damage to the targets caused by the high intensity beams if the wheel stops spinning for any reason.
Chapter 2

Target Production Methodologies

Targets are an essential component of experimental nuclear science and with recent advances in ion-source technology increasingly intense beams are becoming possible. Higher intensities are of great utility since the current trend in heavy element science is to explore ultra-low cross section nuclear reactions. With improving beam intensities it becomes increasingly important to better understand and improve current target production methods to optimize the performance and longevity of these targets. The heat deposited by the beam in nuclear experiments is often distributed by the use of a rotating target wheel. Such a target wheel can hold multiple banana-shaped target segments, such as the one shown in figure 2.1, in order to minimize localized heating by allowing the target material to cool down during the rotation period.

Figure 2.1: Banana-shaped target frames for mounting target segments on a wheel. These targets are generally produced independently and usually consist of a backing foil and a metal oxide layer.

The target materials used in these reactions are generally plated or deposited onto a backing foil which is attached to these frames via one of many different methods. Molecular plating is one of these techniques and is frequently used in heavy element
science to produce targets of rare isotopes due to its high efficiency. The electrodeposition cell described in this chapter and illustrated in figure 2.2 has been re-designed to produce these banana-shaped target segments with optimum efficiency. The cell used to produce targets for the BGS had an efficiency of 95\% for the production of americium targets. This is a significant increase over previously reported efficiencies on the order of 20-90\%[ESS+04, LEK+05], especially for production of isotopically pure targets of rare actinides that are very expensive. In comparison, techniques such as vapor deposition and sputtering frequently only have efficiencies of 2 to 5\%.

![Image 2.2: Schematic representation of the electrodeposition cell used to produce targets for the BGS. The target segment being deposited is shown in green. Isobutanol is used in this work but other solvents are also possible.](image)

The advantages of electrodeposition as a method for target production are its versatility and ease of reuse. While this protocol yields efficiencies of larger than 90\% for all of the metals that were tested with the new electrodeposition cell, it may still be necessary to recover the remaining target material. Since the deposition solution is almost exclusively isobutanol it can easily be evaporated to concentrate any remaining metal atoms.

While there are many publications describing the electrodeposition efforts of several different institutions, there is no clear protocol that describes the process adequately enough so that other researchers with no experience can easily reproduce a successful plating apparatus. The electrodeposition cell, shown in figure 2.3, and its protocol have been optimized based on the previous publications [ESS+04, CTG+01, TF89, DSY+10, LEK+05, Gre, EGB+10] as well as the personal communications that were necessary to re-establish an effective plating setup at Lawrence Berkeley National Laboratory.

The beginning of this chapter will discuss the electrodeposition cell in detail. Initial characterization experiments will also be described and a detailed procedure will be given. In the later part of the chapter our application of the recently developed polymer-assisted deposition (PAD) procedure as an alternative method to prepare
thin films of metal oxides [JMB+04] will be discussed. This work has been adapted to produce the metal oxide films necessary for nuclear reactions via heavy ion irradiations. The feasibility of the PAD project will be discussed in the conclusion.

2.1 Upgrades and Methods for the new Electrodeposition Cell

The electrodeposition technique can be very temperamental; therefore, different groups have different procedures and approaches to obtain high quality metal oxide targets. The following experiments were based on a basic procedure written by Ken Gregorich in 1995 [Gre]. The initial experiments with the new updated electrodeposition cell largely failed. They are included here to explain the choices made for the final procedure as well as to educate future generations of graduate students that may need to make new targets.

The new electrodeposition cell was designed based on previous iterations by Liv Stavsetra and Paul Ellison [Ell10] to overcome leaks in the seals of previous cells. This new cell uses a custom cut silicone gasket to form a seal with the target frame and reduce the deposition area to the area needed in BGS experiments. The cell is pictured below in 2.3:

Figure 2.3: Image showing the different parts of the electrodeposition cell. A) Anode plate with copper plunger. B) Solution chamber with Viton o-rings on both sides. C) Solution chamber with gasket. D) Stainless steel cathode. E) Entire cell without screws and ground connection F) Anode high voltage connection.
This new design has alleviated these leakage problems with previous iterations. Specifically, the plating area is now restricted to the desired active target area. A series of experiments was performed to optimize parameters for the americium target production for the “Superheavy Element Z identification via the $^{243}\text{Am}(^{48}\text{Ca}, 3-4n)^{287,288}\text{Am}$ reaction” experiment in August of 2012. The results of the electrodeposition cell characterization experiments and the final parameters and procedure are described below.

2.1.1 Characterization Experiments of the updated Electrodeposition Cell

Five experiments were performed to optimize the new electrodeposition cell with terbium, a homologue of americium, prior to producing americium-243 targets. Early experiments are included to help identify and illustrate problems that can crop up during electroplating. All characterization experiments used aluminum as a target backing foil and palladium as an anode. Experiment 1 was the initial test of the new electrodeposition cell. A stock solution of terbium nitrate hexahydrate (Aldrich, Lot #09724JS) was prepared, an aliquot was taken and spiked with approximately 7500 Bq of europium-152 chloride(Container #13311 on RWA 1017). The solution was dissolved in 300 microliters of concentrated nitric acid and brought to dryness twice in order to ensure that the final solid would be a nitrate and not a chloride salt. The dry metal nitrate crystals were then dissolved in isopropanol. Once in the cell, the solution was stirred for 1 minute every 5 minutes using an X-shaped stir bar. Figure 2.4 below shows the voltage ramp applied to the cell as well as the current against time.

![Figure 2.4](image)

Figure 2.4: Plot of time vs voltage and current for electrodeposition cell characterization experiment 1.
It is clear right away that the plating current is much too high. Aumann and Müllen [AM74] state that the current should be limited to 2 mA/cm$^2$. The significant voltage drop in the cell beyond 3000 seconds, is caused by an overall current limit of 16 mA. The problematic high current was addressed in Experiment 2. Activity was measured using a high purity germanium detector. The activity and efficiency results for this experiment are shown in table 2.1.

<table>
<thead>
<tr>
<th>Percent Activity</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Total activity plated on target</td>
<td>58.4</td>
</tr>
<tr>
<td>Activity left in solution</td>
<td>4.8</td>
</tr>
<tr>
<td>Activity left in vial</td>
<td>2.7</td>
</tr>
<tr>
<td>Activity lost in cell</td>
<td>34.1</td>
</tr>
</tbody>
</table>

Table 2.1: Efficiency of the plating cell during experiment 1.

The high deposition current in this experiment was caused by using old isopropanol of insufficient purity (95%). The large proportion of water created greatly increased the current, which in turn caused heat build-up in the plating cell. This increase in temperature of the plating solution may explain the lower than expected plating efficiency.

In experiment 2, the isopropanol solvent was replaced with HPLC grade isopropanol (Sigma Aldrich CHROMASOLV (>99.9% purity). Plating solutions were prepared similarly to experiment 1. An appropriate aliquot (containing 2.5 mg of Tb) was taken from the previously prepared Tb stock in concentrated HNO$_3$ and added to 400 $\mu$L of concentrated HNO$_3$. A small amount of europium radioactive tracer in hydrochloric acid was added to spike the solution. To ensure that no hydrochloric acid remained in the solution, it was evaporated to dryness three times under a heat lamp, each time adding a new 400 $\mu$L aliquot of concentrated HNO$_3$. After the third aliquot was evaporated, the remaining metal nitrate was dissolved in 20 mL of the new, purer isopropanol.

Additionally, other improvements were made to address issues identified during experiment 1. The X-shaped stir bar was sanded down so that it would properly rotate in the liquid chamber of the cell. The cell was not filled entirely with the electroplating solution/isopropanol, instead 1 mm was left at the top such that the open side of the cell could be covered with Parafilm. The Parafilm was deemed necessary because small particles of dust and debris were observed floating in the electroplating solution after experiment 1. This debris may be coming from the hood ventilation system. Therefore, this precaution would not be necessary in a glove box. The voltage and current response of this experiment are shown below in figure 2.5. The current in this experiment was much lower than in Experiment 1, however it was still much higher than reported literature values. A plating efficiency of 69% was achieved. The activity data for this experiment is shown below in table 2.2.
Figure 2.5: Plot of time vs voltage and current for electrodeposition cell characterization experiment 2.

<table>
<thead>
<tr>
<th>Percent Activity</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Total activity plated on target</td>
<td>68.6</td>
</tr>
<tr>
<td>Activity left in solution</td>
<td>10.1</td>
</tr>
<tr>
<td>Activity left in vial</td>
<td>1.3</td>
</tr>
<tr>
<td>Activity lost in cell</td>
<td>19.6</td>
</tr>
</tbody>
</table>

Table 2.2: Efficiency of the plating cell during experiment 2.

The resultant film on the target however, was very grainy which can be attributed to the still very high current. Experiment 3 attempted to address this problem by dissolving the metal ions as chlorides instead of nitrates. The same procedure was undertaken for the experiment 3 stock as it was done for experiment 2, using concentrated HCl instead of concentrated HNO$_3$ in 400 µL aliquots. The stirrer was run continuously on setting 4 during this experiment. However, as the voltage and current plots in figure 2.6 show, this did not solve the high current problem. Experiment 3 achieved a plating efficiency of 61% as shown in table 2.3. Replacing

<table>
<thead>
<tr>
<th>Percent Activity</th>
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</tr>
</thead>
<tbody>
<tr>
<td>Total activity plated on target</td>
<td>60.7</td>
</tr>
<tr>
<td>Activity left in solution</td>
<td>2.5</td>
</tr>
<tr>
<td>Activity left in vial</td>
<td>5.1</td>
</tr>
<tr>
<td>Activity lost in cell</td>
<td>31.8</td>
</tr>
</tbody>
</table>

Table 2.3: Efficiency of the plating cell during experiment 3.
the nitrate ion with chloride showed no improvements. Additionally the deposit on the target backing consisted largely of black crystalline point-like structures. It was deemed that hydrochloric acid introduced more problems and was not worth further investigation.

In experiment 4, the concentration of the nitric acid used to initially dissolve the metal ion was lowered to 0.01M in order to reduce the number of counter ions in solution. 100 µL of new Tb stock (prepared in 0.01 M HNO₃) was spiked with europium tracer. Again, this solution was evaporated to dryness three times. A smaller vial allowed for a much smaller volume of 0.01 M HNO₃, 50 µL, to be used when redissolving the dry solids. The final dried nitrate was again dissolved up in 8 mL of isopropanol. Figure 2.7 shows that this did slightly lower the current, however, not to values normally reported in the literature.

Lowering the nitrate concentration did not greatly reduce the current but it reversed the trend during the final 400 Volt deposition cycle; the current was dropping and not increasing anymore. It should be noted that this experiment was also stirred continuously and a plastic bag was used to cover the cell in lieu of Parafilm, as the Parafilm has the tendency to stick and draw up the electroplating liquid, resulting in drips outside the cell. This experiment obtained an efficiency of 74.4%. Results are shown in table 2.4. However, the film surface was still very non-uniform and grainy.

At this point, in experiment 5, a new solvent was explored, isobutanol. Isobutanol is less polar than isopropanol; therefore, it absorbs less moisture from the air. The dried stock used for this experiment was one that had been previously prepared for

Figure 2.6: Plot of time vs voltage and current for electrodeposition cell characterization experiment 3.
Table 2.4: Efficiency of the plating cell during experiment 4.

<table>
<thead>
<tr>
<th>Percent Activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total activity plated on target</td>
</tr>
<tr>
<td>Activity left in solution</td>
</tr>
<tr>
<td>Activity left in vial</td>
</tr>
<tr>
<td>Activity lost in cell</td>
</tr>
</tbody>
</table>

Table 2.5: Efficiency of the plating cell during experiment 5.

<table>
<thead>
<tr>
<th>Percent Activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total activity plated on target</td>
</tr>
<tr>
<td>Activity left in solution</td>
</tr>
<tr>
<td>Activity left in vial</td>
</tr>
<tr>
<td>Activity lost in cell</td>
</tr>
</tbody>
</table>

Figure 2.7: Plot of time vs voltage and current for electrodeposition cell characterization experiment 4.

