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2017 Report for New LANL Physical Vapor Deposition Capability

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There is an urgent need at LANL to achieve uniform, thin film actinide targets that are essential for nuclear physics experiments. The target preparation work is currently performed externally by Professor Walter Loveland at Oregon State University, who has made various evaporated actinide targets such as Th and U for use on several nuclear physics measurements at LANSCE. We are developing a vapor deposition capability, with the goal of evaporating Th and U in the Actinide Research Facility (ARF) at TA-48. In the future we plan to expand this work to evaporating transuranic elements, such as Pu. The ARF is the optimal location for evaporating actinides because this lab is specifically dedicated to actinide research. There are numerous instruments in the ARF that can be used to provide detailed characterization of the evaporated thin films such as: Table top Scanning Electron Microscope, In-situ X-Ray Diffraction, and 3D Raman spectroscopy. These techniques have the ability to determine the uniformity, surface characterization, and composition of the deposits.

Instrument Details and Setup

A Denton Vacuum 502B (DV-502B) system has been installed and implemented in the ARF, shown in Figure 1. This system is capable of reaching a vacuum of 10^{-8} torr, has a 2kVA evaporation power supply upgrade option, high purity gas lines, deposition controller, and an oscillating quartz crystal monitor. The vapor pressure of deposition material increases dramatically with the temperature; for example, the vapor pressure of Au increases from 10^{-8} torr to 10^{-6} torr when the temperature increases from 677^{o}C to 821^{o}C. The 2kVA evaporation power supply allows for the filaments to reach higher temperatures enabling deposition of a larger variety of compounds.

The evaporation system can be controlled in two ways: 1) a touch screen instrument controller, and 2) the automatic deposition controller (Inficon SQC-310 Controller), both shown in Figure 1. The touch screen controller allows for the manual control of the pressure, current, shutter position, and rotation rate. This mode of operation can be used to determine the initial deposition parameters for each new material. The automatic deposition controller has two ways to control the evaporation process: in a manual power ramp mode or an automatic mode. The manual power ramp mode allows the user to utilize the oscillating quartz crystal while manually controlling the power. In the automatic mode, the evaporator runs predefined deposition protocols. These protocols have many parameters which control the deposition process such as: the ramp rate, soak, idle and set point times, percent power, endpoint thickness, and deposition rate. The automatic deposition controller can store up to 1000 programmable protocols for fully automatic deposition and greatly decreases the man hours needed to produce replicates.

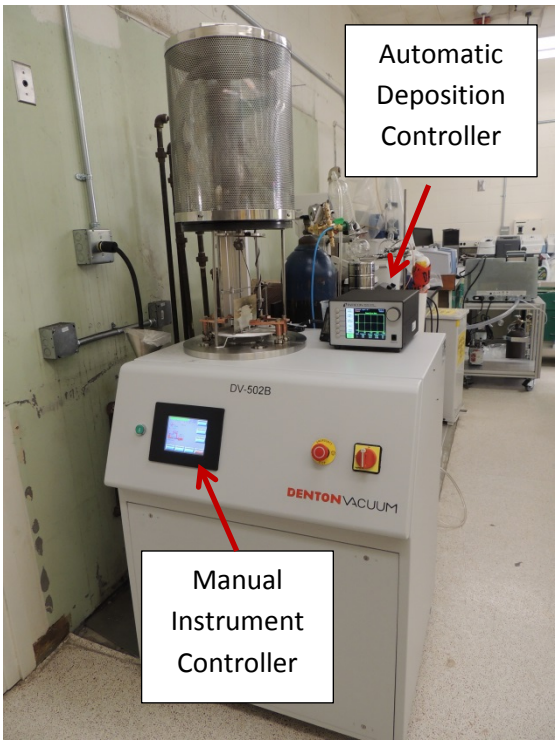

Figure 1. Denton Vacuum 502B Evaporator capability at TA-48 Actinide Research Facility (ARF)

During operation, the evaporator continually measures the internal pressure of the system and displays it on the touch screen. A pressure interlock is also in place to shut down the instrument if the pressure seal is compromised. High purity gas lines are preferred for venting the ball jar with nitrogen or argon, which can aid in keeping the deposit in the same chemical composition, even after evaporation. For example, a premature venting with air after evaporation of ThF_4 introduces oxygen into the system and would change the deposit to a mixture of thorium fluoride and oxides. Therefore, venting with inert N_2 gas will help control the chemical composition of the deposits.

