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**Plasma Sprayed Zirconium for US HPRR LEU Conversion
Fuel Diffusion Barrier**

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ABSTRACT

Plasma spraying of zirconium (Zr) is being investigated for the diffusion barrier between the U10Mo fissile material and the aluminum cladding for US HPRR LEU fuel. Interest in plasma spraying is in part due to the application of the Zr to the U10Mo late in the manufacturing process allowing for the more efficient recycle of scrap fuel material which allows higher LEU utilization. Recent results have explored the scale up from mini-plate sized samples to full sized HPRR fuel samples. In addition, coatings on the edges of the fuel are being investigated to prevent all contact between the U10Mo and the Al cladding. Activities involving plasma sprayed Zr for the upcoming MP-1 reactor test in ATR are also discussed. A subset of the MP-1 irradiation test samples will utilize plasma sprayed Zr for the diffusion barrier.

1. Introduction

In support of the United States' nonproliferation and highly enriched uranium (HEU) minimization policies, the U.S. Department of Energy (DOE)/National Nuclear Security Administration's (NNSA) Global Threat Reduction Initiative (GTRI) is actively working to convert civilian research and test reactors from the use of HEU fuel to low enriched uranium (LEU) fuel. If no suitable LEU fuels are available, the program contributes to the development of new LEU fuels. The Reactor Conversion program is developing a high-density monolithic plate fuel system which uses low enriched uranium 10 wt% molybdenum (U10Mo) foils clad with aluminum (Al) where a zirconium (Zr) diffusion barrier is placed between the U10Mo and the Al [1]. This fuel is intended for use in the five (MURR, NIST, MIT, HFIR, ATR) US high performance research reactors (HPRR).

The application of the Zr diffusion barrier between the U10Mo fuel and the Al cladding by plasma spraying has been an ongoing area of research [2-10]. The goal of the study described

here was to demonstrate the plasma spraying technique for depositing Zr onto full sized U10Mo foils to demonstrate scale up of the process from the mini foil samples (25 mm x 100 mm) to full sized samples. Full size denotes one of the actual fuel foil sizes for one of the five US HPRRs to be converted to LEU fuel in the US DOE Reactor Convert program. The smallest full sized fuel foil is for the NIST reactor with a size of 289 mm x 62 mm. The largest full sized fuel foil is for the ATR reactor with a size of 1219 mm x 87 mm. The widest full sized fuel foil is for the MURR reactor with a size of 610 mm x 103 mm. The current LANL vacuum plasma spray system can accommodate a foil length of 620 mm or less and can coat lengths of 350 mm or less in a single run. Therefore, as a demonstration of the the maximum length foil that can be processed on the current LANL system, a 610 mm long foil was chosen as the goal for scale up activities.

In addition to scale up studies for full sized fuel foils, coating of the edges of the fuel was investigated. Since the plasma sprayed samples are held down around the sample periphery during coating (3 mm of the periphery used to hold down the sample remains uncoated), the uncoated portion of the foil is removed by shearing after the coating process leaving the edge of the fuel uncoated. Plasma spraying and resistance welding techniques were investigated as methods for edge coating. If it is determined in the future that edge coatings are desirable, an understanding of the techniques that produce acceptable edge coatings is needed. Therefore, experiments to demonstrate edge coatings and characterize the coating microstructure were conducted and are reported here.

Plasma spraying for application of the Zr diffusion barrier is one of the techniques to be used in the MP-1 reactor test series planned as part of the US HPRR monolithic fuel evaluation program. The MP-1 test series will evaluate monolithic U10Mo fuel performance in the ATR reactor at Idaho National Laboratory. Several different fuel fabrication techniques giving different microstructures will be utilized to produce the test fuel. The objective of the MP-1 irradiation test is to provide sufficient information on fuel performance, in concert with fabrication studies, to down-select an LEU fuel design that meets US HPRR fuel performance and commercial fabrication requirements. In order to satisfy the specifications for MP-1 test fuel, the thickness of the Zr layer on the LEU fuel must be measured by a non-destructive technique. The non-contact thickness measurement technique of X-ray fluorescence (XRF) is being evaluated in addition to traditional contact metrology methods. Preliminary results of this investigation are reported here.