Experiment 4. It was initially dissolved in isopropanol, but was then brought back to dryness and re-dissolved in 8 mL of isobutanol. Unfortunately, this resulted in higher-than-normal losses of material to the container, as shown in table 2.5. Additionally, the plating voltage was reduced from 400 volts, as described by Ellison and Gregorich independently in their procedures for molecular plating, to 150 volts. The plating time was also increased to 5 hours and the stirrer was run continuously, just as in experiment 4, this time on setting 3. Figure 2.8 shows the resultant plot of current and voltage vs time. Using isobutanol as a solvent lowered the current by a factor of five. Visible inspection showed that the quality of the film was much higher. Additionally, the plating efficiency was increased to 83 percent as shown in table 2.5. An additional
problem was discovered in this experiment; if the cathode surface is not smooth, if it was scuffed due to cleaning for example, plating in that area will be less efficient. Therefore, an area of low activity may be created. This can be visibly seen in target 5 shown in figure 2.9.
Resultant Target | Comments
--- | ---
| | Experiment 1: A thick layer of nitrate and impurities can be seen
| | Experiment 2: Less impurities but very non-uniform. Still too much nitrate deposited. Powdery and flaky, material can be observed in the plastic bag containing the target (low adhesion)
| | Experiment 3: The Chloride deposition was even more non-uniform. Black specks were observed
| | Experiment 4: Less nitrates but still non-uniform
| | Experiment 5: Very uniform deposition except for damaged area on the electrode

Figure 2.9: Images of the five target segments produced during the characterization experiments of the new electrodeposition cell.
2.2 Electroplating Procedure

The procedure developed through these experiments was then implemented to successfully produce americium targets and is described in great detail in this section.

2.2.1 Solution Preparation

A chemical separation may need to be performed prior to creating a deposition solution. However, in most cases where the isotope of interest can be purchased as a solid powder, it simply has to be dissolved. This can often be achieved with 1M nitric acid but may require more aggressive approaches such as aqua regia. No matter how the metal ion is first brought into solution, it should be evaporated to dryness after the initial dissolution. In this process the metal ion will precipitate as a metal nitrate. The metal nitrate should then be re-dissolved in minimal amounts of 1M nitric acid and evaporated to dryness two to three times. This ensures that contaminants such as chlorides and excess nitrates are removed from the solution. Finally the dry metal nitrate should then be dissolved in ultra-high purity isobutanol. No aqueous acid should be necessary after the purification steps. If dealing with dangerous or radioactive isotopes, the entire procedure should be carried out in a fume hood or glove box.

2.2.2 Frame Preparation

First the backing foils must be cleaned because they may have been covered with a thin layer of oil to prevent oxidation or chemical attack. The foil should first be cleaned with acetone, and then rinsed with water, followed by a rinse with isobutanol. In between each step the foil should be allowed to dry thoroughly. The blank stainless steel frames should be checked for rough corners or edges. If any are found they should be filed off with 400 grit sandpaper. The frame should then be glued onto the previously cleaned foil using carbon glue. Use of other glues is possible as long as they are chemically resistant to isobutanol and able to withstand the high temperature required to transform the metal into an oxide. The frame and foil should now be set aside for twelve to twenty-four hours in order to let the glue set. A scalpel should then be used to carefully trim the foil around the edges of the frame.
2.2.3 Deposition and metal oxide creation

When a new cell is commissioned, a leak test should first be performed before any radioactive or hazardous materials are added to the cell. The easiest way to do this is to fill the cell with distilled water and set it on a sheet of paper for fifteen minutes. Leaks will appear on the paper as water marks. The plating cell must be cleaned to prevent cross contamination of target materials, and to insure that no extraneous materials are present. The cell should be cleaned twice with 1M nitric acid, then rinsed twice with distilled water, and finally rinsed twice with isobutanol. The plating is performed as a single layer deposition of $\approx 500 \, \mu g/cm^2$ of the cation of the isotope of interest.

After inserting the prepared metal foil and frame and assembling the cell, the prepared deposition solution and the stir bar are inserted into the cell. A voltage of 150 V is applied for five hours. If a radioactive metal ion is being deposited the progress may be monitored by taking small samples of the solution and measuring the activity. After five hours the now depleted plating solution is removed and the frame is removed from the cell. After drying the metal nitrite layer on the target backing foil with a UV-lamp the frame is fired to create a more durable oxide in a muffle furnace for one to two hours. The temperature of the furnace is determined by the metal oxide formation temperature of the plated metal. The maximum temperature is limited by the heat tolerance of the backing material. A maximum temperature of 300°C is suggested.
2.2.4 Detailed Procedure

1. Solution Preparation \(\approx 3\) hours: Measure the desired weight of metal cations on a precision scale. The active target area of this setup is 4.5 \(\text{cm}^2\). To make targets with a thickness of 500 \(\mu\text{g/cm}^2\), a mass of 2.5 mg is necessary, assuming a 90\% efficiency. It may be prudent to create a stock solution to avoid having to repeat this step for each target frame.

2. Dissolve the metal ions using five to twenty \(\mu\text{L}\) of 1M nitric acid. If the metal ions do not dissolve higher concentrations of acid or aqua regia may be necessary.

3. After the metal ions are dissolved, raise the temperature of the vial to 80 \(^\circ\text{C}\) while flushing argon gas into the vial. This will remove water as well as excess counter ions.

4. Repeat step 2 and 3 as necessary.
   CRITICAL: These purification steps are essential as they lower the amount of impurities and thus create a much smoother deposition layer.

5. Dissolve the pure metal nitrate now present in the vial in high purity isobutanol. CRITICAL: It is very important to use isobutanol of the highest possible purity. Contaminations and dissolved water will raise the deposition current and lower the efficiency and surface uniformity.
   PAUSE POINT: At this point the solution may be capped and stored until the next day.

6. Electrodeposition \(\approx 5.5\) hours: Insert the Viton O-rings into groves of the cleaned solution chamber as shown in figure 2.3-B.

7. Insert the rubber gasket into the recess for the frame.

8. Place the cleaned Ti-backed-target frame onto the O-ring at the side of the cell, which should be upside down or tilted. The blank target should be inserted such that the stainless steel frame touches the gasket and the titanium foil against the stainless steel cathode.

9. Place the solution chamber on the base plate and Ti-foil.

10. Tilt the cell on its side such that the solution chamber is sitting on top of the stainless steel cathode. Place the palladium electrode (anode) on top such that it does not overlap with the Viton o-ring.

11. Fasten the cell to the base with the four long socket head screws. Screw tension should be just sufficient to fully compress the O-rings.

12. Attach the ground wire to the cathode.
13. Add a few mL of isobutanol before adding a measured aliquot of the stock solution to the cell. The aliquot should be enough to produce a layer of 500 $\mu$g/cm$^2$ of the metal based on the expected efficiency.

14. Add isobutanol to fill the cell (taking into account the use of the stirrer and its volume) The volume of the cell described in Appendix is 26 mL.

15. Be sure the High Voltage Power Supply (HVPS) is switched off and is set to deliver positive high voltage once turned on! Connect the high voltage cable to the connector on the plating cell.

16. Lower the stirrer into the plating cell. The stirrer should never touch the electrode. The edges of the stir bar may have to be shaved off to be able to freely rotate in the plating cell.

17. Start the stirring motor. A low rotation (10-60 Hz) should be sufficient. CAUTION: The stirrer should not touch anything.

18. Turn up the HVPS to 150 V. Monitor the current during the plating. Aumann and Müllen [AM74] suggest keeping the current density below 2 mA/cm$^2$. I suggest a much lower value: 0.5 mA/cm$^2$.

19. After 5 hours the plating should be complete. Turn off the HVPS. Remove the HV cable from the plating cell.

20. Pour the isobutanol solution from the plating cell into a designated and clean beaker. The target material in the used plating solution may have to be recovered. The isobutanol in the beaker tends to evaporate on its own. When it is dry, cover the beaker to prevent contamination with dust, etc.

21. Remove the four screws, which hold the cell to the base plate.

22. Remove the target and dry it under heat lamp. PAUSE POINT: The target may be stored indefinitely before baking. However, the nitrate layer is very fragile and should be oxidized as quickly as possible to avoid damage due to improper handling.

23. Metal Oxidation and Target Characterization $\approx$5 hours: Place the target segment into the furnace and raise the temperature slowly (1-2.5 °C per minute) until the oxidation temperature of the metal being plated is reached. Hold the temperature for 15 minutes.

24. Remove holder from furnace and set on a heat resistant material to cool

25. When cool, remove the target from the tube furnace.

26. Measure thickness of the target using the appropriate method.

27. Repeat for other target segments.
2.2.5 Expected Results

This procedure makes it possible to produce uniform targets of varying thicknesses, depending on the amount of material used in the plating solution. The procedure can easily be adapted for other target materials.

2.2.6 Troubleshooting

1. A large deposition current will result in non-uniform target segments. This is most often caused by impurities in the deposition solution. It is essential to use high purity reagents and follow the purification procedure described in steps 2-5. Additional sources of impurities may be dust falling into the deposition cell or water dissolving into the isobutanol because it was left uncapped for too long.

2. If the titanium backing foil is no longer attached to the stainless steel frame, a glue was used that is not chemically compatible with isobutanol. It is important to use glue that does not dissolve in isobutanol and is heat resistant.

2.2.7 Electrodeposition Conclusion

Two wheels consisting of four banana-shaped americium oxide ($^{243}\text{Am}_2\text{O}_3$) targets were produced with the upgraded electrodeposition cell. The target material was

Figure 2.11: Americium targets produced with the new electrodeposition cell and improved procedure.
deposited onto 2.7 um thick titanium after chemical purification. The target wheels had average americium thicknesses of 650 and 430 µg/cm², respectively. The metal oxide layer on these targets was of very high quality. As an alternative to metal oxide layer creation via electrodeposition, target production via polymer-assisted deposition has also been investigated.
2.3 Polymer-Assisted Deposition (PAD) Method

Across most low-energy nuclear facilities, there has recently been a push for higher intensity stable and radioactive beams at Coulomb barrier energies. To further study nuclear structure, reactions, and astrophysics, higher energy and intensity beams are necessary, as outlined in the Nuclear Science Advisory Committee (NSAC) Long Range Plan 2007 [Sav08]. In the time leading up to the opening of the FRIB at Michigan State University, ATLAS at Argonne National Laboratory is being refit to provide a ten-fold ion beam intensity increase for all species [Sav09]. The high beam intensities produced in these next generation particle accelerators necessitate target materials especially resilient to radiation damage. The non-flaking, smooth, and thermally resistant targets produced by PAD show potential for use with higher intensity ion beams.

In earlier work, the heavy elements group has shown that the PAD method yields highly uniform metal oxide films with characteristics that fulfill the requirements for targets in nuclear reactions. Research is now being performed on expanding this methodology for the production of actinide targets, which often exhibit complex chemical behavior. The work is being conducted at the 88-Inch Cyclotron at Lawrence Berkeley National Laboratory (LBNL), where Garcia et al. have produced and irradiated several thulium(III) oxide targets prepared by the PAD method as a model system for americium. The stationary targets prepared by the PAD method proved resilient up to a beam current of 200 particle nanoamperes (1 electrical µAmp) of argon beam [GAC+08].

![Figure 2.12: Left: Radioactive solution spin coating apparatus with inner canister closed. Right: Apparatus with canister open; four Si$_3$N$_4$ target segments are visible.](image)

To utilize PAD with a target wheel, a spin coating apparatus was designed to create high quality radioactive thin films. In keeping with radiation safety principles, special consideration was given to minimizing contamination due to either equipment failure or user error. These design considerations include a small foot print for use
in glove boxes, primary and secondary containment, safety interlocks, and capacity for interchanging spin canisters. The spin coating apparatus designed around these features is shown in figure 2.12, which showcases many of the design goals. First, the primary (tan PEEK disk closed on left, open on right) and secondary (large black lid on left) containment are visible. The tan inner canister is removable to allow for the production and swapping between multiple canisters for different radioisotopes. The apparatus has been wired and programmed to allow for its control through simple command input into the front digital panel. It has been programmed to interlock the opening of the secondary containment while spinning, thereby preventing the possibility of contamination by the radioactive PAD solution in case of device failure.

To determine whether the PAD method would be suitable for the creation of p-block metal targets, lead and indium were tested first. The metal oxide targets were prepared as follows: commercially available single-side polished silicon wafers were purchased with a one \( \mu \mathrm{m} \) Si\(_3\)N\(_4\) coating on both sides. The desired target frame shape (same shape as shown in figure 2.1) was then scribed onto one side of the wafer. These patterned wafers were then placed into a 20\% potassium hydroxide bath at 80 \( ^{\circ} \mathrm{C} \) for 5-6 hours to etch away the now exposed silicon. This results in banana-shaped target frames with a 1 \( \mu \mathrm{m} \) Si\(_3\)N\(_4\) window left over from the unscribed side of the wafer. Figure 2.13 shows frames produced in such a manner.

