Preparation of Actinide Targets by Molecular Plating for Coulomb Excitation Studies at ATLAS

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Abstract

Molecular plating is now routinely used to prepare sources and targets of actinide elements. Although the technique is simple and fairly reproducible, because of the radioactive nature of the target it is very useful to record various parameters in the preparation of such targets. At Argonne, $\sim 200 \mu\text{g}/\text{cm}^2$ thick targets of Pu and Cm were required for Coulomb Excitation (COULEX) Studies with the Argonne-Notre Dame BGO gamma ray facility and later with the GAMMASPHERE. These targets were plated on $50 \text{ mg}/\text{cm}^2$ Au backing and were covered with $150 \mu\text{g}/\text{cm}^2$ Au foil. Targets of ^{239}Pu , ^{240}Pu , ^{242}Pu , ^{244}Pu and ^{248}Cm were prepared by dissolving the material in isopropyl alcohol and electroplating the actinide ions by applying 600 volts. The amount of these materials on the target was determined by alpha particle counting and gamma ray counting. Details of the molecular plating and counting will be discussed.

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1. Introduction

In order to pursue a systematic study of collective behavior in actinide nuclei by the method of Coulomb excitation [1], the high quality heavy-ion beams available at ATLAS as well as high purity isotopic actinide targets are required. A large array of germanium detectors for gamma-ray spectroscopy such as GAMMASPHERE presents the necessary experimental opportunity to accomplish this effort. The Physics Division of Argonne National Laboratory finds itself in the unique position of having the ability to obtain, process and handle actinide elements and, therefore, prepare the thin deposits used as targets for these experiments.

In the technique of so-called "unsafe" coulex, beams of ~ 6 MeV/u ^{208}Pb , a few percent above the Coulomb barrier, impinge on a thin actinide target deposited on a thick backing of Pb or Au. This allows for both the beam and recoil to be stopped in the thick backing layer. The advantage of this method over "traditional" coulex where the recoil escapes out of the target and measured in a particle detector is that now the gamma rays are emitted from the stopped nuclei and can be detected at high resolution using the array. The first studies [2] employed targets of ^{248}Cm and ^{240}Pu and using the Argonne-Notre Dame array comprised of 50 BGO elements and 12 germanium detectors. With the availability of GAMMASPHERE at ATLAS [3], these studies have now expanded to include ^{239}Pu , ^{240}Pu , ^{242}Pu and ^{244}Pu .

2. Purity of Isotopic Starting Materials

The starting material for the molecular plating of thin deposits onto the backing substrates needed to be of high elemental and isotopic purity, and therefore well characterized. The actinide materials, which are available to U.S. national laboratories from the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory, were obtained from the ANL Chemistry Division. For each radioisotope, the isotopic purity was greater than 97%. These materials were chemically pure and in most instances, were used directly for molecular plating. In the case of ^{239}Pu and ^{240}Pu , the plutonium was chemically purified by an anion-exchange column using nitric acid solution.

The gamma ray spectrum obtained using a small, low-energy photon spectrometer (LEPS) of the ^{240}Pu target material before chemical purification is shown in Figure 1a. Evident are the ^{241}Am gamma rays which are due to the ^{241}Am accumulated from the decay of a small amount of ^{241}Pu in the sample. In Figure 1b a spectrum is given of the ^{240}Pu material after chemical purification which shows a very small amount of ^{241}Am and less background under the peak.

3. Target Preparation

The preparation of actinide targets by electrodeposition for use in heavy-ion physics experiments is well covered in the literature, beginning with the work of Trautman and Folger [4], and more recently, on the excellent techniques developed at the Institute for Reference materials and Measurement (IRMM) by Ingelbrecht [5]. The method is inherently simple, involving a Teflon electroplating cell and using a platinum wire as the anode with the substrate resting on a brass base acting as the cathode. A potential is applied to the Pt electrode and the resulting current produces a migration of the dissolved actinide ion toward the cathode where it accumulates and deposits onto the substrate. Beginning with only small amounts of starting material, high efficiency (>90%) is obtained using this method.

Based on earlier experience with electroplating of ^{248}Cm [6] for experiments at the Indiana University Cyclotron Facility [7] on thin backings, the deposition of the plutonium isotopes proceeded in a similar fashion. We employed a plating cell based on the design first put forth by Sedlacek [8], with modifications. A schematic of our cell design is shown in Figure 2. In the place of the thin carbon film on a microscope slide used previously, a brass plate was substituted as a fixture for holding the gold backing foil already mounted on the target frame to be used in the experiment. This is a prudent adaptation, as it eliminates the need for any further handling or preparation of the target. The anode consists of a 1.5 mm diameter Pt wire approximately 18 cm in length. The end is fashioned into a circular shape of about 5 mm in diameter. To obtain a more uniform deposit, the electrode is rotated often during the molecular plating process.