2. Experimental Procedure

During plasma spraying of the Zr layer on the U10Mo, a spinning sample holder was utilized in order to reduce the localized heat loading and subsequent steep thermal gradients in the U10Mo sample that could lead to part distortion in the foil during coating. This fixture is shown in Fig. 1 mounted in the plasma spray chamber. The rotation speed used for coating foils was 125 RPM and the torch translation speed was 34 mm/s. Plasma spray parameters were: Ar torch gas 35 standard liters per minute (slm); He torch gas 18 slm; Ar powder gas 3 slm; chamber pressure 40 kPa; standoff distance 100 mm; powder feed rate 0.5 RPM; torch current 850 A; torch voltage 25 V; TA current 70 to 100 A AC; 6 preheat passes; 28 powder passes; 4 post spray passes. Two

full sized U10Mo (depleted) foils were available for Zr plasma spray coating. The first (FS1) was 343 mm x 81 mm x 0.36 mm and the second (FS2) was 610 mm x 73 mm x 0.38 mm. The first foil is slightly larger than the NIST reactor foils and the second foil is similar in size to one of the MURR foils.

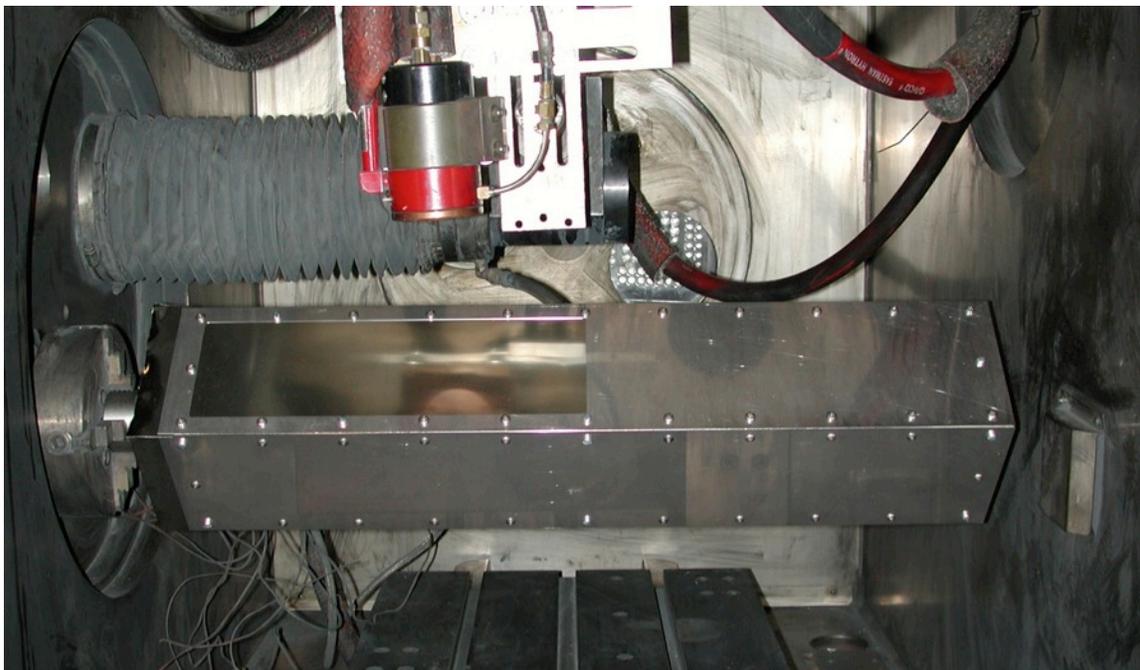


Fig. 1. Spinning foil holding fixture (lower center of image) mounted in the plasma spray chamber under the plasma torch (red on bottom and black on top).