Solutions of 6\% by weight (b.w.) lead and 6\% (b.w.) indium with 15\% (b.w.) polyethylenimine (PEI) in water were prepared in the same fashion as the thulium solutions described by Ali et al. [AGPMN10]. In this case, lead nitrate (\( \text{Pb(NO}_3\text{)}_2 \)) and indium chloride (\( \text{InCl}_3 \)) were used as the precursors to the metal-polymer solutions. A one milliliter aliquot of the lead solution was then dripped onto the polished side of the silicon nitride and spread evenly over the surface. This was repeated on three other segments. The four target segments were then mounted in the spin canister (figure 2.13) and spun at 1500 RPM for one minute. They were then removed, and baked for three hours in a muffle furnace to a final temperature of 600 \( ^{\circ} \mathrm{C} \). This process created a thin layer of lead dioxide (\( \text{PbO}_2 \)) and on other targets, indium oxide (\( \text{In}_2\text{O}_3 \)), while the polymer decomposes to volatile oxides.

The same process was carried out to make the indium oxide targets, except a square target was scribed out instead, to be used as a stationary irradiation target. A second layer of lead or indium oxide can be coated on the wafer by adding an additional one mL aliquot of the 6\% by weight solution and repeating the steps described above.

After the first application of the PAD method the thickness of the single layer PbO\(_2\) film was \( \approx \)150 nm (140 \( \mu \mathrm{g/cm}^2 \)) as found by measuring the energy loss of alpha particles passing through the film. The determined energy loss was converted to a thickness using energy loss coefficients calculated from SRIM. Through reapplication of the PAD method, subsequent layers were found to add approximately 140 nm (130 \( \mu \mathrm{g/cm}^2 \)) per iteration. Typically, lead targets used at the 88-Inch cyclotron are 500
Figure 2.13: Target wheel consisting of four lead oxide targets on a thin Si$_3$N$_4$ backing in a storage cassette.

$\mu g/cm^2$ or greater, requiring at least four layers of lead oxide to be deposited.

The In$_2$O$_3$ film was $\approx 100$ nm ($96 \mu g/cm^2$) as measured by taking the difference in weight of the target before and after adding the metal oxide film. A target wheel suitable for irradiations at the BGS was then assembled from the PbO$_2$ segments produced with this procedure. Figure 2.13 below shows the completed target wheel. The lead oxide film had a mirror finish, indicating a surface roughness on the nanometer scale.

With the successful creation of oxide targets, the PAD method has been tested with p-block metals, transition metals, and lanthanides. Actinide targets so far remain more problematic.

### 2.3.1 Irradiation Test of a PAD produced rotating Target Wheel

Before irradiating PAD-produced radioactive targets that could contaminate the BGS, it is necessary to understand the limits of the PAD-produced metal oxide and the Si$_3$N$_4$ backing material with non-active targets. To determine these limits, the PAD PbO$_2$ targets were irradiated at the 88-inch cyclotron. The $^{208}$Pb($^{40}$Ar, 4$n$)$^{244}$Fm nuclear reaction was chosen for these tests because of the large energy loss of the $^{40}$Ar beam in the targets and the high intensities at which the $^{40}$Ar beam is available at the 88-inch cyclotron. Silicon PIN diode detectors mounted at 27 degrees
from the beam axis were used to detect argon-40 beam ions elastically scattered in the target and used to monitor the product of beam intensity and target thickness. The $^{208}\text{Pb}(^{40}\text{Ar},4n)^{244}\text{Fm}$ nuclear reaction has a cross section of about 1.7 nanobarns [KHHb$^+$08], which translates to a rate of detection of six $^{244}\text{Fm}$ atoms per hour in the BGS focal plane detector. To produce this nucleus, the cyclotron was tuned to provide a 191 MeV beam of $^{40}\text{Ar}$. It was planned to test the durability of the PAD targets by slowly increasing the argon-40 beam intensity to the maximum possible.

Figure 2.14: Irradiation damage to targets produced via polymer assisted deposition. A wave-like pattern indicative of target degradation can be observed.

During the irradiation of the targets, the beam intensity was slowly ramped up over the first two days of the experiment. However, due to a power failure which affected the entire LBNL facility the experiment was abruptly shut down and could not be continued due to damage to the cyclotron. The targets were then inspected and it was found that one was broken and three exhibited a wave-like pattern that indicates melting of the silicon nitride target backing. The CHOPPER spectrum, a spectrum of Rutherford scattered particles vs. rotation phase of the target wheel, enables the analysis of each individual target during irradiation. It showed that the targets were highly uniform and that the target that was broken did not break during irradiation but likely due to the vacuum failure caused by the power outage.

While it was not possible to measure the maximum beam intensity that the targets were capable of withstanding in this experiment, it still lead to the conclusion that the target backing material likely needs to be changed. It already showed signs of degradation after irradiation with an intensity of one particle micro ampere as shown.
in figure 2.14. It may be desirable to test the PAD process on more heat conductive backings such as thin metal foils. Specifically, the silicon nitride backings may be more susceptible to damage caused by the beam because it has a crystalline structure and may not be able to self anneal. Possible alternatives to Si$_3$N$_4$ backings such as titanium and beryllium will be explored.

### 2.3.2 Investigation of the Production of Actinide targets with the PAD Spin Coater

As a first step in producing actinide targets, the long-lived radioactive element uranium was used. While PAD has previously been successful in preparing various metal oxide targets [GAC+10], the complicated chemistry of uranium has created several problems with the production of uranium PAD targets. In the first step of the spin coating procedure, a solution of PEI is prepared and then mixed with a solution of dissolved uranyl salt to produce the solution that will be loaded into the spin coater. However, the pH of this PEI solution is very basic, near pH = 11. When mixing such a basic solution with dissolved uranyl nitrate (UO$_2$(NO$_3$)$_2$), the uranium precipitates, resulting in a suspension-like solution that is impossible to spin coat. To solve this problem, the pH of the PEI solution was first lowered by adding hydrochloric acid and then mixed with the uranium salt solution. Unfortunately, the acidic nature of these solutions greatly destabilized the PEI solutions, resulting in its rapid degradation and decreased spin coating effectiveness. PAD targets produced with the acidic PEI uranium spin coating solutions produced significantly thinner UO$_2$ layers than expected (only 10 - 30 µg/cm$^2$) likely due to the decreased viscosity of the solution matrix.

Work is currently being performed to optimize the chemistry of the spin coating solution to balance between the high pH necessary for PEI and the low pH necessary to prevent the precipitation of uranium. One possible solution under investigation is the addition of anionic species to the uranium salt solutions to prevent the precipitation of the uranium when mixed with the high pH PEI solutions.

Due to the intrinsic insolubility of the uranyl ion at pHs above five, complexes of uranyl were explored in hopes to find a procedure that stabilized the ion at higher pH values. Initial work focused on carbonate ligands. However, the maximum solubility of uranyl achieved was 2% by weight. As reported by Garcia et al. [GAC+10], this concentration is not feasible for target production because a concentration of 6% is needed. Other ligand alternatives are being explored in order to adapt this method for the production method for actinide targets.
2.3.3 Proposed Production of Actinide Targets

More work on actinide target production is necessary. Finding a ligand that can be used to complex actinides such as uranium is not simple. The ligand must easily complex the actinide and leave the combined molecule in a negatively charged state such that the polymer can still complex the system. Additionally the ligand must undergo a clean combustion reaction such that it does not leave any residue during the baking of the targets. An ideal ligand candidate is ethylenediaminetetraacetic acid (EDTA), which complexes most actinides and burns cleanly. However, EDTA does not form stable complexes with uranium(VI) [MPTI93, Dav61]. Finding an appropriate ligand for uranium is complicated by the fact that uranium forms uranyl ions in solution. These ions are photochemically active in the presence of organic ligands [OL82, AS60].

Future research in this project will explore the behavior of carboxylate ligands such as citric acid to determine their ability to complex uranium and other actinides without interfering with the polymer chemistry in the spin coating solution. Once an appropriate ligand has been characterized the PAD method will be applied to create targets made out of actinides such as thorium, americium, and neptunium. Creating these targets is highly important for new generation nuclear experiments.

Even though metal oxide targets of unparalleled quality can be created that could increase the beam intensity that these targets can withstand, silicon nitride was not a suitable backing material. In addition to optimizing the metal oxide layer on the target, new target backings to further increase stability under high-energy beam intensities should be explored.

2.3.4 Proposed Investigation of ultra thin Titanium as an Alternative to Silicon Nitride

Generally, targets in low energy nuclear science use titanium, carbon, or beryllium backings [Rab54]. Titanium backings are created by cold rolling titanium foils [Max67] and then gluing them to metal frames. This technique limits the thickness of titanium foils to a minimum of approximately two $\mu$m due to pinhole formation. Carbon foils are traditionally floated on water and then picked up with a frame [LBE+08]. Neither of these procedures are suitable for producing targets via the PAD method because they do not result in ultra smooth backings. It should be possible to create ultra thin, ultra smooth titanium backings using etching procedures similar to the ones used by our group for making silicon nitride backings. The production of such one $\mu$m thick titanium backings will be explored.

Higher intensities are of great utility since the current trend in heavy element
science is to explore ultra low cross section nuclear reactions [Mah00]. The advances in source technology [OUL+09], as well as the drive to perform measurements at the sub pico-barn cross section level, make it increasingly important to create stronger and more resilient targets.

Reducing the thickness of the titanium backing will reduce the amount of heat deposited by the beam. The energy deposited in a two $\mu$m film is 12 MeV (calculated with SRIM 2004 [LLL+10]), which corresponds an energy deposition of 12 Watts for a one particle $\mu$A. Being able to halve the thickness of titanium would make it possible to double the beam intensity used for these targets. There are several different methods that can be explored for the production of these backings. Ideally they would be created via commercial plasma etching [Cob79]. However, due to cost and the specialized equipment necessary for this method, alternatives such as solution etching similar to silicon nitride backing production procedure described above could be explored. A potential candidate for this procedure is a copper wafer coated with titanium. Copper etchant type CE-200 by Transene does not interact with titanium and may thus be used to etch copper selectively [Cob79], creating an ultra smooth and ultra thin titanium surface onto which PAD solutions can be spun.

In conclusion, continuing research in producing targets via polymer assisted deposition and using micro machining procedures to produce high quality target backings should make it possible to produce targets of unparalleled quality, greatly increasing the effectiveness of the low energy nuclear science program at the 88-inch cyclotron and at other irradiation facilities.
Chapter 3

Nuclear Structure Measurements of Fermium-254

This work is focused on new capabilities that have been developed at the BGS, making it possible to remove evaporation residues from the high background areas of their production and to study their nuclear structure. High-intensity stable ion beams from the 88-inch cyclotron, together with the highly selective and efficient separation of compound nucleus evaporation residue products using the BGS, and the rapid delivery to a low-background detector facility via the Berkeley Recoil Transfer Chamber (RTC), open up many new possibilities for nuclear decay and structure studies in the neutron deficient actinides and transactinides. These actinides predominantly decay via electron capture which produces vacancies in the K-shell. Electrons filling these vacancies result in x-rays uniquely identifying the Z of the decay products. These decays also populate excited states in their daughters which subsequently de-excite by γ decay. These γ-rays are observed in coincidence with the characteristic x-rays in a two-clover γ-ray detector setup similar to the one described by Dababneh et al. [DPA+04]. This new method was used to study the electron capture of the nucleus $^{254}\text{Md}$ populating excited states in $^{254}\text{Fm}$, replicating the previously known states as well as observing a new level at 985.7 keV.

In addition to single particle states, excited states are expected at approximately 1 MeV in spheroidal nuclei due to quadrupole and octupole vibrations around the equilibrium shape. Neergard et al. [NV70] predict these states for several fermium isotopes. These states are generally described by the quantum numbers $K$, the projection of the spin $J$ onto the nuclear symmetry axis, and $I$, the total angular momentum. The quadrupole vibrations in this region are characterized by $K=2$, $I=2,3,4...$ and even parity, whereas the octupole vibrations are characterized by $K=2$, $I=2,3,4...$ and negative parity (calculations performed by Jolos et al. [JMSS11]).

Even-even nuclei are commonly known to have ground state rotational bands with
K=0, I=0,2,4... and even parity. In the fermium region the 2+ rotational state occurs around 45 keV above the 0+ ground state and the 4+ state occurs around 145 keV. These transitions are highly converted with $\alpha$-values (ratio of internal conversion decays to $\gamma$-decays) of approximately 1300 and 30 respectively as calculated by the theoretical conversion coefficient calculation model BRICC [KBT+08]. This makes them very difficult to observe directly in $\gamma$-coincidence spectroscopy. These transition energies can be inferred from energy differences in the $\gamma$-rays feeding the ground state band or can be measured in conversion electron coincidence spectroscopy.

This work is focused on studying the higher excited particle and vibrational states in fermium-254. Results in neighboring isotopes of fermium are included for comparison purposes. In this region of the chart of nuclides, particle, vibrational and rotational excitations are expected to be observed. This chapter will begin with a theoretical section discussing the underlying concepts providing a foundation for understanding the experimental results. The experimental setup as well as experiments characterizing the apparatus will then be discussed before treating the experimental results.