The oscillating quartz crystal, seen in Figure 2, which monitors the deposition rate is used by the controller to measure the thickness of a deposit and regulate the deposition rate in automatic mode. The controller when runs in automatic mode, the controller applies the power to maintain a set deposition rate and automatically terminate the deposition once a set thickness is reached (based on the oscillating quartz crystal), therefore producing consistent targets.

The rotating plate and shutter will also aid in controlling the quality of the deposition. The rotation of the substrate will increase the uniformity of a deposit, especially on a non-uniform surface such as carbon foil[1]. The rotating plate is 6 inches in diameter and allows for mounting multiple samples at a time. However, the accuracy of the thickness deposited may change depending on the position on the rotating plate. The shutter can be used to cover the filament until the desired deposition rate is achieved. Controlling the deposition rate is important for uniformity and thickness control.

The two electrodes and shadow bar give the ability to make samples with two layers in one deposition protocol. The shadow bar ensures that there is no cross contamination between the two electrodes. An example of its application would be to evaporate UF_4 and then cover it with a stable metal such as Al or Au. Covering a radioactive deposit with a stable metal would allow for easier handling of the sample.

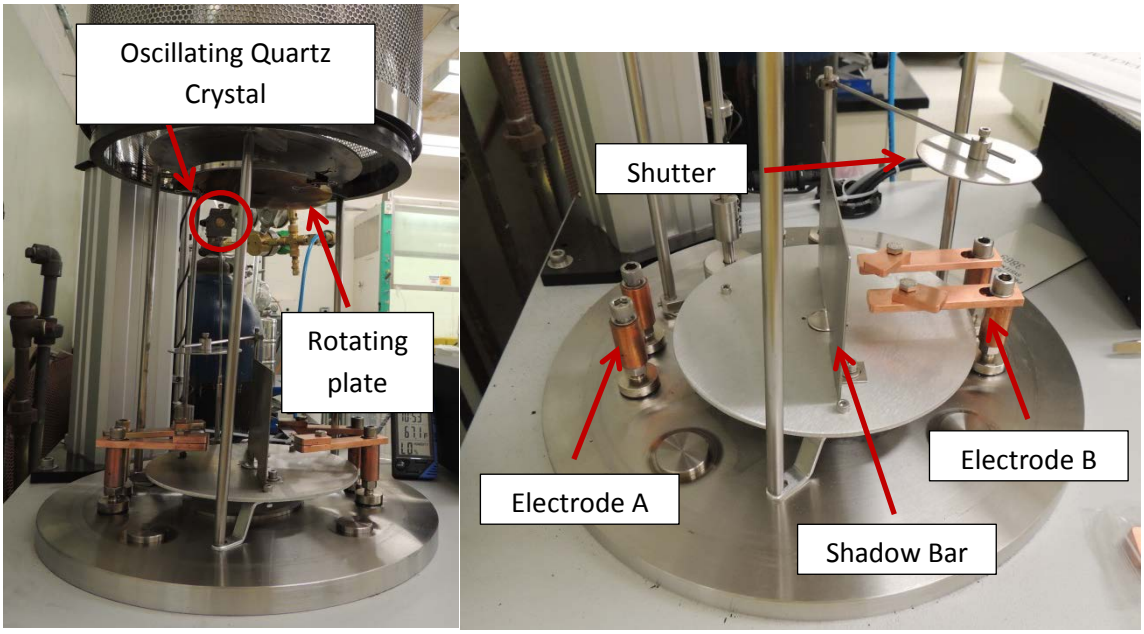

Figure 2. Bell Jar Internal Components

Once the evaporation system is thoroughly optimized, it will be able to provide thin and uniform deposits of actinide compounds on various backing materials such as Al, C, Ti or polymeric materials. Due to safety restrictions and our current setup, we will most likely only be able to evaporate Th and U in the near term. However, with additional funding we would be able to alleviate some of the safety restrictions and evaporate other actinides such as Cf and Pu.

