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Fusion & Materials for Nuclear Systems

FABRICATION OF NATURAL URANIMUM UO_2 DISKS (PHASE II): TEXAS A&M WORK FOR OTHERS SUMMARY DOCUMENT

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ABSTRACT

The steps to fabricate natural UO_2 disks for an irradiation campaign led by Texas A&M University are outlined. The process was initiated with stoichiometry adjustment of parent, U_3O_8 powder. The next stage of sample preparation involved exploratory pellet pressing and sintering to achieve the desired natural UO_2 pellet densities. Ideal densities were achieved through the use of a bimodal powder size blend. The steps involved with disk fabrication are also presented, describing the coring and thinning process executed to achieve final dimensionality.

1. OVERVIEW

As part of a work for others agreement with Texas A&M University (TAMU) in support of their work for "Fabrication and Irradiation of Natural Uranium UO_2 Disks (Phase II) Proposal Number NFE-12-04134", Oak Ridge National Laboratory (ORNL) has agreed to fabricate and characterize sample disks of natural uranium UO_2. Following fabrication ORNL has agreed to ship said samples to TAMU for continued analysis. The requirements for the final sample disks are as follows: disks should be approximately 3-mm in diameter and $\leq$0.240 mm thick while the density of the fully sintered disks should be ~10.4 g/cm^3.

2. COLD-PRESS AND SINTERING OF NUO_2 DISKS

The parent material identified for the natural UO_2 disk fabrication was sub-micron natural U_3O_8 powder. The parent powder was reduced to achieve the UO_2 stoichiometry necessary for pellet fabrication. The reduction heat treatment was carried out in a Lindberg tube furnace. The powder was loaded in a clean Al_2O_3 boat and exposed at 900°C for 24 hours under flowing Ar-4%H_2. After the furnace heat treatment, the reduced powder cooled in the furnace under the Ar-4%H_2 flowing gas for four hours. This material is known as heat-one powder (HT-1). The powder remained fine grained and was suspected to be hyperstoichiometric. A subset of this material was further heat treated to coarsen the powder and further reduce the material prior to sintering. The secondary reduction step involved a 1000°C for 4 hours under flowing Ar-4%H_2 heat treatment. Material from this lot is known as heat-two powder (HT-2).

Pellet pressing was conducted using a Carver press with a ½" die set as shown in Figure 1. Prior to pellet pressing, the die bore and rams were cleaned with stearic acid. An initial exploratory set of pellets (Pellets 1-6) was pressed using HT-1 and HT-2 powders. Pellets 1 and 2 were a blend of HT-1 and HT-2 and were pressed at pressures of 75 MPa and 150 MPa respectively. Pellets 3 and 4 were 100% HT-2 and were pressed at pressures of 75 MPa and 150 MPa respectively. Pellets 5 and 6 utilized a binder addition, where a binder was added to the powder to assist in set-up during pressing. The binder addition was accomplished by combining 0.003 g PEG (8000 mw) with 1 g powder along with 5x volume of NERL water. The slurry was stirred, rested, and stirred again followed by an overnight drying step at 75°C after which, pellet 5 and 6 were pressed at pressures of 75 MPa and 150 MPa respectively.

Figure 1: Carver press utilized for natural UO_2 pellet fabrication.

The exploratory pressed pellets were then transferred to the ASTRO graphite furnace for sintering, as shown in Figure 2. The pellets were spaced evenly on a Ta boat and loaded into the furnace. The furnace was evacuated and backfilled with Ar with an Ar flow rate of ~200 cc/min set during thermal exposure. The samples were then sintered according to the following furnace program: ramp to 450°C at 2°C/min, hold at 450°C for one hour, ramp to 1750°C at 5°C/min and hold at 1750°C for 6 hours, cool to room temperature at 5°C/min. Inspection of the sintered pellets indicated the quality was not sufficient as the pellets were frangible and not intact. The use of the binder resulted in a discoloration of the pellets, suggesting a reaction of the natural UO_{2+x} powder with the binder.