3.1 Structure Models

Nuclear structure models fall in one of two camps: the microscopic and the macroscopic approach. In the microscopic approach the nucleus is treated as a collection of particles creating an average potential. This potential is felt by a few valence nucleons and can be used to approximate their properties. In contrast, the macroscopic approach describes the nucleus as an incompressible liquid drop and is characterized by charge, volume and surface interactions. Much of the theory covered here is covered in more detail by Krane [Kra88].

3.1.1 Mean-field Approximation

To precisely describe a nucleus, an exact solution to the Schrödinger equation:

$$H\Psi(1, .., A) = E\Psi(1, .., A)$$

where $H$ is the Hamiltonian operator, $\Psi$ is the wave function, and $E$ is the energy of the system, is required. However, except in very light nuclei, this many body problem becomes impossible to solve. Larger nuclei are described via a Hamiltonian for the mean field interaction, $H_{mf}$, as well as a Hamiltonian for the residual interaction,
Figure 3.1: Calculated nuclear potentials for a nucleus with a radius of 5 fm. Radius is calculated from \( r = r_0 A^{1/3} \) with \( r_0 \) being 1.25 fm. Shown are the square well potential in green, the harmonic oscillator potential \((x^2)\) in blue and the Woods-Saxon potential \((-\frac{V_0}{1+\exp\frac{r-R}{a}})\) in red.

\[
H_{res}.
\]

\[
H = \sum_{i=1}^{A} \left( \frac{p_i^2}{2m_i} + U_i \right) + \sum_{i=1}^{A} \sum_{k>i}^{A} (V_{ik} - U_i) = H_{mf} + H_{res} \tag{3.2}
\]

where \( p \) is the momentum of the nucleon, \( m \) is its mass, \( U \) is the average potential created by the nucleons, \( V \) is the 2-body interaction between nucleons, and \( i \) and \( k \) count all \( A \) nucleons. Analytically, a harmonic oscillator provides an easy solution. However, it does not accurately describe the nucleus. Many different potentials are used in nuclear calculations. Figure 3.1 shows a few of these standard potentials.

Most commonly, a rounded square well potential (such as the Woods-Saxon potential in figure 3.1), with spin orbit coupling taken into account is used in these calculations. Such a potential easily reproduces the proton and neutron shells as well as their magic numbers observed in experiment. Figure 3.2 shows the resulting level structure highlighting the shell closures and magic numbers.

The Woods-Saxon or rounded well potentials can be improved with even more realistic self-consistent potentials obtained via variational methods. However, such Hartree-Fock type methods will not be discussed here.
Figure 3.2: Calculated nuclear structure levels based on a rounded square well potential. Spin orbit coupling is taken into account on the right side of the figure. The orbital angular momentum quantum number $l=0,1,2,3...$ is labeled with the s,p,d,f notation style. Magic numbers are indicated on the right of the chart. Taken from W. Loveland et al., Modern Nuclear Chemistry, Wiley, New York, 2005 [LMS05].
3.1.2 Spherical Shell Model

In contrast to mean field approximations, the independent particle model is based on a spherically symmetric potential and each nucleon is considered to be an independent particle. Consequently closed spherical shells have no impact and only valence nucleons affect the properties of the nucleus. The independent particle model generally describes nuclei very well in cases where there are only few nucleons outside of filled shells. Limiting the calculations to just the valence nucleons is accomplished by assuming that the residual interaction beyond the mean field interaction is dominated by the pairing interaction. It is convenient to treat pairing via quasi-particles. Quasi-particles are linear combinations of particle and hole wave functions. In this case, the single particle energy, $\epsilon_i$, becomes a quasi-particle energy, $\epsilon'_i$, based on the following relation:

$$
\epsilon'_i = \sqrt{(\epsilon_i - \lambda)^2 + \Delta^2}
$$

(3.3)

where $\lambda$ is the Fermi energy and $\Delta$ is the pairing-gap parameter. The spherical shell model is particularly useful for nuclei of low $A$. It, however, breaks down at high atomic numbers due to the large number of valence nucleons. With increasing valence nucleons, the attractive proton-neutron interactions become so strong that they cause the nucleus to deform. This deformation is proportional to the distance from the closed shells. To better describe such deformed nuclei, the Nilsson model has been developed.

3.1.3 Nilsson Model

When nuclei become deformed, a central mean field is no longer applicable. In such a case a field deformed in one of the axial directions ($x = y \neq z$) can be used. In this case the single particle Hamiltonian in equation 3.2, when including $l \cdot s$ and $l^2$ coupling, becomes:

$$
H = \frac{p^2}{2m} + \frac{m(\omega_x^2(x^2 + y^2) + \omega_z^2z^2)}{2} + 2\hbar\omega_0 \kappa l \cdot s + \mu \kappa \hbar \omega_0 l^2
$$

(3.4)

Here, $l$ is the orbital angular momentum and $s$ is the particle spin. The second term changes the spherically symmetric potential to an anisotropic potential where $\omega_x$ and $\omega_z$ define the deformation of the nucleus based on deformation parameter $\epsilon_2$ developed by Nilsson. The last two terms parametrize the $l \cdot s$ and $l^2$ interactions. In the Nilsson model several new quantum numbers are introduced for deformed nuclei. Figure 3.3 illustrates the geometric interpretation of these quantum numbers. For axially symmetric nuclei the quantum number $K$ is used in place of $\Omega$. This most often occurs in studies of $K$-isomers. Figure 3.4 and figure 3.5 show the solutions of equation 3.4 as a function of the deformation parameter $\epsilon_2$. Nilsson states are
Figure 3.3: Nilsson state quantum numbers. $\Lambda$ is the angular momentum projection onto the symmetry axis, $\Sigma$ is the spin projection on the symmetry axis, and $\Omega$ is the total angular momentum projection onto the symmetry axis. The labels $l$, $j$, and $s$, correspond to the traditional definitions of orbital, spin, and total angular momentum.

labeled as:

$$\Omega^\pi[Nn_z\Lambda]$$ (3.5)

(where $\Omega$ gives the total projection of nucleons angular momentum ($l \cdot s = j$) onto the $z$-axis, $\pi$ is the parity of the state, $N$ is the principal quantum number of the major shell, $n_z$ is the number of nodes in the wave function along the $z$-axis, and $\Lambda$ is the orbital angular momentum projection onto the $z$-axis. The Nilsson model is particularly applicable to neutron deficient actinides since they are generally highly deformed nuclei. In the case of fermium, its atomic number $Z$ falls in between the two shell closures 82 and 114. The power of the Nilsson model lies in predicting the ground state as well as possible single particle excited states of a nucleus of known deformation. By comparison, the liquid drop model describes the macroscopic properties of the nucleus.

3.1.4 Liquid Drop Model

In the liquid drop model developed by Weizäcker [Wei35], the nucleus is treated as an incompressible charged liquid. In this model the mass of the nucleus is given by the sum of the individual components, neutrons and hydrogen atoms, minus the binding energy of the system.

$$M(Z,A)c^2 = [ZM(^1H) + (A-Z)M(n)]c^2 - B_{tot}(Z,A)$$ (3.6)
Figure 3.4: Plot showing the energy of individual Nilsson states for protons $Z \geq 82$ as a function of the nuclear deformation parameter $\epsilon_2$ taken from [Fir96].
Figure 3.5: Plot showing the energy of individual Nilsson states for neutrons $N \geq 126$ as a function of the nuclear deformation parameter $\epsilon_2$ taken from [Fir96].
Here \( B_{\text{tot}} \) is given by what is now called the semi-empirical mass equation which fits nuclear binding energies with pairing effects:

\[
B_{\text{tot}}(A, Z) = a_v A - a_s A^{\frac{5}{3}} - a_c \frac{Z^2}{A^{\frac{1}{3}}} - a_a \frac{(A - 2Z)^2}{A} \pm \delta
\]  

(3.7)

where \( a_v, a_s, a_c, \) and \( a_a \) are fit parameters. The first term of the semi-empirical mass equation takes into account the volume of the nucleus. This is a constant term since the nuclear force saturates. It is, therefore, expected that all of the central nucleons contribute equally to the overall binding of the system. Only the nucleons on the surface are not completely surrounded and, therefore, have a different contribution. The second term accounts for decreased binding at the surface by correcting for the overestimation of the binding in the first term. In addition to the volume and surface term, the reduction in binding due to the Coulomb repulsion between protons is taken into account with the third term. The fourth and fifth terms represent the quantum mechanical effects of asymmetry energy and pairing. While this model gives good estimates for properties of the nucleus such as mass and binding energy, it does not account for nuclear properties such as deformation, rotation, and vibration.

### 3.1.5 Macroscopic Deformation, Rotation, and Vibration

When picturing the nucleus as an incompressible liquid drop, it is also easy to imagine that such a liquid drop can be deformed, rotated, or made to vibrate. These modes of excitation are particularly interesting because in the case of even-even nuclei they are the only available excited states below the pairing gap (\( \approx 1 \text{MeV} \)). In general, nuclei that have closed shells, both proton and neutron numbers close to the magic numbers 2, 8, 20, 28, 50, 82, 126, and 184, are spherical. However, nuclei that are located far away from these numbers are in either the prolate or oblate deformation. Prolate deformations are most often described as football shaped, whereas oblate deformations are considered to be door knob shaped. The quadrupole deformation of a nucleus can be characterized simply by describing the nucleus in terms of an ellipsoid shape. In such a description the deformation parameter \( \beta \) is defined by:

\[
\beta = \frac{4}{3} \sqrt{\frac{\pi}{5}} \frac{(b - a)}{R_{\text{avg}}}
\]  

(3.8)

where \( b \) and \( a \) are the semi-major and semi-minor axes and \( R_{\text{avg}} \) is the average radius. For prolate nuclei \( \beta \) is positive, whereas for oblate nuclei it is negative. In the literature, however, deformations are generally reported with the deformation parameter developed by Nilsson, \( \epsilon \), which is related to \( \beta \) via the relation \( \beta \approx 1.05\epsilon \). Higher order deformation, such as octupole, deformations are possible as well and will become important for octupole vibrations later on. The order of the deformation is denoted by a subscript on the deformation parameter \( \lambda \), 2 for quadrupole and 3 for octupole deformations.
The energy levels of a rotating nucleus can be described via the relation:

\[ E_{\text{rot}} = \frac{J(J+1)\hbar^2}{2I} \]

where \( J \) is the rotational angular momentum quantum number and \( I \) is the moment of inertia of the nucleus. However, it is very difficult to predict the energies of rotational states of a nucleus because it is not possible to predict the moment of inertia. This is due to the fact that in most cases the nucleus doesn’t behave like a perfect rotor. While the deformed moment of inertia can be determined, the experimental values are often lower by a factor of two to three. This indicates a significant violation of the assumed independent motion of the nucleons. Rotations of the nucleus can be based on any of its nuclear excitations such as vibrations or single particle excited states. The moment of inertia can be different for each of these rotations due to varying deformations or numbers of unpaired nucleons.

The final form of collective excitations considered here are nuclear vibrations. These are geometric vibrations around the ground state and can either be quadrupole, octupole or higher order vibrations. Dipole vibrations are not possible since they would correspond to a translation of the nucleus. Figure 3.6 shows a schematic two dimensional representation of these different vibrational modes. In even-even nuclei such as fermium-254, the rotational and vibrational states can be standardized as follows. The rotational band based on the 0+ ground state has spacings of two angular momentum units. This is due to the fact that a single step in angular momentum would violate the symmetry of the nuclear wave function. However, once the nucleus is excited into a vibrational state that symmetry by default is broken and rotational states of vibrational excitations only differ by one angular moment unit. Therefore, in general, quadrupole vibrations have a rotational band based ground state of 2+. Rotational quanta with \( l = 0, 1, 2, 3, \ldots \) result in a total angular momentum of 2+, 3+, 4+, 5+... In contrast octupole vibrations have a rotational bandhead of 3-, 2-, 1-, or 0- in order of decreasing likelihood. In the case of fermium-254 the octupole
vibrational ground state is a 2- state; therefore, the band would be 2-, 3-, 4-... In the case of rotations based on vibrational states the rotational spacing is now determined by the relation:

$$E = \frac{\hbar^2}{2I} [J(J + 1 - K^2)]$$  \hspace{1cm} (3.10)

where the only difference is the $K^2$ term which represents the bandhead of the vibrational state. $K$ is defined as the sum of the individual single particle angular momenta ($\sum_i \Omega_i$) in multi-particle excitations. After octupole vibrations single particle excited states generally take over.