An Integrated Work Document (IWD) was written and approved for evaporating thin films. The IWD was approved after working with Industrial Health and Safety, Radiation Protection, Pressure and Electrical Safety, and Facilities. Under this IWD, we will be able to work with no more than 1 g of solid or

slurries of low-level radioactive elements such as Th and U; there is no mass limitation on stable compounds. This will provide us with the ability to coat radioactive samples with stable materials, such as Al, Au, and Cu. By coating the sample with a layer of stable material the sample handling much easier, as it will then can be considered as a sealed source. This option would only be appropriate if the coating does not alter the physics requirements of the source.

Experimental Work

A manual has been written on how to use the evaporator. For the most part, this instrument is fairly easy to use safely due to the pressure, voltage, and temperature interlocks. There are also auto processes available for venting, pumping, and depositing. We also used the Inficon SQC-310 controller for automatic film deposition of desired thickness. The process needs to be optimized for each element and setup as a protocol for the controller, once this is done the user will be able to reuse the same protocol to achieve a well-defined thickness each time. Further work is needed to understand the calibration of the quartz crystal that determines the thickness.

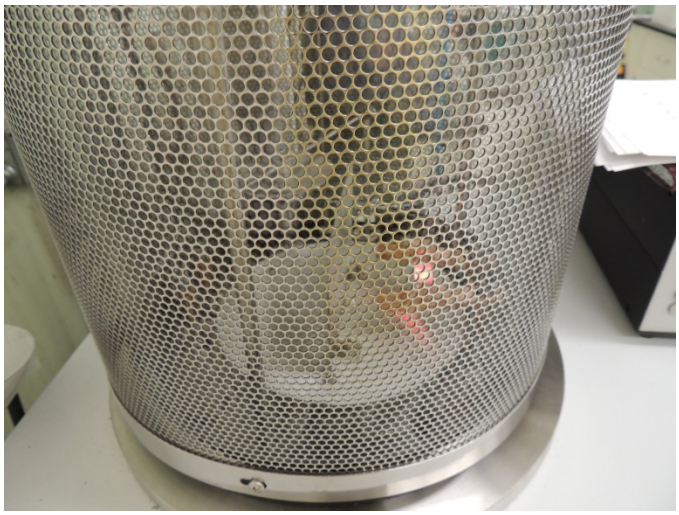

Figure 3. Bell Jar During Gold Deposition using Tungsten filament

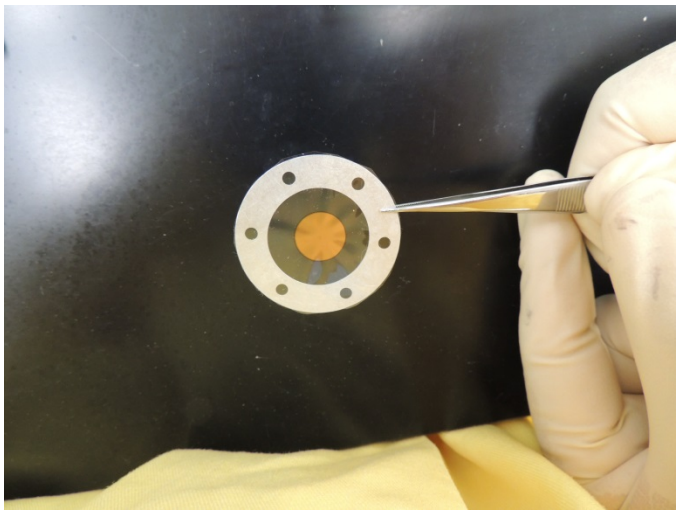

Figure 4. Gold Deposited on Mylar

A manual deposition of gold on thin Mylar, 300 μg/cm^2 thick, placed in a DANCE ring was achieved (Figure 4). Mylar was chosen since its melting point is 260^{o}C, which is low compared to the melting points of the metals that we planned to deposit. Since the Mylar showed no signs of degradation, we assume any substrates with a melting point above 260^{o}C will be compatible this evaporation deposition procedure. This deposit was made by utilizing an aluminum mask with an inner diameter of a 0.5 inch and an outer diameter of 6 inches, which matches the rotating plate diameter. The mask and ring was held on to the rotating plate by binder clips. The rotation speed was set at 80%. This evaporation used 77 mg of gold foil in a tungsten boat under a vacuum of approximately 10^{-7} Torr, utilizing 28% power for 2 minutes. As this was our first attempt, the mass of gold evaporated on the Mylar target was not measureable on a balance. The evaporation protocol, including ramp time, was 15 minutes. From the beginning to end, the process took approximately two hours; the majority of which was spent pumping and venting the system, which required 30-45 minutes each.

Utilizing the deposition controller, we deposited 13.2 nm of copper onto a thin carbon foil, as shown in Figure 5. Before preparing this deposit, we ran multiple experiments to determine the optimal running conditions for depositing copper at a rate of 1 Å/s. Maximum power was set at 25%, with rotation at 100%. Although deposition for copper tends to start closer to a power of 16% it is necessary to set the maximum power higher so that the controller had room to optimize the power until the desired rate was reached. The deposition rate over time is displayed on the controller and is shown in Figure 6. It took approximately 2 minutes to reach the desired deposition rate. With more working experience, we will be able to run much more complex protocols to meet a variety of target specifications.