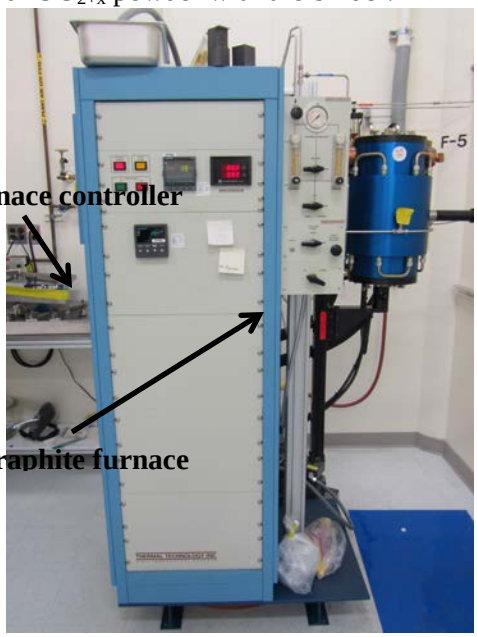

Figure 2: ASTRO graphite furnace utilized for sintering pressed pellets.

A second set of pellets (Pellets 7-10) were pressed and sintered at 1800°C according the conditions listed in Table 1. The second set of pellets did not use a binder addition and were fabricated using either HT-1 or HT-2 powders. The thermal exposure followed the same ramp and cooling cycle discussed in the exploratory pellet fabrication. Here the effect of a higher sintering temperature was explored as well as the heat powder and pressing conditions. The weight of the pellets and the measured dimensions were used to estimate the density. Pellet 7 did not have sufficient integrity to determine it's density. The density for Pellets 8-10 were below the density requirement of ~10.4 g/cm^3 and showed no strong correlation with the pressing conditions. Based on these results, efforts to improve the packing density prior to sintering were sought.

Table 1: Pellet 7-10 fabrication conditions and results, sintered at 1800°C.

Pellet ID	Powder Fraction		Pressure	Diameter		Thickness	Weight	Dimensional Density	
	HT-1	HT-2	MPa	X (mm)	Y (mm)	Z (mm)	g	g/cm^3	%TD
7	0	100	69	-	-	-	-	-	-
8	100	0	69	10.92	10.87	1.14	0.999	9.40	85.69%
9	100	0	55	10.86	10.87	1.27	1.030	8.75	79.74%
10	100	0	83	10.90	10.96	1.21	1.002	8.83	80.45%

Improved packing fractions were obtained by using a bimodal powder size distribution during pressing. Grain growth was expected during the sintering of Pellets 7-10. Coarse powder material was obtained from Pellets 7-10 by milling the pellets in a Spex mill for 1 hour using 5 min intervals. The milling media was 4-mm alumina beads and resulted in approximately 3.5 g of recovered coarse powder. This milled material is denoted as HT-M.

A systematic approach to the effect of powder ratios (HT-1 to HT-M) was examined. A higher sintering temperature of 1900°C using the same ramp and cool recipe as described earlier was employed to improve densification. Table 2 lists the experimental test matrix and associated results. Density measurements were obtained by two methods for Pellets 11-16, by dimensional analysis and by Archimedes' principle. Figure 3 shows the nature of pellets 11-16 after firing. Dimensional density analysis of pellets 11-12 was not accurately employed as the samples experienced chipping of their edges leading to material loss. The new conditions led to a 7-8% improvement in density compared to pellets 7-10 using the dimensional density analysis, however, the density was still below the requirement of ~10.4 g/cm^3. The Archimedes' principle density measurement removes the error caused by variations is surface curvature and surface flaws and variable dimensions, which are captured by the dimensional density analysis. The Archimedes' principle analysis indicates pellets 13-16 met the density requirement.

Table 2: Pellet 11-16 fabrication conditions and results, sintered at 1900°C.

Pellet ID	Powder Fraction		Pressure	Diameter		Thickness	Weight	Dimensional Density		Archimedes Density	
	HT-1	HT-M	MPa	X (mm)	Y (mm)	Z (mm)	g	g/cm^3	%TD	g/cm^3	%TD
11	75	25	34.5	10.81	5.65	1.31	0.526	-	-	10.030	91.43%
12	75	25	69	10.81	10.87	1.21	0.823	-	-	10.127	92.32%
13	50	50	34.5	10.60	10.59	1.29	0.983	8.64	78.79%	10.330	94.17%
14	50	50	69	10.83	10.87	1.10	0.987	9.70	88.46%	10.460	95.35%
15	50	50	124.1	10.88	10.89	1.10	0.981	9.58	87.36%	10.456	95.31%
16	25	75	124.1	11.08	11.01	1.06	0.986	9.71	88.53%	10.508	95.79%

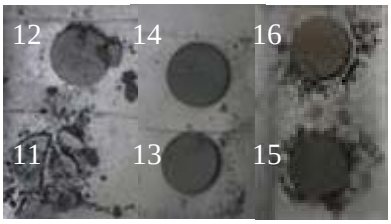

Figure 3: Images of pellets 11-16, label indicates the pellet identity.

