A novel technique for the production of robust actinide targets

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The success of accelerator experiments is highly influenced by the availability of targets with specific and well-defined properties. Actinide targets in particular are in high demand due to the importance they hold for stockpile stewardship as well as nuclear science. The main goal of this project is the development of revolutionary new approaches in the preparation of actinide targets that are isotopically pure, cost efficient, reliable, robust, and highly uniform with controlled thicknesses and dimensions (Stewardship Science Academic Alliances Program Topic Research Area # 3: Radiochemistry). The actinides will be provided by the Actinide Center of Excellence in Research in the Engineering College at the University of Notre Dame.Leading up to the experiment, I. Richardson participated in the $^{54}Fe+^{58}Ni$ beam production experiment with the Momentum Acromat Recoil Separator (MARS) [1]. For that experiment, a $^{54}Fe^{18+}$ beam at 36 MeV/u from the K500 cyclotron impinged on a natural nickel target 50 μm thick. Analysis of this data for production rates is ongoing.

The method used to produce these targets is electrospray deposition of chemically reactive layers that can be converted to actinide oxides by simple heat treatments. The experimental setup used consists of the following components as shown in Fig. 1:

Fig. 1. The Electrospraying setup. The components are marked as follow.

a. Ozone cleaner, for the preparation of substrates before the spraying.
b. Syringe pump – Syringe – Capillary nozzle.
c. Distance regulator between the tip of the nozzle and the substrate.
d. Copper base,
e. Hot plate,
f. High voltage power supply,
g. furnace

The first step was investigating the different combinations of parameters that will produce the best possible targets with respect of uniformity, thickness, and robustness (see Figs. 2 and 3). The surface treatment of both the aluminum and carbon substrates prior to spraying is where the investigation started, by determining the best treatment in order to have a thin target layer and not just droplets on the surface. Following that, the investigation turned to the parameters of the spraying itself. More specifically, the spraying time (from 15min to 2hr), the Uranium concentration (from 0.1 M to 1M) in the solution, the flow rate (from 3μl/h to 30μl/h) , the heat treatment temperature (from 350oC to 550oC) as well as the stability and robustness of the targets under irradiation with an Argon beam (from 2×10^{16} to 1.3×10^{17} ions/cm^2).

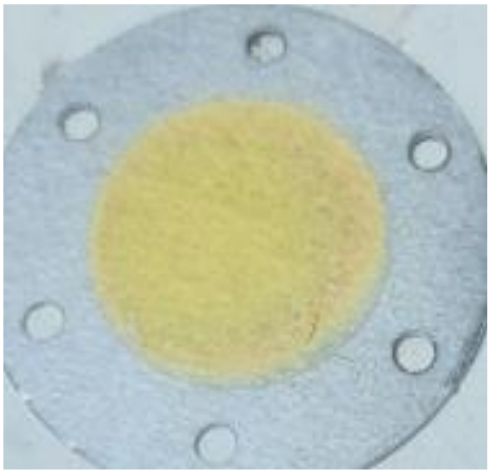

Fig. 2. UO_2 target on pure Al substrate.

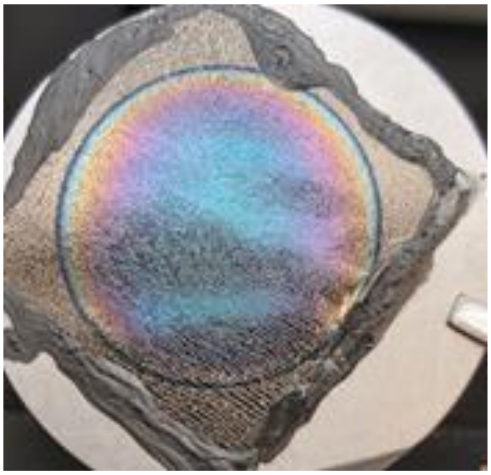

Fig. 3. UO_2 target on thin Carbon substrate.