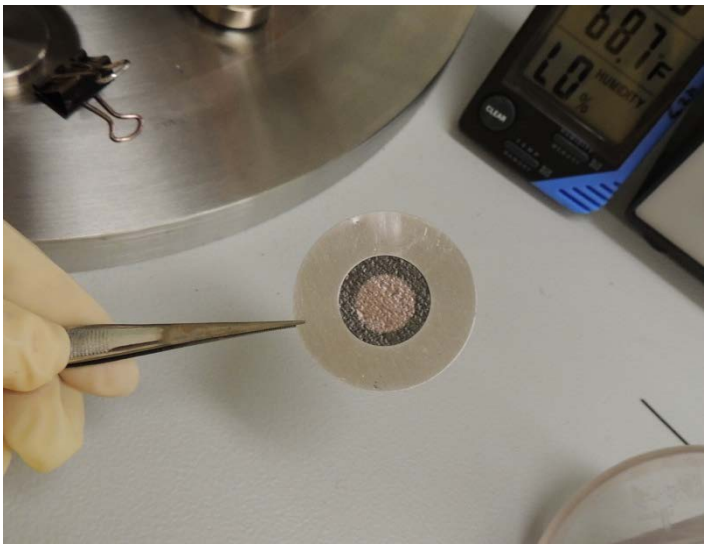

Figure 5. 13.2nm (132 Å) Copper Deposit on Thin Carbon Foil

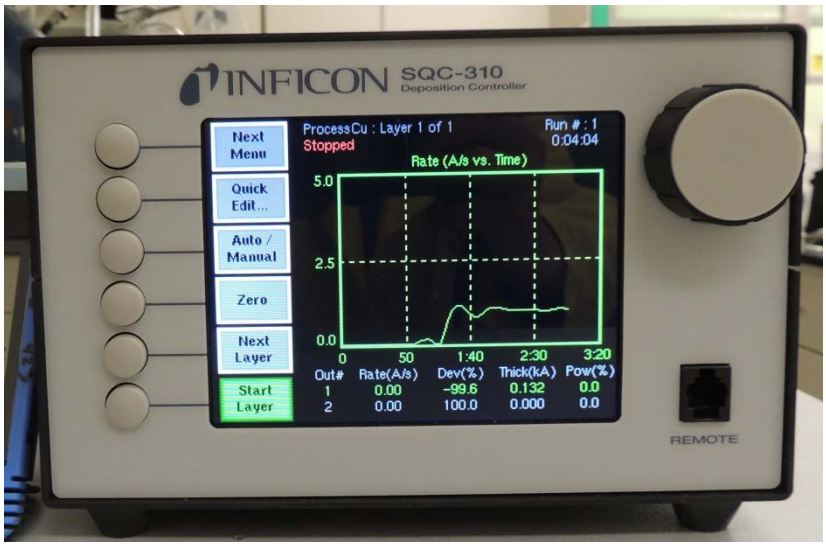

Figure 6. Rate of Copper Deposited on Thin Carbon Foil over Time

Future Work

In order to take advantage of real time deposition rate measured by the deposition controller, we need to fully optimize the automatic protocols for different sample material. The manual is lacking in some detail on this. Therefore, we will systematically go through all of the various controls and protocol parameters for different materials. Once the system has been sufficiently studied, a how-to document will be written for future users.

Optimal targetry for nuclear physics measurements requires a well-defined geometry, e.g. deposit size and shape. To explore this, we will make and test various mask designs. Once an optimal design is found, we will be able to make a large variety of sized masks to shorten the waiting time for target deposition. We will optimize the deposition parameters for the source material that is most likely to be used.

We plan to calibrate the film thickness using laser interferometry. We have a laser interferometer setup, approved and working. To measure the film thickness, we can deposit the thin film on a small area of a highly reflective mirror and use this modified mirror to replace the high reflector of the interferometer. The thin film deposit creates an additional set of weaker optical interference fringes shifted from the original set. The frequency shift between the two sets of optical fringes gives a precise measurement of the film thickness.

We will start with optimizing Al or Au and then move onto ThF_4 or UF_4. The deposition of actinides will most likely take a little longer as it will be coordinated closely with the Radiation Protection office. The main challenges are to reduce the contamination risk and optimize the evaporation parameters which are not published. We plan to use a quartz glass tube chimney between the source and the target to contain the radioactive material inside the tube to aid in contamination control and material