Stainless steel (a surrogate for uranium) foil (250 μm thick) sample edges were covered with Zr by plasma spraying or resistance welding. The plasma spray parameters were: Ar torch gas 35 (slm); He torch gas 20 slm; Ar powder gas 2.5 slm; chamber pressure 9.3 kPa; standoff distance 286 mm; powder feed rate 0.6 RPM; torch current 950 A; torch voltage 28 V; transferred arc (TA) current 40 A AC; translation speed 26 mm/s, 2 preheat passes; 4 powder passes; 2 post spray passes. Zr foil with thickness 30 μm and 80 μm was welded onto stainless steel samples using capacitive discharge resistance welding. The weld output voltage was 5 V and the weld repetition rate was 4 Hz. The welding was performed by hand so the welding pressure was not precisely controlled. Prior to spraying or welding, the substrates were cleaned with isopropanol.

3. Results and Discussion

3.1 U-10Mo Full Sized Sample Coating

A plasma sprayed Zr deposit of length 305 mm was applied on both sides of the FS1 foil. The backside of the foil reached a maximum temperature of 700°C during Zr coating as measured by a thermocouple in contact with the back of the sample. The foil before coating is shown in Fig. 2. The foil had a significant curvature in the long dimension as can be observed from the figure.

The foil in the plasma spray chamber after coating the second side is shown in Fig. 3. The foil coated on both sides is shown in Fig. 4 and was much flatter than the uncoated foil but had some small scale waviness.



Fig. 2. FS1 U10Mo foil before coating showing significant curvature in the sample.

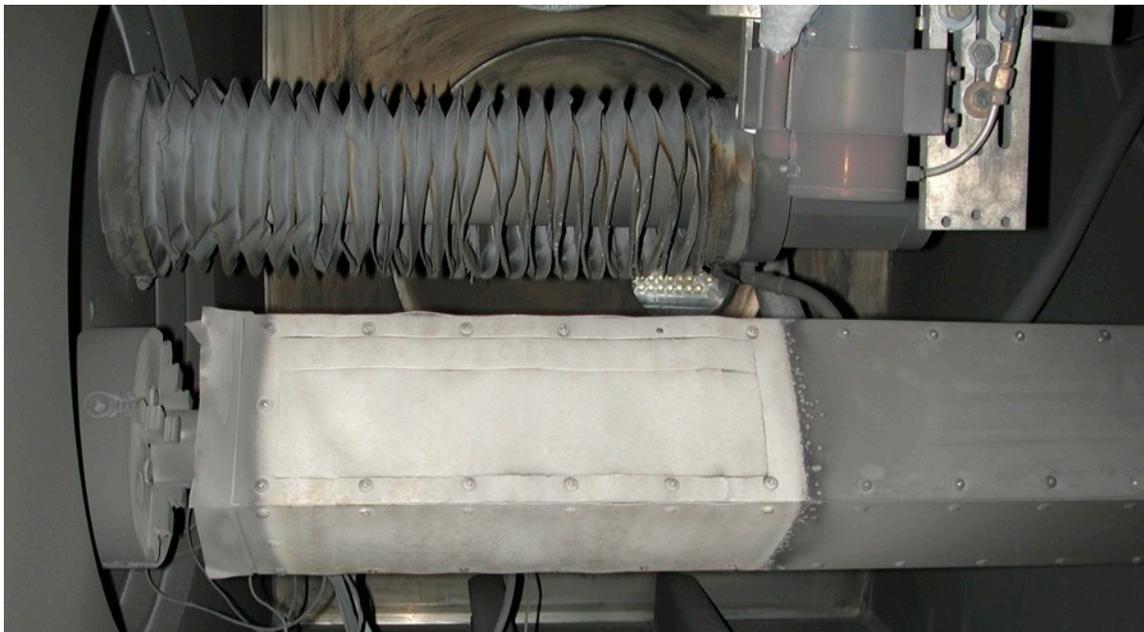


Fig. 3. FS1 U10Mo foil coated with Zr shown in the spray chamber attached to the rotating fixture.



Fig. 4. FS1 U10Mo foil coated with Zr on both sides showing reduced curvature compared to the uncoated foil (Fig. 2).

The FS2 foil was coated in four separate runs in order to cover the full length and both sides.