Verification of the pellet composition was determined using x-ray diffraction (XRD). Pellet 12 was examined, as XRD analysis requires a powder sample. Obtaining a powder sample requires destruction of the parent pellet and pellets 13-16 were identified as acceptable candidate for coring. Figure 4 shows the spectrum associated with pellet 12. The spectrum confirms the pellet is UO_2 with no observed secondary phases. The successful fabrication of natural UO_2 pellets with densities ~10.4 g/cm^3 was achieved and pellets 13-15 were selected for disk fabrication.

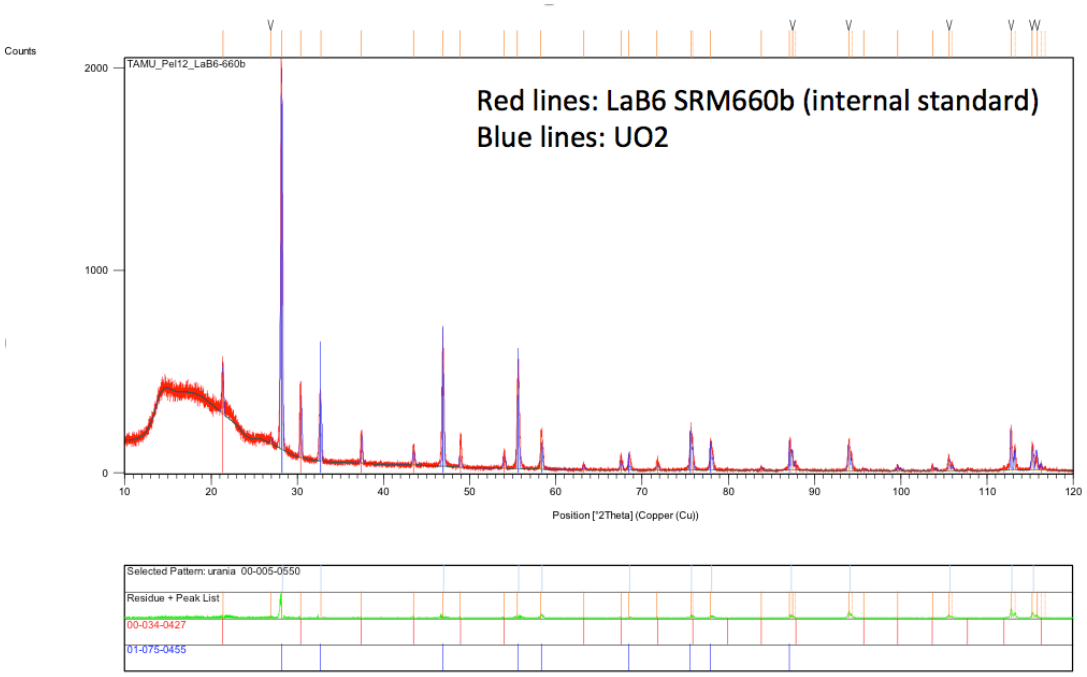

Figure 4: XRD spectrum of pellet 12, confirming UO_2 stoichiometry.

3. DISK FABRICATION AND VISUAL INSPECTION

A series of disk samples were prepared from the larger pellets with acceptable density. Three, 3-mm disks were cored from pellets 13-15. The coring was completed using a diamond-tipped coring bit on a small Sherline end-mill operated in a hood in the Uranium Development Laboratory at ORNL. Prior to coring, the pellets were attached to an aluminum mount with crystal bond. After coring the small disks were liberated from the aluminum mount by dissolving the crystal bond in acetone. The yield was not 100% as flaws were present in the pellets in the form of cracks. The flaws present after pressing and sintering led to only two disks being recovered from each parent pellet (pellets 13-15).

The 3-mm disks were thinned to ≤0.240 mm using a Buehler Mini-MET system with a micron-scale thinning attachment. The cored samples were attached on a circular glass slide and polished to a 6-µm finish. After polishing one side, the disk was removed from the glass slide and placed polished side down then attached to the glass slide to ensure the side remained parallel during polishing and thinning. The micrometer thinning attachment was then slowly advanced to remove material. The thickness of the disk was periodically checked during thinning using a micron-scale height gauge measuring the difference from the base of the glass slide to the top of the UO_2 disk to determine thickness. Once the disk was thinned to ≤0.240 mm the sample was removed from the glass by soaking it in acetone. The final finish on both sides was 6-µm.