In order to fully characterize these targets, alpha spectroscopy (thickness), X-Ray Fluorescence (XRF) analysis (uniformity), Transmission Electron Microscopy (TEM) analysis (crystal structure), Scanning Electron Microscopy (SEM) analysis (surface analysis) and X-Ray Photoelectron (XPS) analysis.(oxygen coefficient $k_0 = 2 + x$ and ionic (U^{4+}, U^{5+}, U^{6+}) composition of the UO_{2+x} layer) were used. Some of the results are presented below:

From the figures below as well as all the measurements we performed we can draw certain conclusions regarding our targets.

- The surface of our targets, (Fig 4) is smooth with minimum imperfections.
- The heat treatment temperature in-fluences the crystal structure of our deposited layer. Higher temperatures result in Mg leaking from the substrate into our UO_2 layer, thus obstructing the

crystallization of our layer and forming a MgO layer (Fig 5) inside our UO_2 layer. Otherwise, we have good crystallization of our target layer (Fig 6).

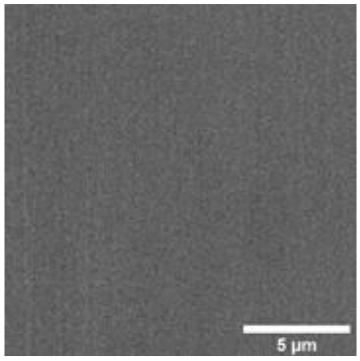

Fig. 4. SEM image of the surface after 350° C heat treatment and no irradiation.

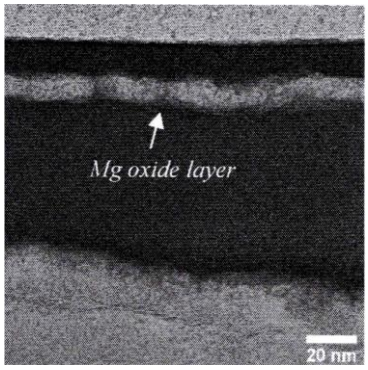

Fig. 5. TEM image of a target after heat treatment at 550° and irradiation with 7.7×10^{16} ions/cm^2

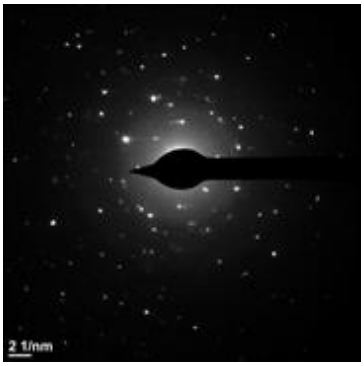

Fig. 6. Diffraction pattern of the UO_2 layer after heat treatment at 350ºC and irradiation with 2×10^{16} ions/cm^2.

- The amount of U in our target linearly depends on the duration of the spraying (Fig 7), which allows us to control the thickness of the target by varying the spraying duration.
- Higher fluence results in higher non uniformity of our targets, (Fig 8) but still less than 10%.
- The overall thickness of the targets does not change after the irradiation (Fig 9).

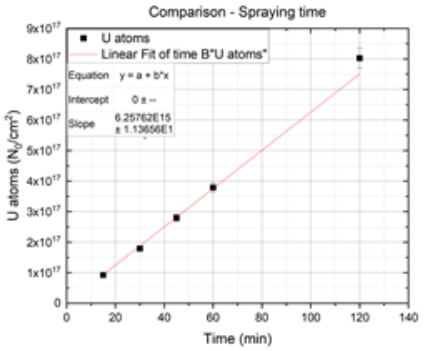

Fig. 7. Alpha spectroscopy results for targets with different spraying duration**.**

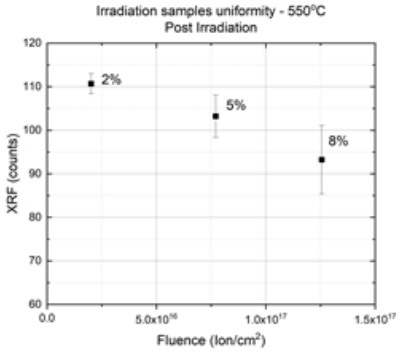

Fig. 8. XRF results for targets with different irradiation fluence and heat treatment at 550ºC.