The TA current was increased from 70 A on FS1 to 100 A on FS2. Increasing the TA current causes more substrate heating and cathodic arc cleaning. The FS2 foil was annealed at 650°C for 1 hour followed by an oil quench prior to Zr coating. The annealed foil FS2 was noticeably more flexible than FS1 which was not annealed after cold rolling. The FS2 foil before coating is shown in Fig. 5 and did not have the curvature like that shown for FS1 in Fig. 2. An image of the plasma spray operation during coating is shown in Fig. 6. Foil FS2 after coating is shown in Fig. 7. The maximum temperature on the back side of foil FS2 during spraying was 750°C. The foil had a slight amount of waviness after coating similar to the FS1 foil.



Fig. 5. FS2 U10Mo annealed foil prior to Zr coating.

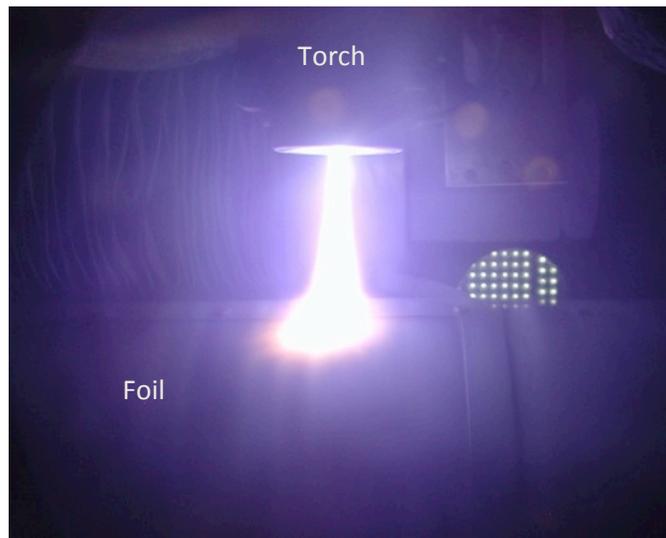


Fig. 6. Plasma torch spraying Zr powder onto the rotating fixture holding the FS2 U10Mo foil.



Fig. 7. FS2 U10Mo foil plasma spray coated with Zr. The dark line in the center of the sample shows the limits of the two separate spray runs needed to cover the entire top surface.

The FS1 U10Mo foil coated with Zr was sheared along the edges of the coating to remove the uncoated periphery. The sheared edges of the foil showed no Zr delamination as can be seen in Fig. 8. The LEU foils needed for the US HPRR reactors would have the uncoated edges sheared

off in a similar fashion before being clad in the Al outer layer.

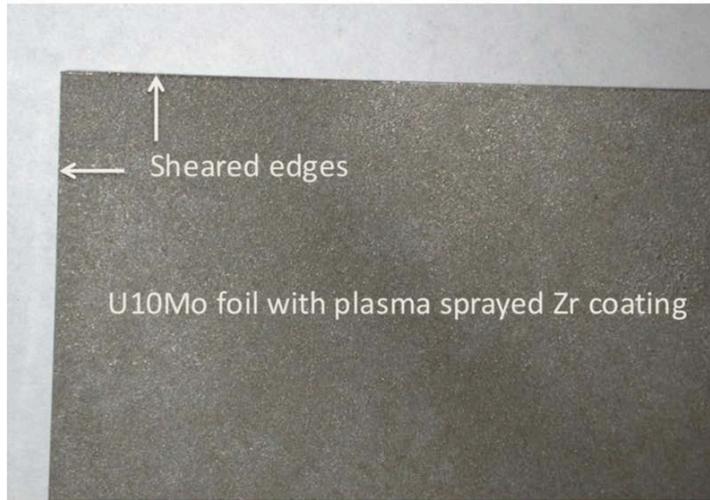


Fig. 8 Sheared edges of Zr plasma spray coated U10Mo FS1 foil.

