UO_2 target preparation with spin coating assisted solution combustion synthesis

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Abstract. A simple and efficient target preparation method is developed combining spin coating and solution combustion synthesis. Multiple smooth and uniform UO_2 targets have been prepared using this method on a variety of backings (aluminium, carbon, silicon) used in nuclear physics experiments. The thicknesses of the targets can be precisely tuned by changing the number of coatings within the range of ~50-1000 μm/cm^2. These targets are highly uniform (<5% deviation), robust, and remain strongly adherent to their backings even after being irradiated by high doses (10^{17} ions/ cm^2) of 1.7 MeV Ar^{2+} ions.

1 Introduction

High quality, robust, and uniform actinide targets are important for a broad range of studies from nuclear astrophysics to the stockpile stewardship.
For obtaining accurate experimental results, uniform, crack- and contamination-free actinide targets are required. These should also be stable, structurally robust and adhere to the target backings and withstand high beam irradiation during experiments.

The various methods available for actinide target preparation have certain advantages and disadvantages. For example, mechanical processes such as cold rolling of metallic actinides pose safety hazards due to the pyrophoric nature of the metals [1]. In physical methods, vacuum evaporation requires a complex and high vacuum experimental set-up. The efficiency for this method is low and not suitable for making targets with expensive actinide materials [2]. The targets prepared by chemical methods (electrostatic deposition and molecular plating) often exhibit low stability under irradiation due to poor electrical and thermal conductivity [3]. Another proposed method for actinide target preparation is polymer assisted deposition (PAD). Although, PAD produces targets of high uniformity, this process requires long annealing periods at high temperature (1000 ºC) post deposition and specific backing materials [4].

At the University of Notre Dame, we have developed a simple and cost-efficient method for uranium dioxide (UO_2) target preparation using spin coating assisted solution combustion synthesis. The UO_2 targets are produced on different backings, and are found to be uniform, smooth and homogeneous. The target thicknesses can be well tuned within the ~50-1000 μm/cm^2 range. These show high thickness uniformity (<5% deviation) within a spot diameter of 25 mm. The targets are also found to not disintegrate or lose material under high intensity irradiation. The material yield of this method ~30%. In our previous work, we discussed the details of irradiation induced structural changes in UO_2 and $UO_{2.12}$ targets on aluminium backings [5-8]. In the current work, we provide a summary of this novel target making method for making highly uniform UO_2 targets that can be extended for preparing other actinide targets with a wide range of thicknesses on a variety of backings.

2 Experimental Methods

The optimal combination of the precursors (reactive solution, fuel and solvent solution) for the UO_2 target fabrication using solution combustion synthesis is determined using differential scanning calorimetry (DSC) coupled with thermogravimetric analysis (TGA) [7]. In this case, the reactive solution, fuel and solvent are isotopically depleted uranyl nitrate hexahydrate ($UO_2(NO_3)_2 \cdot 6H_2O$, International Bio-Analytical Industries, ≥98.0%), acetylacetone fuel ($C_5H_8O_2$, TCI, >99.0%) and 2-methoxyethanol ($C_3H_8O_2$, Alfa Aesar, 99%) respectively. The final working solution is a 0.25 M uranyl nitrate solution in a 0.5 molar ratio with the fuel.

A volume of 100 μL of the uranyl solution is spin coated on the atmospheric plasma treated backings at a

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speed of 3500 rpm/s for 35 seconds with a spin coater (SPIN 150i NNP). The angular acceleration is 100 rpm/s. Then the samples are annealed for 20 minutes at 400 ºC in a furnace, in order to produce the UO_2 layers. With this procedure, targets are produced on a variety of backings (aluminium alloy 6061, 99.997% pure aluminium, carbon and silicon) that are widely used as backings in nuclear experiments. The dimensions of both McMaster-Carr 6061 aluminium alloy and 99.997% puratronic grade Alfa Aesar aluminium discs are 42 mm in diameter and 0.7 mm in thickness. Carbon foils from ACF Metals used as backing are made of 98.9% ^{12}C and 1.1% ^{13}C. The dimensions of carbon backings made from these foils are 25 mm × 25 mm × 0.3 µm. The p type (boron doped) silicon wafer backings have a diameter of 50.8 mm and thickness of 0.3 mm. To prepare targets with different thicknesses, the spin coatings and combustion are repeated 1, 3, 5 and 7 times on aluminium alloy backings. The target made on pure aluminium is coated with the solution followed by a combustion of 30 times. In addition, three targets with identical parameters are made on aluminium alloy to investigate the reproducibility of the procedure.