recovery or disposal. Once completed we will have a specific procedure written for evaporating ThF_4 and/or UF_4 using the evaporation deposition system. Ideally, we would like to take this technology and apply our knowledge to transuranic actinides, most specifically Pu. To evaporate Pu it will require a dedicated system housed in a hood or glovebox. There are a few tabletop versions of this system, the smallest of these available would take up an entire hood but has similar capabilities as the full sized version currently being used.

Another consideration for future work is the fact that ThF_4 and UF_4 are not commercially available. Also these compounds oxidize over time, for example ThF_4 will oxidize in air. The degradation of these compounds would hinder the quality of the optimized protocols. Therefore, we will need to develop the capability to synthesize ThF_4 and UF_4 as needed. An aqueous process has been well characterized for UF_4 and ThF_4 production, however anhydrous UF_4 and ThF_4 have not been formed [2,3,4,5,6]. Upon an initial literature search, it is not clear if an anhydrous form would affect an evaporation yield; therefore, this would need to be investigated further. If a completely anhydrous form is necessary, it is most likely that we will need to synthesize UF_4 and ThF_4 using HF gas at elevated temperatures. To use this method, a new IWD would have to be developed, which will be a considerable task considering the safety requirements associated with this type of work.

For characterization of the Th and U deposits, we will utilize the instrumentation available in the ARF. For example, we will use the Table Top SEM and 3D Raman, which will give chemical composition, surface structure, uniformity, and pore size characterization. Additional characterization instruments available are autoradiography for uniformity, XRF for chemical composition, gamma and alpha spectroscopy for yield assessment. However, some effort may be required to properly calibrate these instruments before they can provide quantitative results.

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