Method Development for Thin Radioarsenic Targets

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Abstract. Experimental neutron-capture cross-sections of arsenic radioisotopes, especially arsenic-73 (^{73}As) and arsenic-74 (^{74}As) have not previously been measured. However, these cross-section measurements are important to obtain for applications such as stockpile stewardship and astrophysics. Although many methods for developing thin-film arsenic targets are available in literature, they are not directly applicable to the fabrication of radioarsenic thin-film targets, where a carrier is needed. Gold can be used as a substrate backing for neutron-capture cross-section measurements and arsenic has been shown to co-deposit with gold in hydrochloric acid. Therefore, this study investigations the feasibility of using gold as a carrier to develop radioarsenic targets on a gold substrate backing.

1 Introduction

Accurate cross-section measurements are important to a wide variety of fields of research, including astrophysics, reactor performance, nuclear medicine, and defense applications. Current data for neutron-induced cross-section measurements exist for most stable isotopes; however, very few measurements on radioisotopes have been performed. Although statistical model predictions exist for many radioisotopes, the experimental values may be vastly different and uncertainties associated with these predictions are typically high [1]. As a result, direct and indirect neutron-induced cross-section measurements are desired to update current nuclear databases.

Arsenic is one element of interest for obtaining neutron-induced cross-sections measurements from [2]. During underground nuclear tests, the stable isotope arsenic-75 (^{75}As) was used as a detector material for neutron fluence monitoring. The ratio of radioarsenic isotopes arsenic-73 (^{73}As, $t_{1/2}$ = 80.3 d [3]) and arsenic-74 (^{74}As, $t_{1/2}$ = 17.8 d [4]) allows for the calculation of the total neutron activity via the (n,2n) or (n,γ) reactions, which can be used to evaluate testing performance [5]. In astrophysics, arsenic is relevant to the *r*- and *s*-processes of nucleosynthesis [5]. For both of these applications, a method to produce thin films of radioarsenic is necessary to perform direct neutron-induced cross-section measurements on. Methods for developing thin-film arsenic targets can be found in the literature [5, 6], but these methods are not directly applicable when manufacturing radioarsenic thin-film targets, where a carrier is not naturally present. Because gold is monoisotopic, can be used as a substrate backing for neutron-capture cross-section measurements [7], has well established neutron-capture cross-sections [8], and arsenic has been shown to co-deposit with gold in hydrochloric acid [9], investigations into whether it is feasible to use gold as a carrier to develop thin film radioarsenic targets on a gold substrate backing were conducted.

2 Materials and Methods

For each electroplating experiment, a 20 mL 0.1M HCl (Fisher Chemical, TraceMetal) solution containing either 50 or 100 ppm of Au (Inorganic Ventures, 10% v/v HCl, 1000 μg/mL) and a small spike of ^{73}As (National Isotope Development Center in 0.1N HCl) was used. Gold discs (Goodfellow, 99.95% metals basis, 0.025 mm thick, 25.4 mm diameter) were used as a cathode. A platinum anode (BeanTown Chemical, 99.99% metals basis, 0.025 mm thick) was used. A custom PEEK plating cell with a 7 mm diameter opening and a maximum volume capacity of 2.5 mL was utilized for all experiments. A plating volume of 2 mL was utilized for all experiments, with the plating time kept consistent at 1 hour. A small stir bar within the plating cell ensured that the plating solution remained homogeneous.

The gold deposition efficiency was determined by inductively coupled plasma - optical emission spectroscopy (ICP-OES). For each gold concentration (50 ppm or 100 ppm), the stock solution was made to be used for ICP-OES measurements prior to and post-deposition. The arsenic deposition efficiency was determined by liquid scintillation counting (LSC), where a 500 μL aliquot was removed prior to deposition and homogenized with a liquid scintillation cocktail (Ultima-Gold LLT). Post-deposition, a 500 μL aliquot was removed and homogenized with Ultima-Gold LLT. The concentrations were compared prior to and post-deposition (either in ppm or cps) to evaluate the deposition efficiency.