3.2 U10Mo Coated Sample Characterization

The interface between the Zr and the U10Mo is an important feature to examine in order to predict the bond quality. Having little or no oxide at the interface promotes a tougher bond between materials. Examination of the U10Mo/Zr interface reveals the presence of an oxide as is shown in the SEM-BSE image in Fig. 9 and the elemental maps shown in Fig. 10. The presence of the oxide generally reduces the bond strength between the U-Mo and the Zr. Due to the presence of the uranium oxide, higher TA current levels in subsequent full scale foil coating runs is warranted.

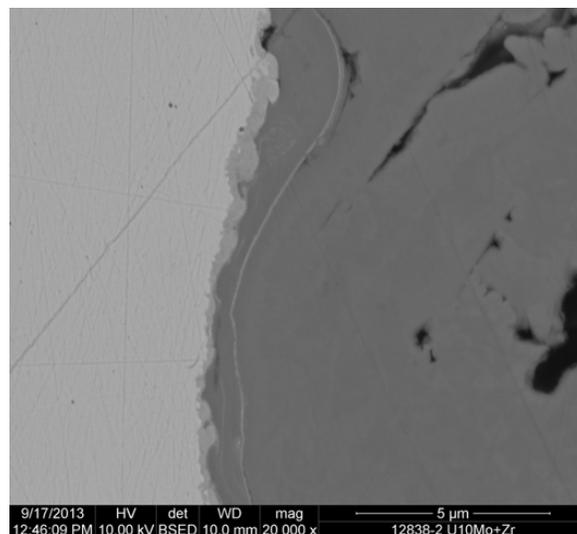


Fig. 9 SEM-BSE image of U10Mo (left side)/Zr (right side) interface in the FS1 foil.

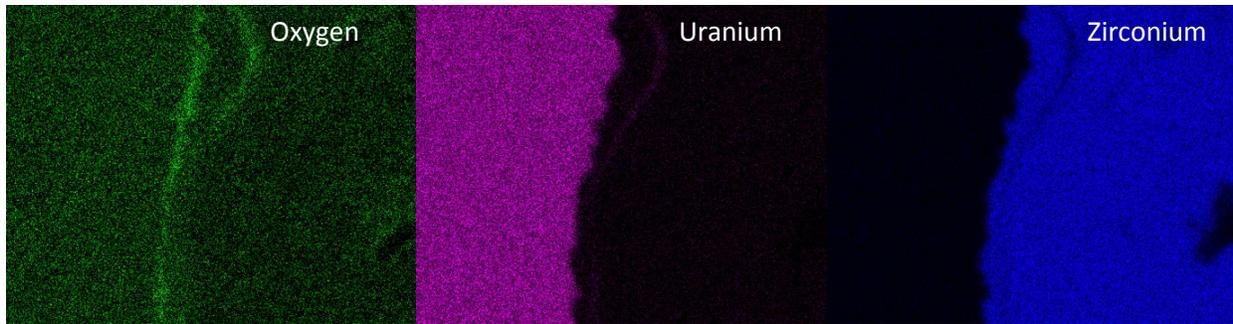


Fig. 10. Elemental maps of oxygen, uranium and zirconium for the area shown in Fig. 9.

3.3 Sample Edge Coating by Plasma Spraying

A plasma sprayed Zr coating applied to the edge of a surrogate stainless steel (SS) foil is shown in Fig 11. The Zr thickness is approximately 35 μm . The density of the Zr is high (no porosity visible) and the interface between the SS and Zr shows continuous close contact between the two materials. The contrast in the SEM image indicates that some mixing of the SS and Zr took place. This was due to the high temperature of the substrate during Zr deposition, which at the very surface, was close to the melting point of SS.

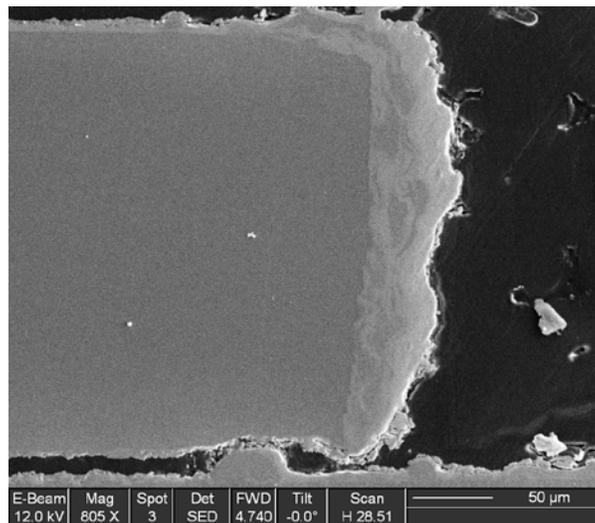


Fig. 11. Cross section of a stainless steel (left side) sample plasma sprayed on the edge with Zr (right side on edge of SS).