Measuring and understanding Nilsson numbers, K-values, vibrational, and rotational energies of nuclei at the extremes of the nuclear landscape is essential to advancing and constraining nuclear models. Particularly in the region of the neutron deficient actinides there is a dearth of nuclear structure data. Exploring this region will enable theorists to examine trends in nuclear properties and enhance our understanding of these nuclei in general.

### 3.2 Previous Experiments on the Structure of Fermium Isotopes

Fermium was originally discovered in 1952 as part of the debris of a hydrogen bomb explosion \cite{GTH+55}. It has also been produced in fusion evaporation reactions and multiple neutron capture on $^{252}$Cf \cite{HTGC54}. The following was previously known about the nuclear structure of fermium isotopes near the neutron shell $N=152$.

#### 3.2.1 Fermium 250

Greenlees \textit{et al.} \cite{GHK+08} studied the deformed shells around $N=150$ in order to develop a theoretical model with which to predict the shell structure of even heavier elements. Fermium-250 was produced in the reaction $^{204}$Hg($^{48}$Ca, 2n)$^{250}$Fm at the University of Jyväskylä. Fusion evaporation products were recoil tagged in the Recoil Ion Transport Unit (RITU) gas filled separator and prompt $\gamma$-rays were detected with JUROGAM. By summing conversion electrons with $\gamma$-rays they were able to measure the ground state rotational, the 8- isomeric, and the 2- vibrational states. Figure 3.7 below shows the level scheme measured in this experiment.

The authors were able to measure highly excited states since they were measuring $\gamma$-rays directly at the location of the fusion evaporation reaction. The authors also
Figure 3.7: Partial level scheme of fermium-250 measured by Greenlees et al. [GHK+08] via in beam $\gamma$-spectroscopy. M1 transitions are labeled for clarity even where they were not observed.
compared the 2- vibrational level to analogous levels in $^{250}$Californium and $^{246}$Curium [FAP$^+77$][MTMM71].

### 3.2.2 Fermium 254

The nuclear structure of fermium-254 was first studied by Unik et al. [UDV62]. They observed the population of several levels via the decay of meta stable einsteinium-254. Unik et al. produced large quantities of einsteinium-254 by irradiating curium-246 with a high flux of neutrons. The einsteinium was then chemically isolated and measured in a high yield beta spectrometer. All transitions in their spectrum were measured via internal conversion spectroscopy and had relatively large errors. They identified the 2+ excited state at $692 \pm 2$ keV and its first rotational band member at $734 \pm 4$ keV. Additionally they measured hints of $\gamma$-rays with energies of $989 \pm 7$ and $1010 \pm 20$ keV. However, it was later determined that these $\gamma$-rays were a part of the $\beta^-$-decay of $^{250}$Bk, the $\alpha$-decay daughter of $^{254}$Es and are not part of the fermium-254 level scheme shown in figure 3.8.

Porter and Freedman made more precise measurements ten years later [PF71]. Porter et al. prepared einsteinium-254m via a neutron irradiation of 0.4 $\mu$g of einsteinium-253. The einsteinium was chemically isolated from its fission products and the material was deposited onto an aluminum foil. They measured the level energies discovered by Unik et al. to much higher precision through the use of the Argonne toroidal-field spectrometer [FWP$^+60$] with errors on the eV scale.

![Figure 3.8: Level scheme of $^{254}$Fm prior to this work taken from Unik et al. [UDV62].](image-url)
Ahmad et al. [ADM+73][AL84] confirmed these energies in a series of experiments using Ge(Li) detectors. The source was produced in an analogous fashion to the one used by Porter. However, neither Porter et al. nor Ahmad et al. make any mention of higher lying excited states.

### 3.2.3 Fermium 256

The isotope studied in most detail in this region is fermium-256. Originally discovered in 1976, Lougheed et al. [LLH+76] proposed a simple level scheme of fermium-256 based on the beta decay of einsteinium-256m. Subsequently experiments were performed with greatly increased accuracy. The level scheme of fermium-256 shown in figure 3.9 was measured by Hall et al. [HGH+89] while studying the $\beta$-delayed fission properties of einsteinium-256m.

The authors chemically purified a 0.1 $\mu$g sample of $^{254}$Es and electrodeposited it in a 0.2 cm diameter circle on a palladium coated beryllium foil. This target was then irradiated with a 16 MeV tritium beam for approximately 7 hours to produce 7.6 hour
$^{256m}$Es in the reaction $^{254}$Es(t,p)$^{256m}$Es. The reaction products were caught on a gold catcher foil and then counted in $\beta - \gamma$ and $\gamma - \gamma$ coincidence detection apparatuses after another round of chemical purification. This level scheme is an excellent guide to the level scheme of fermium-254 and most of the structures should appear in both. Features of particular interest are the 2+ $\gamma$ vibration at 682.3 keV, the 2- octupole vibration at 881.8 keV, and the 3+ particle excitation at 1100.0 keV.

3.3 Experimental Setup

Nuclear structure measurements were carried out utilizing the 88-inch cyclotron described in section 1.2 and the BGS described in section 1.3. Both the C$^3$ detector as well as the RTC were employed for these experiments. The beam was produced at LBNL using the Versatile ECR ion source for NUclear Science (VENUS) [LLL$^\dagger$06] and accelerated through the 88-inch cyclotron. It was then guided to the BGS target area where it hit the target wheel. The target wheel generally rotated at a frequency of 11 Hz and the product of beam current and target thickness was monitored via Rutherford scattering measured in two independent silicon PIN diode detectors mounted at a 27$^\circ$ angle to the beam line.

Compound nucleus evaporation residues then recoiled out of the target and were separated from unreacted beam and other reaction products in the BGS based on their different magnetic rigidities in 500 millitorr of helium. The separated products then implanted in the detector at the end of the BGS where their decay signature was measured. The $\alpha$-decays of the reaction products were measured in the Corner Cubed Clover (C$^3$) detector and used to center the beam. After centering the reaction products in the focal plane, the C$^3$ detector was replaced with the Berkeley Recoil Transfer Chamber (RTC) [Kir99, SCD$^\dagger$06, KFGG02, SGA$^\dagger$05] which is described in much greater detail in section 3.4. A honeycomb structure has been machined to support a 6 $\mu$m Mylar foil as the interface between the BGS and the RTC. This foil can easily be changed or replaced. The differential pressure between the BGS and the RTC was kept at close to 1500 torr.

The RTC was flushed with helium and KCl aerosol clusters which were produced by heating KCl crystals to 640$^\circ$ Celsius in a tube furnace through which high purity helium was being flushed.

After the EVRs were thermalized in the RTC, they became attached to the KCl aerosol particles and were efficiently transported through a gas-jet capillary away from the high $\gamma$-background (20 kHz per crystal) caused by interactions of the beam with the beam stop and other materials in the BGS. The EVRs were transported to the low background counting setup which consisted mostly of 10 cm thick lead walls around two clover style $\gamma$-detectors [DBT$^\dagger$99] facing each other as shown in more
Standard mixed $\gamma$-ray sources were used for energy and efficiency calibrations and a total photopeak efficiency of 41.83\% at 122 keV was measured. Both calibrations were processed in Radware as described by Radford [Rad95]. No addback was performed; all crystals were treated individually.

### 3.4 Recoil Transfer Chamber

A recoil transfer chamber is a device designed to separate the products of nuclear reactions from the harsh conditions under which they are produced. In the case of heavy element research, RTCs are placed at the focal plane instead of the standard silicon chip detectors. While the underlying concepts and ideas for RTCs have been around since the 1970’s [Wol76, TBS87], they were mostly used to study fission and other decay products. The first RTC to study the super heavy elements was developed in Berkeley in 1999 [Kir99]. The goal of the Berkeley RTC was to stop and transport super heavy elements to one of several apparatuses to study their chemistry. Originally RTCs were placed right behind the target. However, this led to very low gas jet efficiencies due the high energy of the beam. Specifically efficiency...
drops due to convection in the heated gas inside the RTC as well as the destruction of aerosol clusters. Additionally, RTCs at the target also transport activities produced in unwanted side reactions, making $\gamma$-ray studies for sub millibarn cross-sections very difficult. Consequently, the Berkeley RTC was placed at the end of the BGS where evaporation residues have already been removed from the majority of unwanted reaction products, the beam, as well as the plasma produced by the beam. Figure 3.11 shows the RTC attached to the end of the BGS instead of the silicon strip detectors.

Figure 3.11: Image of the focal plane area of the BGS with the recoil transfer chamber in place.

This system was designed and successfully used to study the chemical properties of some of the heaviest known elements. There are several technical challenges in providing an interface between the BGS and the experimental environment:

- In order to stop the reaction products after physical pre-separation they need to be thermalized in a gas cell. The barrier between the BGS and the gas cell needs to allow the EVRs to pass through while still maintaining structural integrity over a high pressure differential. In case of structural failure, the targets of the nuclear fusion reaction would be very likely to blow out due to the rapid expansion of the stopping gas into the BGS.

- The thermalized EVRs need to be transported to a low background area such that the high background in Cave 1 does not interfere with measurements.

The Berkeley RTC [SCD+06, KFGG02, SGA+05] is a $322 \text{ cm}^3$ chamber machined from stainless steel. There are six inlets on the top, bottom left and right side of the
chamber through which high purity helium containing KCl aerosols is flushed into the chamber. The outlet is located at the back of the chamber. A honeycomb structure has been machined to support a Mylar foil as the interface between the BGS and the RTC. It consisted of 5 mm holes arranged in a hexagonal pattern. The BGS usually kept around 0.5 torr and the RTC kept between 760 and 1520 torr. In general, a 6 µm Mylar foil is used however for asymmetric reactions thinner foils were needed. There is a linear relationship between the thickness of the Mylar foil and the pressure at which it fails. Figure 3.12 shows this trend. The measured data points were taken in triplicate with the error being the standard deviation of the three measurements.

![Figure 3.12: Absolute upper pressure limits in the RTC for different thickness of Mylar foil.](image)

These are absolute upper limits and during experimental operation a significant safety margin should be employed since weakening due to the elastically scattered target material as well as reaction products can be expected. Rolls of Mylar foil, for the production of Mylar windows, should not be more than three years old because the foils deteriorate and develop pinholes with age.

Additionally, thinner foils are more permeable to helium and thus cause leakage into the BGS which can lower the efficiency of transporting EVRs to the focal plane. Generally, a 6 µm Mylar foil should be used if possible, however, in each reaction, energy calculations should first be performed in order to verify that the EVRs have enough energy to pass through the foil. These calculations are performed with SRIM [Zie04] as described in section 1.2.1. Performing these calculations also yields the residual energy that the EVRs have after passing through the foil. This energy is necessary to determine the range of the products in helium to verify that they can be stopped before colliding with the back wall of the chamber.

After the atoms have been thermalized in the RTC they need to be transported
toward the detection area. This is accomplished via a KCl-aerosol gas jet. The aerosol particles are produced by sublimating KCl in a tube furnace and flushing it into the RTC with a helium jet. Thermalized reaction products will attach themselves to the KCl clusters and under the correct conditions these particles can be transported to an experimental area through a capillary tube where they will be further analyzed.

Aerosol transport efficiency is often difficult to optimize but can be understood based on the following theoretical considerations. The distance that a cluster that is moving through a capillary diffuses towards the side walls to which it can attach itself in an arbitrary time interval is given by:

$$d_{\text{dif}} \propto \sqrt{T/m}$$  \hspace{1cm} (3.11)

where $d_{\text{dif}}$ is the diffusion distance in capillaries, $T$ is the temperature, and $m$ is the mass of the cluster. A derivation of this equation is given by Wollnik et al. [Wol76]. The diffusion distance can be minimized by lowering the temperature, $T$, or by increasing the mass of the aerosol. Since lowering the temperature of long capillary is not feasible the mass of the particle must be controlled well in order to ensure high efficiency. However, if the clusters become too large, they will deposit on the side walls due to gravitational effects. According to Wollnik et al. this effect is proportional to the mass, $m$, as follows:

$$d_{\text{grav}} \propto m^{2/3}$$  \hspace{1cm} (3.12)

where $d_{\text{grav}}$ is the diffusion distance due to gravitational effects and $m$ is the aerosol mass again. With temperature being constant, the diffusion distance in capillaries, $d_{\text{dif}}$, decreases with mass, whereas the diffusion distance due to gravity, $d_{\text{grav}}$, increases with aerosol mass. Therefore, there is an ideal mass which minimizes to total diffusion distance. This mass was calculated by Wollnik [Wol76] and correlated to a particle of size 0.1 to 1 µm for the case of KCl aerosols.