The surface morphology of the targets is measured with a dual electron/ion beam Helios Nanolab 600 (FEI – Thermo Fisher Scientific). Their cross-sectional images are captured after depositing a protective platinum layer on top of the target layers using Helios. An Orbis (EDAX) X-ray fluorescence (XRF) spectrometer equipped with a Rh X-ray tube, poly-capillary optics, a sample stage with automated XYZ positioner and an 80 mm^2 Si(Li) drift detector is used to map the compositions of the targets and determine the uniformity as well as elemental abundances of the targets. The mapping is done by attaching the targets to the stage and scanning the target surface with a 30 µm X-ray beam spot under vacuum (~6 Pa absolute). The spatial resolution of images obtained from the mapping is ~160 µm. The experimental conditions are 40 keV and 400 mA with 0.6 second sampling time per spot. For analyzing elemental abundances, spot scans are done over the area of the targets with a 2 mm beam spot and 100 live seconds for each spot. An α-particle spectrometer (ORTEC-ULTRA-AS) with an ion-implanted silicon detector (active area – 900 mm^2, resolution - 29 keV at 5.486 MeV) is used to determine the absolute amounts of uranium isotopes in the targets prepared. The details of the calibration of the instrument and data analysis method are described in our previous work [7].

3 Results

The essential characteristics for targets are to be uniform and smooth, which are necessary to produce reliable data during nuclear experiments. The targets on aluminium, carbon and silicon backings prepared with spin coating assisted combustion method are found to be smooth without any defects. The nanoscale images of the cross sections of these target layers on all three different backings are uniform and pore-free (figure 1 A-C).

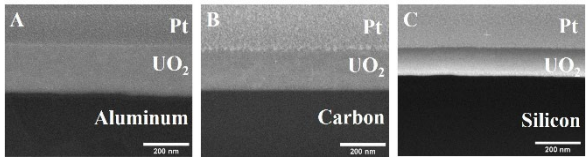

Fig. 1. Nanoscale images of the cross sections of uranium dioxide layers obtained on aluminium (A), carbon (B) and silicon (C) backings using the spin coating assisted solution combustion synthesis method. The images show smooth and uniform cross sections. Platinum is used for imaging purposes.

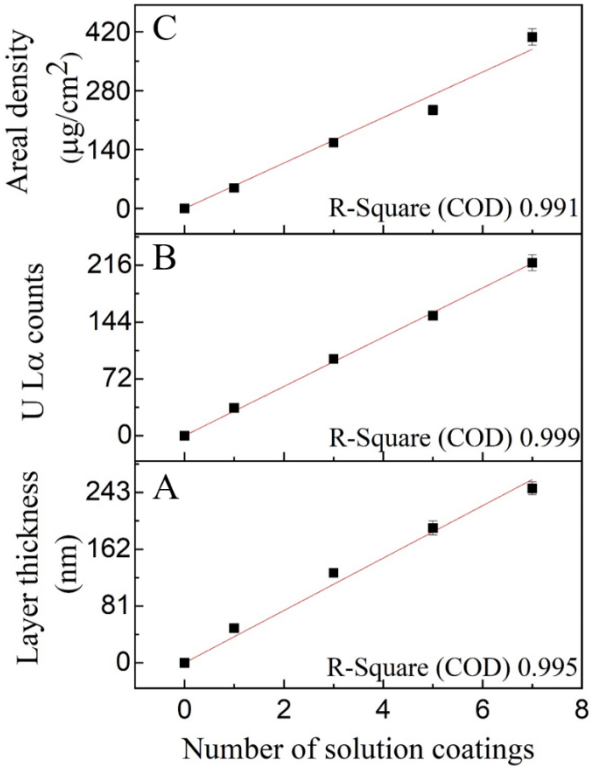

Fig 2. Number of solution coatings in function of layer thickness (A), U Lα counts (B) and areal density (C) derived from SEM, XRF and α–particle spectroscopy measurements, respectively. It is shown that the uranium content in the targets increases linearly with increasing number of coatings of the solutions.