3.4 Sample Edge Coating by Resistance Welding

An optical cross sectional view of a SS foil covered by an 80 μm Zr foil which was resistance welded into place is shown in Fig. 12. The top and bottom of the SS foil are covered in addition to the edge. In this early development sample, there are some areas of incomplete bonding between the SS and Zr. However, the concept of using a Zr foil to cover the thin edges of the

sample and also the larger area top and bottom sides as well is demonstrated. An SEM back scattered electron (BSE) image with a superimposed line scan elemental analysis for a sample of the SS foil covered by a 30 μm Zr foil is shown in Fig. 13. The interface between the Zr and SS is quite distinct as indicated by the abrupt contrast change in the image and rapid transition in elemental composition in the line scan plot. The SS and Zr are in intimate contact with a mixed region on the order of 1 μm thick. The mixed region in other locations of the SS/Zr interface was measured to be between 1 and 25 μm in thickness depending on the local electrode pressure and the the electrical contact between the Zr and the SS during the initial portion of the weld.

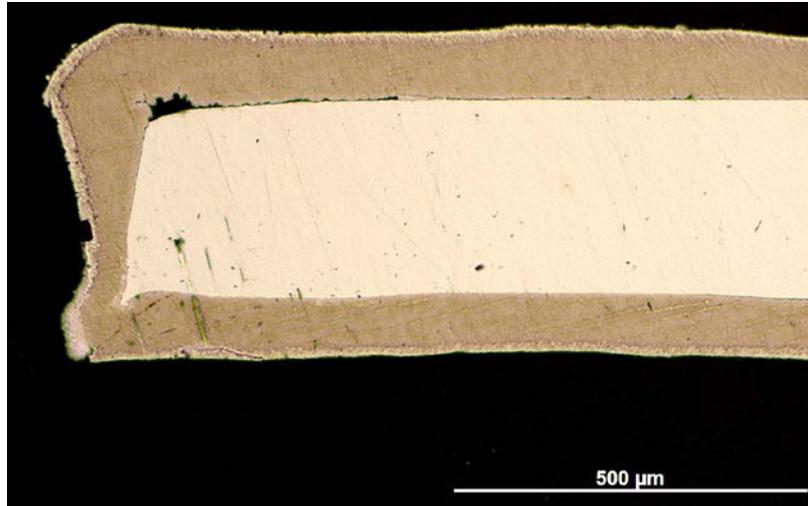


Fig. 12. Optical metallographic cross section of a stainless steel foil (center lighter object) with an 80 μm Zr foil resistance welded to the top, bottom and side edge.