Such clusters can be produced by sublimating KCl around 640 degrees Celsius. Alternatively, a commercial KCl aerosol generator can be utilized. At the BGS KCl particles are produced by letting high purity helium in-line run through a tube furnace filled with KCl crystals that is kept at 640 degrees Celsius. As the sublimated KCl cools down when leaving the tube furnace it produces KCl clusters. The temperature of the tube furnace has independently been determined to be the ideal temperature in order to maximize efficiency by several labs. Figure 3.13 below shows the dependence of efficiency on KCl oven temperature measured at Berkeley by Düllman et al. [DFG+05]. This is in agreement with similar curves measured at Riken by Haba et al. [HKK+07]. This temperature was verified in this work and is described in greater detail later in this chapter. Taking all separate measurements into account, the peak efficiency was also determined to be at 640 degrees Celsius.

KCl clusters are not just created at one specific size, but rather a size distribution.
Therefore, after the helium jet passes through KCl generator, it is passed through a 15.3 meter long 6.4 mm Polytetrafluoroethylene (PTFE) tubing. This serves as a sedimentation loop which removes KCl clusters that are too large and would lower the efficiency of the transport as they are more likely to collide with and attach to the wall. This is purely a gravitational effect and ensures that only those aerosols that can actually travel through the capillary make it to the RTC. With the RTC optimized, the reaction products of interest are now trapped in the coincidence detection setup between two clover detectors.

3.4.1 Clover Detector Setup

Semiconductor detectors where first developed in the 1960s. Since then, production and purification technologies have greatly advanced, and greatly improved detectors have been developed. By far the most significant improvement was the development of high purity germanium detectors. These detectors have a much smaller band gap as opposed to their sodium iodide predecessors. This gives them a much higher resolution. Larger effective volumes also allow them to detect $\gamma$-ray energies up to a few MeV. High purity germanium detectors have to be cooled down with liquid nitrogen because at room temperature thermal motion would be enough to excite electrons across the band gap. High purity germanium detectors were used in some of the earliest $\gamma$-ray tracking arrays such as TESSA 3, GASP [Alv93], EUROGAM I [Bec92]. Early attempts to improve these arrays largely failed because their larger crystal sizes suffered from sensitivity to neutron damage. Additionally, such large

![Figure 3.13: Efficiency dependence on temperature in the KCl oven. Lines are drawn to guide the eye. Taken from [DFG+05].](image)
crystals struggled with Doppler broadening of $\gamma$-rays due to being placed up to 90° with respect to the beam directions. These issues were addressed with the development of segmented high purity germanium detectors. These detectors, called clover detectors [DBT+99], are now used in the most modern $\gamma$-ray tracking arrays such as GRETINA [LCC+04]. Clover detectors contain four separate crystals in one cryostat allowing for larger germanium volumes and a great photopeak efficiency.

Figure 3.14 shows the arrangement of these $\gamma$-detectors in the experimental setup. They are connected to a common ground and in most cases to a common bias. The crystal assembly as well as 4 independent pre-amplifiers are contained in a 1.5 mm thick aluminum cryostat which is pumped down to approximately $10^{-8}$ torr. Germanium detectors are generally shielded from background radiation with lead shielding.

### 3.4.2 Shielding and Trap Design

**Lead Castle**

In the absence of anti coincidence shields 10 centimeters of lead were used to minimize the effects of background radiation. By moving the setup to a low background
area on top of Cave 2 at the 88-inch cyclotron and optimizing the stacking of lead bricks, background rates were lowered from 200 to 15 Hz. For comparison purposes during beam on target conditions, a clover in Cave 1 has rates on the order of 20000 Hz.

At these low event rates accidental coincidences due to background are minimized to 0.5 to 1 coincident events per second. This rate is even lower when only considering events in opposing clovers.

**Aerosol Trap Design**

The traps, shown in figure 3.16, to catch the aerosols were built using empty 3 ml syringe cartridges with two frits (pre-inserted) produced by Agilent Technologies (Part # 12131014). The tip on the exit side of the syringe is roughed up with sandpaper to improve the epoxy bond between the syringe and a piece of 6.4 mm PolyTetraFluoroEthylene (PTFE) tubing. The syringe reservoir is then filled with 12.7 mm of activated charcoal which is sealed in with a small amount of fiberglass wool. The trap is connected into the main gas jet system with a rubber stopper that has a 3.2 mm PTFE tube passed through a pre-drilled hole. These filters are highly effective due to high surface area as well as excellent vacuum tightness. Previous
generations of traps clogged with KCl aerosols after approximately twenty minutes. These aerosol traps last up to three hours.

### 3.4.3 Data Acquisition System

Clover detectors contain an internal pre-amplifier powered by a ± 12 Volt 9-pin D-Sub cable. Pre-amplified signals are then routed into the BGS counting shack into a MSCF-16 Mesytec spectroscopy module via four Lemo cables per clover. The MSCF-16 unit combines 16 channels of shapers, timing filter amplifiers and constant fraction discriminators. Having all of these combined into a single module greatly simplifies the BGS data acquisition system over its previous generations.

### 3.4.4 Calibration and Efficiency Measurements

Working with isotopes produced in fusion reactions is often difficult due to very low production rates. Our transport system has been optimized for high throughput. However, due to the nature of this type of spectroscopy, the overall efficiency is only around 2%. This efficiency was calculated from the individual components of the system consisting of: the BGS (60%) [GLP+05], RTC window (80%), gas jet (85%), \(\gamma\) efficiency at 200 keV (40%) and gating conditions [67.5%]. Total geometric and intrinsic efficiency of the detector setup is shown in figure 3.17. To further characterize the experimental setup it was tested with a high cross-section reaction.
3.5 Characterization Experiment - Production of Erbium-154

The purpose of this experiment was three fold; to measure the attenuation of the degraders in front of the Rutherford detectors, to measure the absolute efficiency of the BGS and to attempt to measure and optimize the gas-jet transport efficiency into Cave 2. Because no coincident $\gamma$ and x-rays were detected in previous experiments a reaction with a much larger cross-section was chosen in order to show that these types of experiments can yield $\gamma$-ray spectra of quality high enough to propose level schemes. In regard to the coincident $\gamma$ and x-ray experiment the following criteria were used to determine the reaction:

- The beam needs to be heavy in order to be able to use a 6$\mu$m Mylar foil in the RTC window.
- The cross section needs to be high to produce large amounts of $\gamma$-activity.
- The product needs to decay by both alpha and electron capture decay. Alpha decay is necessary in order to identify the products via recoil-alpha correlations in the $C^3$ detector. And the electron capture decay is necessary in order to reproduce the experimental conditions for finding an x-ray $\gamma$-ray coincidence in the germanium detector setup.

Through an iterative process of looking at many target and projectile combinations the following two reactions were chosen: $^{118}\text{Sn}(^{40}\text{Ar}, 6n)^{152}\text{Er}$ first reported by Gauvin
and $^{165}$Ho$(^{40}$Ar, 4$n$)$^{201}$At. These reactions are ideal candidates for showing proof of principle as well as to characterize the BGS efficiency and Rutherford attenuation coefficients. The decay schemes of $^{152}$Er and $^{201}$At are shown in Figure 3.18 below: With the properties listed in table 3.1.

![Decay schemes for Er-152 and At-201](image)

Figure 3.18: Decays schemes for $^{152}$Erbium and $^{201}$Astatine showing branching ratios and half lives.

<table>
<thead>
<tr>
<th>Reaction Product</th>
<th>Er-152</th>
<th>At-201</th>
</tr>
</thead>
<tbody>
<tr>
<td>Half-life</td>
<td>10.3 seconds</td>
<td>85.2 seconds</td>
</tr>
<tr>
<td>Branching Ratio</td>
<td>10% EC and 90% $\alpha$</td>
<td>29% EC and 71% $\alpha$</td>
</tr>
<tr>
<td>Coulomb barrier</td>
<td>129 MeV</td>
<td>162 MeV</td>
</tr>
<tr>
<td>Range in Mylar</td>
<td>11.2 $\mu$m</td>
<td>7.1 $\mu$m</td>
</tr>
<tr>
<td>Center of Target</td>
<td>178 MeV</td>
<td>178 MeV</td>
</tr>
<tr>
<td>Lab Frame Energy</td>
<td>unknown</td>
<td>unknown</td>
</tr>
<tr>
<td>Cross-section</td>
<td>70 mb</td>
<td>unknown</td>
</tr>
</tbody>
</table>

Table 3.1: Decay properties and experimental conditions for the two reactions that were run to characterize the BGS and the gas transport system.

The efficiency of the BGS was measured by plunging an aluminum foil just behind the target where it collected all EVRs from the argon + holmium reaction. The plunger was then pulled back out, in the process moving the foil in front of an alpha detector. An absolute efficiency was obtained by comparing the event rate in the focal plane C$^3$ detector to the event rate on the plunged foil after decay and efficiency corrections. Figure 3.19 shows a schematic of the plunger setup which is explained in more detail in the following section.

Additionally, the attenuation of the Rutherford degraders was measured by comparing the Rutherford rates with and without the degrader normalized to the focal

72
plane detector event rate. In summary, this experiment characterized the current state of the BGS and verified that the transport of EVRs to the detection setup was effective and efficient.

### 3.5.1 Attenuator and Efficiency Measurements

The amount of activity on the catcher foil is a function of the production rate, the plunging frequency and the half-life of produced nucleus. The two equations determining the buildup and subsequent decay of activity are:

\[
A_0 = R(1 - e^{\lambda t_{irr}}) \quad (3.13)
\]

and

\[
A_d = A_0 e^{\lambda t} \quad (3.14)
\]

Here \( R \) is the rate of production, \( A_0 \) is the activity at the end of \( t_{irr} \), \( A_d \) is the activity during the decay interval, \( \lambda \) is the decay constant, \( t_{irr} \) is the irradiation time, and \( t \) is the time during the decay measurement interval. For the following calculations \( R \) is normalized such that the saturation activity is 1. Plotting these two equations then yields the following graph for the expected \(^{201}\)At activity. The plunging program was set to a 60 second irradiation followed by 120 seconds of decay, the first 60 of which were used to record an alpha spectrum on the PIN diode at the front of the BGS. The remaining 60 seconds were used to let most of the remaining activity decay away. Due to the plungers movement speed, there is a 3.2 second delay that shortens the actual irradiation and measurement to 56.8 seconds.
Figure 3.20: Theoretical activity during plunger measurements for the argon + holmium reaction. The shaded area shows the time during which the α-decay data were being recorded.

In figure 3.20 the shaded area shows the time interval in which the plunger is in front of the PIN diode and α-decay data are recorded. The integral of this area is used to compare the measured alpha rate to the actual production rate in the BGS. It is used to calculate the plunge correction factor shown in table 3.2 below. The measurement in the PIN diode yielded the alpha spectrum shown in figure 3.21.

Figure 3.21: Alpha diode spectrum during the $^{165}$Ho($^{40}$Ar, 4$n$)$^{201}$At efficiency test.

To determine the efficiency of the BGS, the rate determined from this diode was compared to the rate determined at the focal plane of the BGS. The broad peaks are a result of varying implantation depth in the aluminum foil. Figure 3.22 below shows the integrated alpha peak at the focal plane.
Figure 3.22: Focal Plane Alpha Spectrum for the $^{165}$Ho($^{40}$Ar, $4n$)$^{201}$At reaction.

The peak integrals were then normalized by the correction factors shown in table 3.22 to compare them directly. Normalized rates are determined via the following equation:

$$A_n = \frac{N_\alpha}{\epsilon_p \epsilon_{det} t_{irr} N_r}$$

(3.15)

where $N_\alpha$ is the total number of $\alpha$-decays measured, $\epsilon_p$ is the efficiency of the plunging apparatus, $\epsilon_{det}$ is the efficiency of the detector, $t_{irr}$ is the total irradiation time, and $N_r$ is the number of Rutherford events. The efficiency from the plunging apparatus is determined from figure 3.20 as described above. A geometric calculation was performed to determine the detector efficiency for the PIN diode and the efficiency of the focal plane detector is well understood from previous experiments. The rate is then normalized to the differing irradiation times and finally it rate is normalized to the beam intensity via the Rutherford integral.

The ratio of the normalized rates is the transmission efficiency of the BGS. For the $^{165}$Ho($^{40}$Ar, $4n$)$^{201}$At reaction $69 \pm 3\%$ was measured, which agrees with previously measured and simulated BGS efficiencies.
Table 3.2: Measured, corrected and normalized $^{201}$At alpha rates used to determine the BGS efficiency.