The starting material was obtained in nitric acid solution. The solution was evaporated to dryness, the residue dissolved in 2 ml of isopropyl alcohol and then transferred to the cell using a plastic pipette for ease of disposal. Approximately 2 ml is needed to fill the cell. The Pt wire electrode is then immersed into the liquid, maintaining a distance of 6 mm above the gold substrate. Following the method set forth by Aumann and Mullen [9], a potential of 600 V dc is applied across the apparatus and the plating commenced. Duration of the plating was 90-120 minutes. Plating efficiencies of >90% have been achieved. A photograph of a target of ^{239}Pu is given in Figure 3. Because of the heavy-ion beam encountered in the experiment, sputtering of the target material is expected to occur to some degree. In order to avoid this possible spread of contamination, a thin gold foil is placed in front of the target. Indeed, a small amount of material has been observed to collect on this foil layer.

Targets of ^{239}Pu , ^{240}Pu , ^{242}Pu , ^{244}Pu and ^{248}Cm have been produced by this method and used in the Coulex experiments. Two or three targets of each isotope were prepared. In most cases, only one target was used for the 4 day run with ~ 0.3 pA of ^{208}Pb beam.

The target thickness ranged from 200 to 300 $\mu\text{g}/\text{cm}^2$ and the area of each deposit was 0.28 cm^2 , which was determined by the diameter of the plating cell. Table 1 gives a summary of targets produced.

4. Assay and Characterization

Although the isotopic composition of the actinide materials used in the present experiment were available from Oak Ridge, the materials were further characterized by measuring their alpha spectra. The amount of actinide in the target was determined in two ways. In the first method, the amount of material placed in the molecular plating cell was determined by the alpha disintegration rate of a small aliquot of the solution. After the plating was completed, the amount of material left in the solution was similarly measured. The difference in these measurements yields the amount of material in the target.

The second method utilized alpha and gamma-ray counting. For ^{242}Pu and ^{244}Pu which has low specific activity, alpha counting was used. For ^{239}Pu and ^{240}Pu , the gamma ray spectra were measured using LEPS counting system mentioned above.

5. Results and Discussion

For the experiment carried out at the BGO, the ^{248}Cm target performed well in the 0.1-0.3 pA $\sim 6 \text{ MeV/u}$ ^{208}Pb beam from ATLAS. Three rotational bands were identified with the yrast band observed to a spin of 28 $\hbar$. A second band was observed in coincidence with low-spin transitions of the ground state. The two lowest-lying transitions in the third band correspond to the ground state band of ^{246}Cm and was observed to 20 $\hbar$. This isotope was present at the 3% level, based on the activity of the target. The ^{240}Pu targets used in this same experiment performed in a similar fashion. The ground state band in this nucleus has been extended by 12 $\hbar$ to the 26+ state. Ten new states in a side band were also identified.

In experiments conducted using GAMMASPHERE, the ^{242}Pu and ^{244}Pu performed well in 0.3-0.5 pA beams of $\sim 6 \text{ MeV/u}$ ^{208}Pb .

Measurements on the previously used ^{240}Pu targets now using GAMMASPHERE confirm the results obtained earlier. Investigations involving the use of ^{239}Pu targets at GAMMASPHERE is currently underway.

6. Conclusion

Molecular plating is now routinely used to prepare targets of actinide elements required for Coulomb excitation (COULEX) studies at ATLAS with GAMMASPHERE. The technique is simple and fairly reproducible for the preparation of such targets. Targets of ^{239}Pu , ^{240}Pu , ^{242}Pu , ^{244}Pu and ^{248}Cm were prepared by this method. The targets were plated on 50 mg/cm^2 Au backing and were covered with $150\text{ }\mu\text{g/cm}^2$ Au foil in order to contain any material sputtered by the heavy-ion beam. The amount of material contained within the target was determined by standard counting methods. Further research involving Coulomb excitation of these nuclei is currently in progress.

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Table 1. Actinide Targets Prepared by Molecular Plating

Isotope	Thickness ($\mu\text{g}/\text{cm}^2$)	Backing Foil
^{248}Cm	203	50 mg/cm^2 Pb (with a Au "flash")
	210	53 mg/cm^2 Au
	180	50 mg/cm^2 Pb
^{239}Pu	250	53 mg/cm^2 Au
	150	48 mg/cm^2 Au
		48 mg/cm^2 Au
^{240}Pu	150	53 mg/cm^2 Au
	350	63 mg/cm^2 Pb
	310	53 mg/cm^2 Au
	180	53 mg/cm^2 Au
^{242}Pu	240	53 mg/cm^2 Au
	220	53 mg/cm^2 Au
	330	53 mg/cm^2 Au
^{244}Pu	260	53 mg/cm^2 Au
	175	53 mg/cm^2 Au

Figure Captions

- Figure 1a. LEPS spectrum of the ^{240}Pu target material before chemical purification, ^{241}Am gamma rays are due to the ^{241}Am accumulated from the decay of a small amount of ^{241}Pu in the sample.
- Figure 1b. Spectrum of the ^{240}Pu material after chemical purification which shows a very small amount of ^{241}Am and less background under the peak.
- Figure 2. Schematic of the Electrochemical Cell Based on the Original Design of Sedlacek [8] with Modifications
- Figure 3. Photograph of a ^{239}Pu Target Prepared by Molecular Plating