The thicknesses of the UO_2 target layers are measured from the nanoscale cross section images. The target on the puratronic grade aluminium backing with 30 coatings is the thickest (~1000 µm/cm^2). The thicknesses of the 1, 3, 5 and 7 layered targets on aluminium alloy increase linearly with the increased number of layers (figure 2A). Counts of characteristic uranium X-ray lines (U Lα) are measured using XRF and are plotted (figure 2B) with respect to the number of target layers. The linear trend among them resembled the previous one in figure 2A. The U Lα counts are obtained from averaging 2 mm beam spot scans of 17 different points of each target, and the standard deviation in each case is less than 5%. A third method, α-particle spectroscopy is used to confirm a linear trend on an even larger (~cm) scale. Alpha-particles emitted from the targets are measured and used to determine the areal density of the total uranium layer in µg/cm^2. The areal density of the total uranium layer also showed a

similar trend with respect to the number of layers as seen from the SEM and XRF techniques (figure 2C).

We also investigated the overall uniformity of these targets using an XRF mapping. As seen in the first column of the figure 3, the blank target does not show any uranium content. Absence of bright green spots (U M lines) anywhere on the backing or the XRF stage indicates that there is no detectable uranium. Al K lines emitted from the aluminium backing are denoted by red. In the targets with 1, 3, 5 and 7 coatings, the bright green circular spot at the centre of the backing signifies the presence of uranium. The brightness of the green spots increases with the number of coatings. At the same time, the intensity of the Al K line distribution decreases with increasing coatings. Lesser number of characteristic X-rays of aluminium can be detected in these cases due to the higher content of uranium on the aluminium backing.

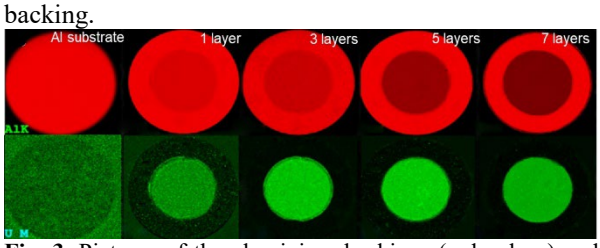

Fig. 3. Pictures of the aluminium backings (red colour) and uranium layers (green colour) derived by XRF mapping showing an overall uniformity of the UO_2 targets with different layers.

Three UO_2 targets prepared with identical parameters are also mapped using the same method and they show a uniform presence of U M lines in the active target region. The intensity of the U M lines is the same for each, which suggests similar thicknesses. A same amount of decrease in the intensity of the Al K lines in the target region confirms that as well. Several spot scans are done all over these three targets and the standard deviation of the U Lα counts among them is less than 1%.

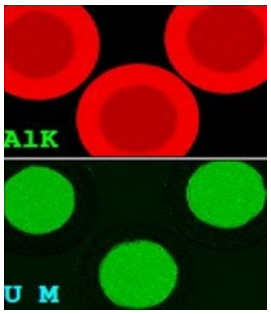

Fig. 4: Pictures of the aluminium backings (red colour) and uranium layers (green colour) of three UO_2 targets prepared with identical parameters derived by XRF mapping showing a reproducibility of the target preparation method.

These targets on aluminum backings are irradiated with 1.7 MeV energy Argon ions (Ar^{2+}) to a fluence (~10^{17} ion/cm^2) under a controlled temperature. The uranium content on the targets is checked using α-particle spectroscopy after irradiation and no significant material loss is found.

4 Discussion and Conclusion

Several UO_2 targets are produced using a spin coating assisted combustion technique on different backings (aluminium, carbon and silicon) used in nuclear experiments. In each case, the surfaces as well as the cross sections are found to be smooth and uniform. A linear relationship is determined between the number of layers coated on the backings and the final UO_2 content on the targets using SEM, XRF and α-particle spectroscopy. This target preparation method provides a precise control of the target thicknesses within the ~50-1000 μm/cm^2 range. The targets produced are highly reproducible. The targets are exposed to high doses of Argon ion irradiation (~10^{17} ion/cm^2). They did not disintegrate or lose adherence to the backing during the experiment. The total uranium content of the targets is also found to remain the same after the irradiation. Overall, this novel method proves to be an excellent candidate for making UO_2 targets of a variety of thicknesses on different backings and can potentially be extended for other actinide target making.

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