<table>
<thead>
<tr>
<th></th>
<th>Peak Integral</th>
<th>Calculated Plunge Correction</th>
<th>Efficiency</th>
<th>Irradiation Time [s]</th>
<th>Rutherford Integral</th>
<th>Normalized Rate per Rutherford per Second</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alpha Diode</td>
<td>1192 ±34.5</td>
<td>0.603</td>
<td>0.005</td>
<td>1858</td>
<td>-4948</td>
<td>0.043±0.0014</td>
</tr>
<tr>
<td>Focal Plane</td>
<td>79602 ±282</td>
<td>N/A</td>
<td>0.85</td>
<td>1288</td>
<td>2403</td>
<td>0.030±0.0006</td>
</tr>
</tbody>
</table>

Figure 3.23: Schematic diagram of the Rutherford detectors and their attenuators in the target area of the BGS. The attenuators are labeled with "U" for upstream and "D" for downstream. The detectors are located at a 27° angle with respect to the incoming beam.

**3.5.2 Rutherford Attenuator Measurements**

During irradiations, the integrated product of the beam intensity and target thickness is monitored via Rutherford scattering of the beam particles in the target. The Rutherford detectors have to be protected via attenuators since the high rate of scat-
tered particles would otherwise overwhelm and damage the PIN diodes. Figure 3.23 illustrates the positions of the attenuators and PIN diode detectors relative to the beam. In this experiment the attenuation of each of the four Rutherford attenuators was measured. There are two Rutherford detectors, Rutherford West and Rutherford East, and each of them has two sets of attenuators. Each attenuator consists of two 1/6 transmission foils mounted inside of a NW25 quick flange gasket. The theoretical total attenuation is, therefore, 6^4, or 1296, and has previously been measured to be 1348 ±20 [Fol04].

In order to measure the attenuation factor the BGS was run with all possible attenuator combinations; first the reaction was run with both attenuators removed, then with just the upstream attenuators, and finally with just the downstream attenuators. The focal plane alpha rate was then used to normalize the Rutherford rates and the ratio of these normalized rates with and without attenuators corresponds to the attenuation factor. Table 3.3 shows the collected data and the calculated attenuation coefficients for the argon + tin reaction.

<table>
<thead>
<tr>
<th>Attenuator Position</th>
<th>Rutherford Events</th>
<th>Alpha Events</th>
<th>Rutherford/Alpha</th>
<th>Attenuation Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>No Attenuators East</td>
<td>64746±254</td>
<td>117502±343</td>
<td>0.551(3)</td>
<td>N/A</td>
</tr>
<tr>
<td>No Attenuators West</td>
<td>61300±247</td>
<td>117502±343</td>
<td>0.522(3)</td>
<td>N/A</td>
</tr>
<tr>
<td>Upstream East</td>
<td>1607±40</td>
<td>103557±322</td>
<td>0.0155(4)</td>
<td>35.5±0.91</td>
</tr>
<tr>
<td>Upstream West</td>
<td>1574±40</td>
<td>103557±322</td>
<td>0.0152(4)</td>
<td>34.3±0.89</td>
</tr>
<tr>
<td>Downstream East</td>
<td>1198±35</td>
<td>80499±284</td>
<td>0.0149(3)</td>
<td>37.0±1.1</td>
</tr>
<tr>
<td>Downstream West</td>
<td>1205±35</td>
<td>80499±284</td>
<td>0.0150(3)</td>
<td>34.9±1.0</td>
</tr>
</tbody>
</table>

Table 3.3: Rutherford event rates and calculated attenuation coefficients for the \( ^{118}\text{Sn}(^{40}\text{Ar}, 6n)^{152}\text{Er} \) reaction ran at a center of target energy of 178 MeV.

The same procedure was repeated for the argon + holmium reaction and yielded the data shown in table 3.4.

Multiplying the attenuation coefficients for upstream and downstream detectors together yields the total attenuation coefficients shown in table 3.5. This table also shows the average result taking both reactions into account. Within the counting errors this agrees with previously measured attenuation factors, thus verifying that the attenuators and Rutherford detectors are functioning as expected.

### 3.5.3 Gas Transport Efficiency Tests

After the efficiency and Rutherford degrader experiments the gas transport system was characterized. In the \( ^{118}\text{Sn}(^{40}\text{Ar}, 3−6n)^{155−152}\text{Er} \) reaction, the tube furnace
Table 3.4: Rutherford event rates and calculated attenuation coefficients for the $^{165}$Ho($^{40}$Ar, 4$n$)$^{201}$At reaction.

<table>
<thead>
<tr>
<th></th>
<th>Rutherford Events</th>
<th>Alpha Events</th>
<th>Rutherford/Alpha</th>
<th>Attenuation Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>No Attenuators East</td>
<td>3383±58</td>
<td>146436±383</td>
<td>0.0231(4)</td>
<td>N/A</td>
</tr>
<tr>
<td>No Attenuators West</td>
<td>3570±59</td>
<td>146436±383</td>
<td>0.0243(4)</td>
<td>N/A</td>
</tr>
<tr>
<td>Upstream East</td>
<td>318±18</td>
<td>523519±724</td>
<td>0.00061(3)</td>
<td>38.0±2.2</td>
</tr>
<tr>
<td>Upstream West</td>
<td>357±19</td>
<td>523519±724</td>
<td>0.00068(4)</td>
<td>35.8±2.0</td>
</tr>
<tr>
<td>Downstream East</td>
<td>313±18</td>
<td>533609±730</td>
<td>0.00059(3)</td>
<td>39.4±2.3</td>
</tr>
<tr>
<td>Downstream West</td>
<td>364±19</td>
<td>533609±730</td>
<td>0.00068(4)</td>
<td>35.7±2.0</td>
</tr>
</tbody>
</table>

Table 3.5: Rutherford Attenuator Results.

<table>
<thead>
<tr>
<th></th>
<th>$\text{Ar + Sb}$</th>
<th>$\text{Ar + Ho}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total East</td>
<td>1314 ± 51</td>
<td>1497 ± 130</td>
</tr>
<tr>
<td>Total West</td>
<td>1196 ± 47</td>
<td>1278 ± 100</td>
</tr>
<tr>
<td>Average East</td>
<td>1341±47</td>
<td></td>
</tr>
<tr>
<td>Average West</td>
<td>1210±42</td>
<td></td>
</tr>
</tbody>
</table>

The three temperatures used were 625°C, 650°C, 675°C, with 640°C being the standard operating temperature based on past experiments. Data were collected for each of these temperatures and, in order to compare the relative efficiencies, the number of 295.8 keV $\gamma$-rays (from one of the decay daughters of $^{153}$Er) was normalized to the Rutherford count for each run. The data, presented below in table 3.6 and figure 3.24, show that the gas jet was slightly more efficient at 625°C. More data points should be collected here to determine whether even lower temperatures increase the efficiency as these data are somewhat in contradiction to previously measured efficiencies. The slightly higher efficiency at 625°C may simply be a miscalibration in the temperature sensor of the oven. The decrease of efficiency at higher temperatures can likely be attributed to the formation of larger aerosol clusters which have a lower transport efficiency through the capillaries.
<table>
<thead>
<tr>
<th>Temperature</th>
<th>Rutherford Counts</th>
<th>295.8 keV Peak Integration</th>
<th>γ-rays per Rutherford</th>
</tr>
</thead>
<tbody>
<tr>
<td>625°C</td>
<td>1767±42</td>
<td>14712±121</td>
<td>8.32±0.2</td>
</tr>
<tr>
<td>650°C</td>
<td>3310±57</td>
<td>26860±164</td>
<td>8.11±0.1</td>
</tr>
<tr>
<td>675°C</td>
<td>1275±36</td>
<td>9884±99</td>
<td>7.75 ±0.2</td>
</tr>
</tbody>
</table>

Table 3.6: Relative efficiency of the gas jet at several temperatures.

![Graph showing the relationship between temperature and relative efficiency](image)

Figure 3.24: Relative efficiency dependence on furnace temperature in the aerosol generator (Line drawn to guide the eye).

Next, keeping the temperature constant at 640°, three different gas jet pressures were used in an attempt to determine if a lower gas jet pressure can be used for future experiments since less symmetric reactions produce recoils with less energy and require thinner Mylar windows which will not be able to handle a pressure differential of 1.5 atm. The standard 1.5 atm was used as well as 1.0 atm and 0.5 atm and the results are as follows in table 3.7 and figure 3.25. It appears a lower gas jet pressure causes a dramatic drop in gas jet efficiency. There are several possible explanations for the efficiency drop at lower pressures/gas flow rates. It is possible that at the lower RTC pressures the EVRs are not slowed down fast enough and impact the back of the RTC. If this is the case Mylar degraders could be used in order to lower their energy to a point where they are stopped in the RTC. Alternatively, it is possible that at lower gas flow rates the flow through the traps is laminar and the EVRs attached to aerosols are able to pass through. At higher flow rates the flow would be more
turbulent and the aerosols impact on the filter and get stuck. In either case, using thin Mylar windows should be avoided if possible.

<table>
<thead>
<tr>
<th>RTC Pressure</th>
<th>Rutherford Counts</th>
<th>295.8 keV Peak Integration</th>
<th>γ-rays per Rutherford</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5 atm</td>
<td>1917±44</td>
<td>587±24</td>
<td>0.31±0.01</td>
</tr>
<tr>
<td>1.0 atm</td>
<td>1980±44</td>
<td>7434±86</td>
<td>3.75±0.09</td>
</tr>
<tr>
<td>1.5 atm</td>
<td>3312±57</td>
<td>26860±164</td>
<td>8.11±0.15</td>
</tr>
</tbody>
</table>

Table 3.7: Relative efficiency of the gas jet at several RTC pressures.

In addition to optimizing the temperature and gas pressure/flow of the system the resulting reaction products from the argon+tin reaction were also measured in the coincidence setup. Figure 3.26 shows γ-rays that were measured in coincidence with dysprosium x-rays. The level scheme of the even-even nucleus $^{154}$Dy, produced in the decay of $^{154}$Er, is very well known [ZHHS80] and the major peaks measured by Zolnowski were replicated in this gas jet experiment. In addition to the transitions in $^{154}$Dy, γ-rays in $^{153}$Dy and $^{155}$Dy from the electron capture decay of $^{153}$Ho and $^{155}$Ho were also observed. Table 3.8 lists the major peaks and their intensity measured in this work. Their corresponding angular momentum transitions taken from Zolnowski are also shown. The same level scheme Zolnowski describes can be deduced from this work.

The decay from $^{154}$Ho is a particularly effective system to study due to the fact that it decays from an 8+ state. It thus has access to the high lying states in the

Figure 3.25: Experimental efficiency dependence on RTC pressure/gas flow rate. Line drawn to guide the eye).
Figure 3.26: Representative γ-rays measured in coincidence with dysprosium K-x-rays. Transitions in $^{154}\text{Dy}$ are shown in black, transitions in $^{153}\text{Dy}$ are shown in red, transitions in $^{155}\text{Dy}$ are shown in blue.

Table 3.8: Major peaks following the $^{154}\text{Ho}$ electron capture and $\beta^+$ decay. Level energies are taken from Zolnowski et al. [ZHHS80] and have undergone minor corrections since the original publication.

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>Counts</th>
<th>Transition</th>
<th>$E_i \rightarrow E_f$ [ZHHS80]</th>
</tr>
</thead>
<tbody>
<tr>
<td>326.4 ± 0.08</td>
<td>108.0 ± 10.4</td>
<td>660.8 $\rightarrow$ 334.6</td>
<td>0+ $\rightarrow$ 2+</td>
</tr>
<tr>
<td>334.9 ± 0.01</td>
<td>2234.1 ± 47.3</td>
<td>334.6 $\rightarrow$ 0.0</td>
<td>2+ $\rightarrow$ 0+</td>
</tr>
<tr>
<td>347.3 ± 0.01</td>
<td>32.3 ± 5.68</td>
<td>1251.9 $\rightarrow$ 905.2</td>
<td>4+ $\rightarrow$ 2+</td>
</tr>
<tr>
<td>366.3 ± 0.2</td>
<td>177.9 ± 13.3</td>
<td>1027.1 $\rightarrow$ 660.8</td>
<td>2+ $\rightarrow$ 0+</td>
</tr>
<tr>
<td>398.1 ± 0.2</td>
<td>106.3 ± 10.3</td>
<td>1057.9 $\rightarrow$ 660.8</td>
<td>0+ $\rightarrow$ 0+</td>
</tr>
<tr>
<td>412.8 ± 0.01</td>
<td>453.7 ± 21.3</td>
<td>747.0 $\rightarrow$ 334.6</td>
<td>4+ $\rightarrow$ 2+</td>
</tr>
<tr>
<td>477.3 ± 0.01</td>
<td>175.9 ± 13.26</td>
<td>1224.1 $\rightarrow$ 747.0</td>
<td>6+ $\rightarrow$ 4+</td>
</tr>
<tr>
<td>505.0 ± 0.02</td>
<td>78.5 ± 8.86</td>
<td>1251.9 $\rightarrow$ 747.1</td>
<td>4+ $\rightarrow$ 4+</td>
</tr>
<tr>
<td>524.1 ± 0.01</td>
<td>44.9 ± 6.70</td>
<td>1747.8 $\rightarrow$ 1224.1</td>
<td>8+ $\rightarrow$ 6+</td>
</tr>
<tr>
<td>570.8 ± 0.01</td>
<td>159.7 ± 12.63</td>
<td>905.2 $\rightarrow$ 334.6</td>
<td>2+ $\rightarrow$ 2+</td>
</tr>
</tbody>
</table>

donna daughter. This may be a limitation in other systems where the ground state spin and parity of the parent may restrict the accessible states in the daughter.