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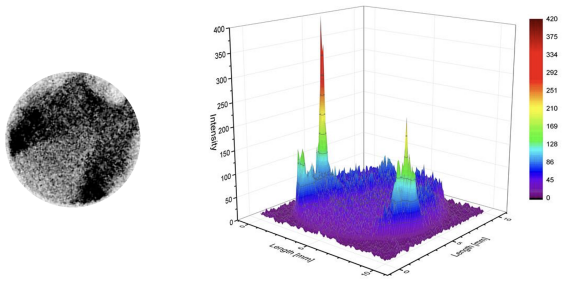

Figure 1: Radiograph of the electroplated ^{73}As target (right), with a stock solution containing 50 ppm of Au, applied voltage of 1 V and deposition time of 1 hour. There was an approximate deposition efficiency of 12% for ^{73}As and 44% for Au. Pixel intensity map of the radiograph (left) to indicate ^{73}As uniformity.

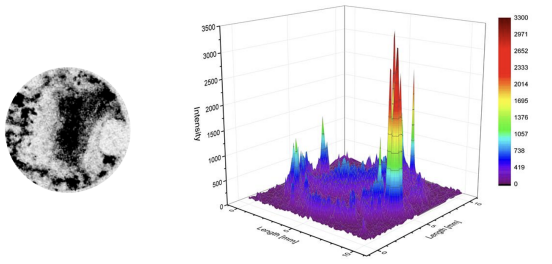

Figure 2: Radiograph of the electroplated ^{73}As target (right), with a stock solution containing 100 ppm of Au, applied voltage of 1 V and deposition time of 1 hour. There was an approximate deposition efficiency of 52% for ^{73}As and 42% for Au. Pixel intensity map of the radiograph (left) to indicate ^{73}As uniformity.

3 Results and Discussion

Upon visual inspection, the electroplated targets did not appear very uniform (see Figure 1 and Figure 2 for radiographs and pixel intensity maps of the targets produced with an applied voltage of 1 V). It appears that that increasing the concentration of the gold solution yielded greater ^{73}As deposition but a less uniform deposition, as indicated by the heat map. Increasing the applied voltage significantly decreased the deposition efficiency of both gold and ^{73}As, likely due to the formation of H_2 bubbles that inhibit access to the target backing substrate.

The deposition efficiencies for different plating conditions are described in Table 1. Based on these experiments, it is predicted that gold stock solutions above 100 ppm and depositions occurring for longer than an hour could have better deposition yields. Improvements in uniformity could be provided by lowering the applied voltage to less than 1 V.

In order for this method to be suitable for nuclear data needs, the production of a uniform target with higher deposition of ^{73}As is essential. Further investigation into the starting solution with respect to the carrier used, carrier concentration, and chemical composition, could lead to a more uniform deposition. Moreover, optimization of this method by varying the applied voltage, deposition time, and substrate backing material, could yield more uniform targets with higher deposition efficiencies. Target characterization to determine the speciation of the gold and arsenic, layer thickness, contaminants, and durability during irradiation could also be explored.

Table 1: Deposition efficiency of ^{73}As and stable Au, under various conditions: specifically, the initial gold concentration of the plating solution (Au stock) and the applied voltage (either 1 V or 2 V). For all electroplating experiments, the deposition time was approximately 1 hour, and the same cell geometry was used.

Au [ppm]	Voltage [V]	^{73}As [%]	Au [%]
50	1	12	44
	2	0.6	23
100	1	52	42
	2	3	24

4 Conclusions

Using stable gold as a carrier is a potential method that is suitable for making thin-film, radioarsenic targets. Such targets could be used for neutron-induced cross-section measurements, where updated data are needed. It appears that higher concentrations of gold, combined with low voltages (around 1 V) and deposition times exceeding one hour would yield the best deposition efficiencies. For an initial gold stock concentration of 100 ppm and an applied voltage of 1 V, a deposition efficiency of above 52% for ^{73}As and 42% for gold onto a gold substrate backing was observed.

Further development and optimization of this method to fabricate thin radioarsenic targets could be beneficial to update current nuclear databases, where experimental data are desired. The desired neutron-induced cross-section measurements could be conducted at facilities such Los Alamos Neutron Science Center (LANSCE), where detector arrays like the Low Energy NZ-neutron induced charged particle detection (LENZ) require a thin film target geometry.

This work is supported by: The U.S. Department of Energy, Office of Science, Office of Nuclear Physics and used the resources of the Facility for Rare Isotope Beams (FRIB), which is a DOE Office of Science User Facility, operated by Michigan State University, under Award No. DE-SC000661; U.S. Department of Energy Isotope Program, managed under the Office of Isotope R&D and Production under Award No. DE-SC0021220; National Nuclear Security Science Academic Alliances Program (SSAA) under Award No. DE-NA0003913; Department of Energy National Nuclear Security Administration

through the Nuclear Science and Security Consortium under Award No. DE-NA0003996.

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