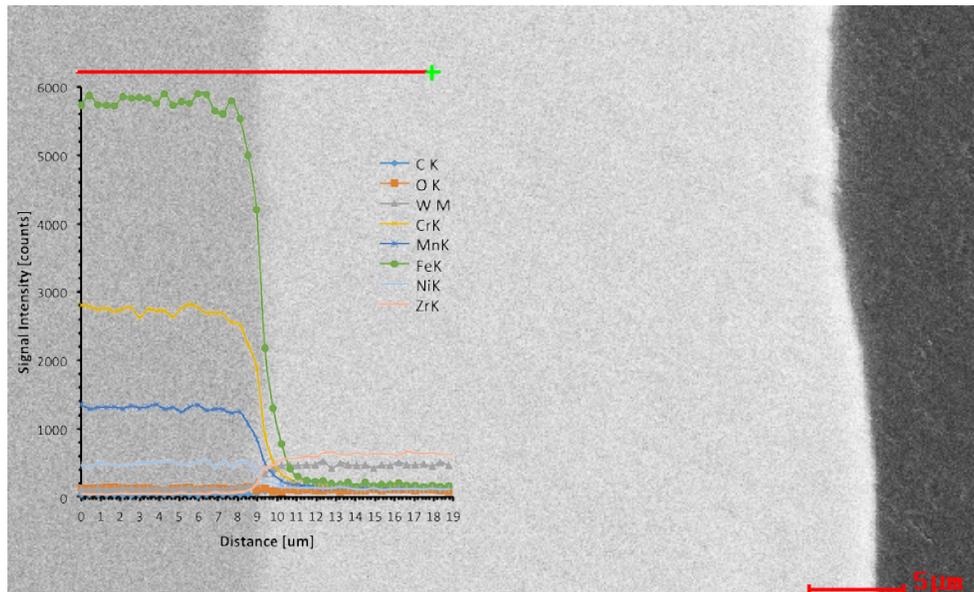


Fig. 13. SEM-BSE image and line scan (along the red line) elemental analysis of the cross section of a SS foil with a 30 μm Zr foil resistance welded to the surface.

3.5 Zr Coating Thickness Measured by XRF

The Zr thickness deposited on sample FS2 was estimated by measuring the mass of the foil before and after coating and assuming full theoretical density of the coating. On the side of FS2 visible in Fig. 7, the Zr thickness was estimated to be 44 μm (left side in Fig. 7) and 45 μm (right side in Fig. 7) using the mass gain method.

In order to gain spatial resolution of the Zr thickness measurement, XRF was performed with a spot size of 10 mm and a spacing between spots of 13 mm. The ratio of the Zr to U line intensities was used to calculate the Zr thickness. Zr foils of known thickness in the 5 to 50 μm range were used to calibrate the technique. The XRF Zr thickness measurements along the centerline (moving down the long dimension of the foil) are shown in Fig. 14. The XRF values indicates a maximum variation in the Zr thickness between points of 27 μm . The average XRF Zr thickness on the centerline is 45 μm . This is in good agreement with the mass based measurement.

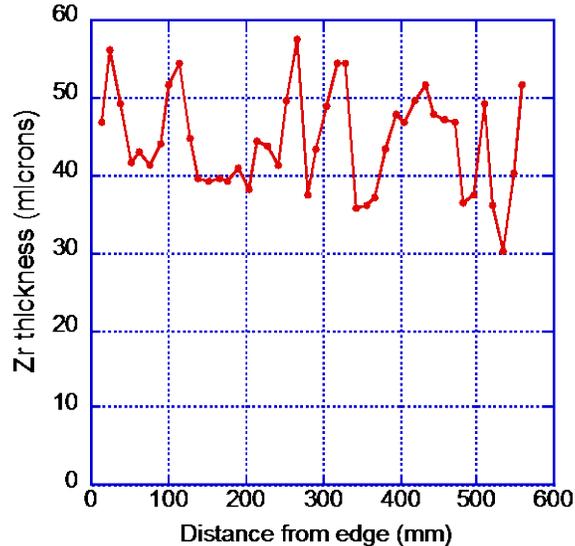


Fig. 14. XRF measurement of Zr coating thickness along the centerline of foil FS2.

4. Summary

Recent results for studies involving the plasma spraying of Zr for US HPRR LEU fuel development have demonstrated the following:

- Full sized U10Mo fuel foils with dimensions approximating the fuel for NIST and MURR have been successfully coated with Zr by plasma spraying.
- One full sized foil (not annealed) showed a significant reduction in large scale curvature after spraying while the other full sized foil (annealed) was flat before and after coating.
- After Zr coating, both full sized foils showed some small scale waviness.
- Coating of the foil edges was demonstrated by both plasma spraying and resistance welding.

- Resistance welding was also demonstrated for applying Zr foil to the top and bottom of a surrogate fuel foil.
- XRF for determining Zr coating thickness has been demonstrated for plasma sprayed Zr on U10Mo. The results are in agreement with a mass based coating thickness estimate.

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