In conclusion, this experiment served four purposes: to characterize the BGS efficiency, to measure the Rutherford attenuator coefficients, to verify and optimize the temperature and pressure settings for the RTC, and to show that transport and
subsequent detection of reaction products with this system is possible. It is, therefore, possible to utilize this technique to study neutron deficient actinides.

3.6 Headline Experiment - The Nuclear Structure of Fermium-254

This new measurement technique developed at the BGS was used to study nucleus $^{254}$Fm as a decay daughter of $^{254}$No the fusion evaporation reaction $^{208}$Pb($^{48}$Ca, $2n$)$^{254}$No. This reaction was chosen due to its high cross section of $2.2 \mu b$ [OUL+01] though only $10\%$ of the fusion evaporation products decay via electron capture giving an effective cross section of $220 \text{ nb}$ for the production of the nucleus $^{254}$Fm from the decay of $^{254}$Md[FAB+70]. Low lying states in $^{254}$Fm have previously been studied by Ahmad et al. by measuring $\gamma$-rays following the beta decay of $^{254}$Es[ADM+73].

The level structure measured in this work confirmed the previously known levels as well as adding one additional level, tentatively identified as the octupole vibration in $^{254}$Fm.

The experiment was performed in the same manner as described in section 3.3. A $^{48}$Ca$^{11+}$ beam was produced using VENUS. The ions were then accelerated to a laboratory energy of $219 \text{ MeV}$ at an average intensity of approximately one particle microampere. At the entrance to the BGS [Fol04] the beam passed through $500 \mu g/cm^2$ of $^{208}$PbO$_2$ on a $47 \mu g/cm^2$ carbon backing. The target material was deposited onto four banana shaped segments via vapor deposition. The purity of the material was $97.85\%$. The calculated center of target energy was $216.4 \text{ MeV}$ corresponding to a compound nucleus excitation energy of $22 \text{ MeV}$.

Compound nucleus evaporation residues then recoiled out of the target and were separated from unreacted beam and other reaction products in the BGS. The separated products then implanted in the focal plane detector of the BGS. The large alpha branch of the $^{254}$No evaporation residues provided clear identification of the reaction products in the focal plane detector via their energy signature. The magnetic rigidity was fine tuned by centering the x distribution of $^{254}$No $\alpha$-particles.

After centering the reaction products in the focal plane the C$^9$ detector was replaced with the RTC. A $6 \mu m$ Mylar foil was chosen as the interface between the BGS and the RTC. The differential pressure between the BGS and the RTC were kept at close to 1500 torr. The RTC was flushed with two liters per minute of helium carrying KCl aerosol clusters produced in a tube furnace set to $640^\circ$ Celsius. This setup effectively transported the reaction products to the $\gamma$-detector setup described in section 3.3.
Figure 3.27: Plot of $\gamma$-$\gamma$ coincidences. Left: Any $\gamma$-coincidences with multiplicity two. A few of the diagonal lines due to Compton scattered photons within one clover are highlighted with red ovals. Right: Only $\gamma$-coincidence with multiplicity two where the two $\gamma$-rays were measured in opposing clovers. Coincidences in opposite clovers are highlighted with red ovals.

The detectors were set up as described in figure 3.14. The detectors were run in singles mode recording each photon event and its time stamp. The collected $\gamma$-spectra were then gated on characteristic fermium x-rays to remove natural background. To reduce background caused by Compton scattering within the crystals of the detectors, only events for which one of the detectors measured either a fermium K$_{\alpha 1}$ or K$_{\alpha 2}$ x-ray and the opposing detector also measured a $\gamma$-ray of any energy within 100 ns of each other were considered. Some of this Compton background, and specifically its removal, is highlighted in figure 3.27. In both plots on $\gamma$-$\gamma$ coincidences with a multiplicity of two are shown. In the left plot, the data were sorted to show all of these coincidences. In contrast, in the right plot events where the $\gamma$-coincidences were in opposite detectors are shown. One can see that the vast majority of diagonal lines caused by Compton scattering have been removed.

A 2.5 cm lead block in between the two clovers minimized backscattered photons that could provide a false coincidence. Figure 3.28 shows representative $\gamma$ spectra using this technique. The measured peaks agree with known transitions as well as adding three new transitions to the level scheme of $^{254}$Fm.

The coincidence data from this experiment makes it possible to propose a more complete level scheme than the ones determined in previous experiments [ADM$^+$73]. The electron capture decay of $^{254}$Md populated a previously unknown level at 985.7 keV which decayed via 251.9, 292.0 and 940.7 keV $\gamma$ rays in $^{254}$Fm. The level was placed into the known scheme based on $\gamma-\gamma$ coincidences as well as energy differences. Table 3.9 shows the measured $\gamma$-rays and their relative intensities. The level scheme
Figure 3.28: Gamma-rays coincident with fermium k_{α1} or k_{α2} x-rays within 100 ns.

including these new transitions is shown in figure 3.29 and can be compared to the previously known level scheme in figure 3.8.

We interpret the level at 985.7 keV as an octupole vibration with a spin and parity of 2-. The assignment is based on relative transition intensities feeding into the 2-, 2+, and 3+ levels. By comparing total measured x-rays with a calculated conversion x-rays using conversion coefficients obtained using the code for Band-Raman Internal Conversion Coefficients (BrIcc) [KBT+08] it was possible to eliminate all but one of the spins and parities possible due to the relative level feeding. No other spin and parity assignment is consistent with the total number of observed x-rays.

In addition, this assignment is consistent with a 2- state observed in Fm-256 at 881.8 keV measured by Hall et al. [HGH+89]. A similar state should be expected in
Table 3.9: $^{254}$Fm $\gamma$-rays as a result of the electron capture decay of $^{254}$Md.

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>Counts</th>
<th>Relative Transition Intensity $\gamma^+e^-$</th>
<th>$J_{\pi i} \rightarrow J_{\pi f}$</th>
<th>Transition (initial $\rightarrow$ final)</th>
<th>Multipolarity</th>
</tr>
</thead>
<tbody>
<tr>
<td>251.9</td>
<td>17.6±4.2</td>
<td>9.88 ± 2.24</td>
<td>2$\rightarrow$ 3+</td>
<td>985.7 $\rightarrow$ 733.5</td>
<td>E1</td>
</tr>
<tr>
<td>292.0</td>
<td>46.2±6.8</td>
<td>29.47 ± 4.47</td>
<td>2$\rightarrow$ 2+</td>
<td>985.7 $\rightarrow$ 693.7</td>
<td>E1</td>
</tr>
<tr>
<td>544.3</td>
<td>3.1±1.8</td>
<td>3.67 ± 2.24</td>
<td>2$\rightarrow$ 4+</td>
<td>693.7 $\rightarrow$ 149.4</td>
<td>E2</td>
</tr>
<tr>
<td>584.2</td>
<td>5.5±2.4</td>
<td>7.07 ± 3.00</td>
<td>3$\rightarrow$ 4+</td>
<td>733.5 $\rightarrow$ 149.4</td>
<td>E2</td>
</tr>
<tr>
<td>648.7</td>
<td>56.7±7.5</td>
<td>80.86 ± 10.73</td>
<td>2$\rightarrow$ 2+</td>
<td>693.7 $\rightarrow$ 45.0</td>
<td>E2</td>
</tr>
<tr>
<td>688.5</td>
<td>28.9±5.4</td>
<td>43.96 ± 8.14</td>
<td>3$\rightarrow$ 2+</td>
<td>733.5 $\rightarrow$ 45.0</td>
<td>E2</td>
</tr>
<tr>
<td>693.7</td>
<td>65.2±8.1</td>
<td>100 ± 12.52</td>
<td>2$\rightarrow$ 0+</td>
<td>693.7 $\rightarrow$ 0.0</td>
<td>E2</td>
</tr>
<tr>
<td>940.7</td>
<td>11.2±3.4</td>
<td>23.84 ± 7.12</td>
<td>2$\rightarrow$ 2+</td>
<td>985.7 $\rightarrow$ 45.0</td>
<td>M1</td>
</tr>
</tbody>
</table>

Figure 3.29: Updated decay diagram of $^{254}$Fm following the electron capture decay of $^{254}$Md.

both of these nuclei.

Numerical calculations based on the Nilsson potential performed by Neergard and Vogel [NV70] predict a 2- octupole vibration at approximately 830 keV and Jolos et al. [JSS12] place it at 1061 keV. Both of these are in good agreement with our work. Figure 3.30 shows both calculated and experimental trends of the octupole vibration $\gamma$ band energy. Based on these new transitions, the level scheme shown in Figure 3.29 for $^{254}$Fm. Large conversion coefficients for rotational transitions prevented deter-
minimization of absolute $\gamma$-ray intensities. All transition energies and relative branching intensity measured in this work agree with previously measured values by Ahmad et al. [AL84].

### 3.7 Conclusion

In this work a previously unknown octupole vibration in fermium-254 has been identified. The new level has been compared to both theory and experimental results in neighboring nuclei showing the trend in octupole vibrations in neutron deficient actinides for the first time.

Transporting evaporation residues to a two clover low background detector setup can effectively be used to perform $\gamma$-spectroscopy measurements of nuclei that are not accessible by currently common methodologies. This technique provides an excellent addition to currently available tools such as in-beam spectroscopy and $\gamma$-ray tracking arrays. The high-intensity stable ion beams from the 88-inch cyclotron, together with the highly selective and efficient separation of compound nucleus-evaporation residue products using the BGS and then followed by rapid delivery to a low-background detector facility opens up many new possibilities for nuclear decay and structure studies in the neutron deficient actinides, such as identifying the Nilsson configurations of the ground state and low lying states by studying the $\gamma$-rays associated with electron capture decay, and determining the nuclear deformations by observation of
rotational transitions populated in electron capture decay.

### 3.8 Experimental Outlook

With the success of the $^{254}\text{No} \xrightarrow{EC} ^{254}\text{Md} \xrightarrow{EC} ^{254}\text{Fm}$ headline experiment, the way is paved for the study of the nuclear structure of many currently unknown neutron deficient actinides. Studying $^{252}\text{Fm}$ would be particularly interesting since there is currently no data available on its nuclear structure. Studying this nucleus would provide an additional data point for the systematics of octupole vibrations in fermium. However, this setup is not limited to the study of just fermium isotopes. Many other nuclei in this region are now accessible. $^{228}\text{Np}$ produced in the reaction $^{209}\text{Bi}(^{22}\text{Ne},3n)^{228}\text{Np}$, with an expected cross section of 3.7 $\mu$B, is only one of many examples that could be studied with this technique.

In preparation for future experiments the background rate should be reduced further. Reducing the background rate by an order of magnitude would greatly enhance the peak to background ratio in gated spectra. This would open up isotopes with lower production cross sections without prohibitively long beam times. One possibility to reduce background would be to install Compton veto detectors around the clover detectors. This would make it possible to both veto $\gamma$-rays that only deposited part of their energy in the detectors as well as vetoing external background. There are several professors in the nuclear engineering department specializing in low background counting with whom collaborations would be a possibility.

Additionally, it would be prudent to develop a method to measure the highly converted, low-lying, rotational transitions in these neutron deficient actinides. One possibility would be to develop an apparatus in which the gas jet deposits on a thin wheel which runs between two Passivated Implanted Planar Silicon (PIPS) detectors for conversion electron detection. This apparatus could then be surrounded by clover detectors to measure x-ray, $\gamma$-ray, and/or electron coincidences.
Bibliography


[MTMM71] L. Multhauf, K. Tirsell, R. Morrow, and R. Meyer. Levels of $^{246}$Cm from the $\beta^-$-Decay Sequence $^{246}$Pu(11 days)$^{246m}$Am(25 min)$^{246}$Cm. Physical Review C, 3(3):1338–1351, March 1971.


AutoCad Drawings for the Electro-deposition Cell

This appendix contains the AutoCad drawings for the electro-deposition so that they are available for any future cells that may need to be produced.