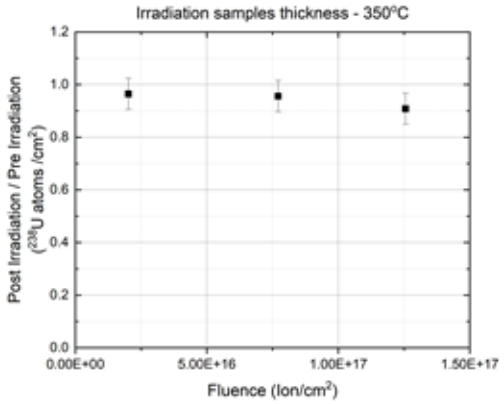

Fig. 9. Alpha spectroscopy results for target stability after irradiation and heat treatment at 350°C.

The next step in the project was to determine the robustness and purity of the targets under a neutron beam. For this investigation we utilized the neutron beam at the Los Alamos National Lab (LANL) using the DANCE detector array [1]. We used targets of varying thicknesses on both pure and alloy aluminum backings in order to determine the significance of the resulting γ-rays from the alloy's impurities in relation to the lines of interest from the uranium. The second aspect of the tests we performed at LANL was a measurement of the average total kinetic energy (TKE) of correlated fission fragments of ^{238}U as a function of incident neutron energy with a twin Frisch-gridded ionization chamber (FGIC) at WNR [2].

The data from the first experiment are currently being analyzed. Some preliminary results are presented below:

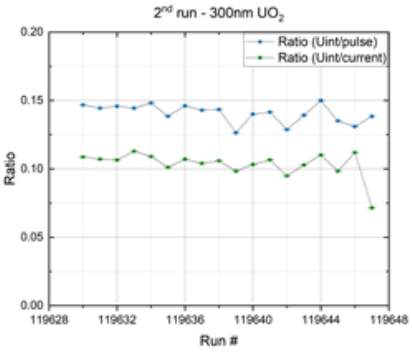

Fig. 10 Preliminary results of our targets stability under neutron beam.

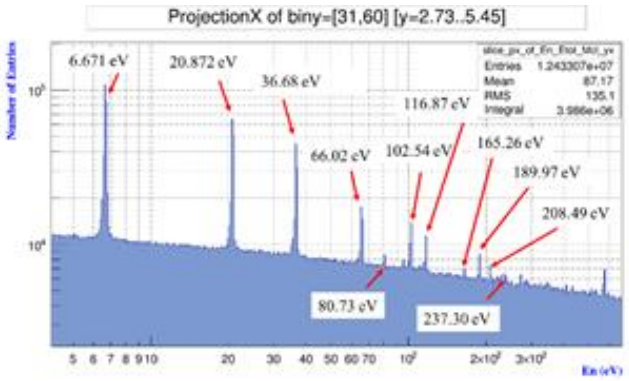

Fig. 11. Preliminary results of the ^{238}Uranium levels we are able to detect.

From the preliminary figures above, we can safely assume that our targets are fairly robust with no impurities that would interfere with our measurements.

Future work

Regarding future work, finalizing the results from the LANL experiment is the top priority. Analysis of our microscopy measurements are also being performed at the moment in order to fully understand our target's damage under the Ar beam, as well as the crystalizing mechanisms conditioning our targets.

Following that, we have started exploring the combustion properties of Eu as a surrogate for Am, which is going to be our next step in the actinide target production.

The final step would be a cross section measurement using the neutron beam that is being developed at University of Notre Dame in the following months.

[1] J.L. Ullmann, T. Kawano, T.A. Bredeweg, A. Couture, R.C. Haight, M. Jandel, J.M. O'Donnell, R.S. Rundberg, D.J. Vieira, J.B. Wilhelmy, J.A. Becker, A. Chyzh, C.Y. Wu, B. Baramsai, G.E. Mitchell, and M. Krtička, Phys. Rev. C **89**, 034603 (2014).

[2] D.L. Duke, F. Tovesson, A.B. Laptev, S. Mosby, F.J. Hambsch, T. Brys, and M. Vidali, Phys. Rev. C **94**, 054604 (2016).