Table 3: Disk properties after coring and thinning.

Disk	Archimedes	Disk Properties		
Pellet/ID	Density (g/cm3)	Thickness (mm)	Mass (g)	Notes
13-A	10.33	0.23	0.0153	Shipped
13-B	10.33	0.17	0.0113	Shipped
14-A	10.46	0.15	0.0106	Crack in sample
14-B	10.46	0.23	0.0166	Shipped
15-A	10.46	0.24	0.0161	Shipped
15-B	10.46	0.24	0.0167	Shipped

Table 3 shows the final dimensions of the thinned disks from pellets 12-15. Some disks did not survive the thinning process due to fractures in the samples. Figures 5-7 show the front and back appearance of successfully thinned samples as taken using an optical microscope. Samples with limited surface flaws were deemed acceptable and shipped to Texas A&M University. The samples selected for shipment are noted in Table 3.

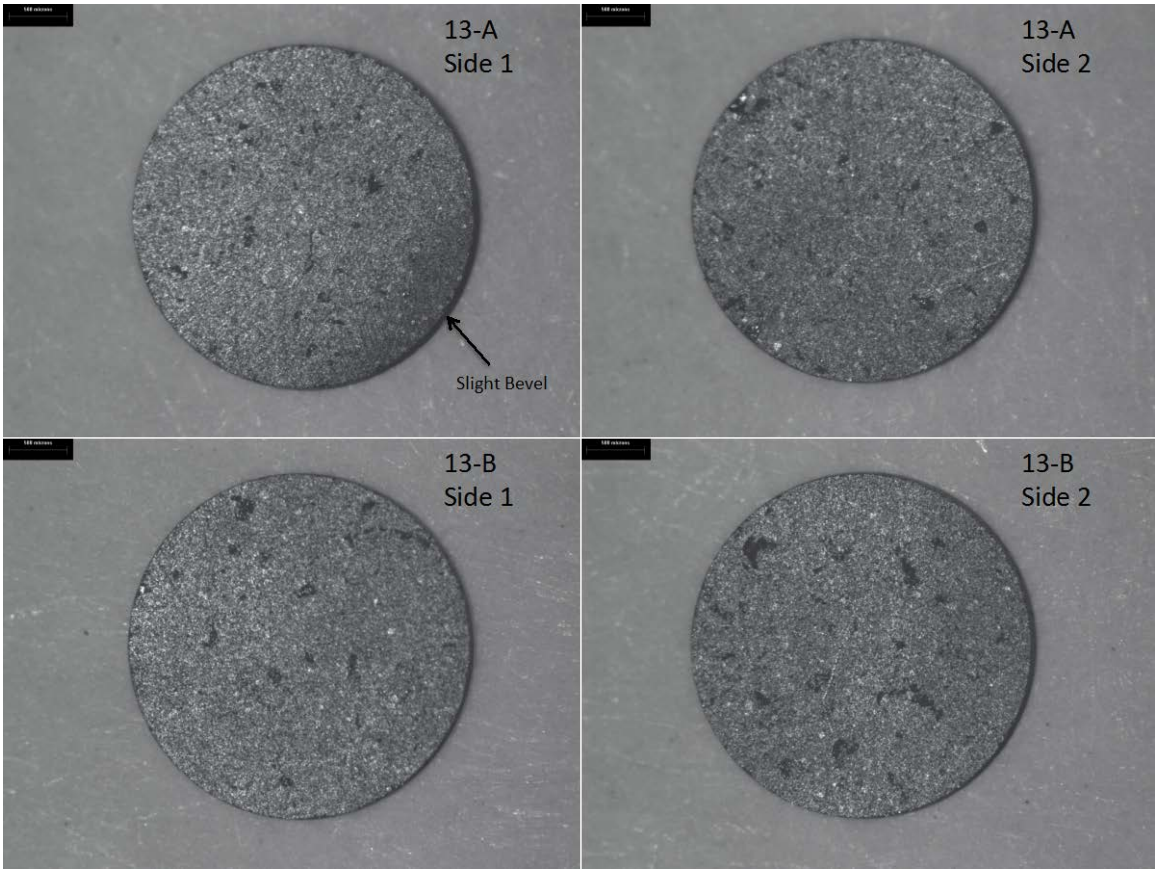

Figure 5: Appearance of natural UO_2 disks cored and polished from pellet 13.

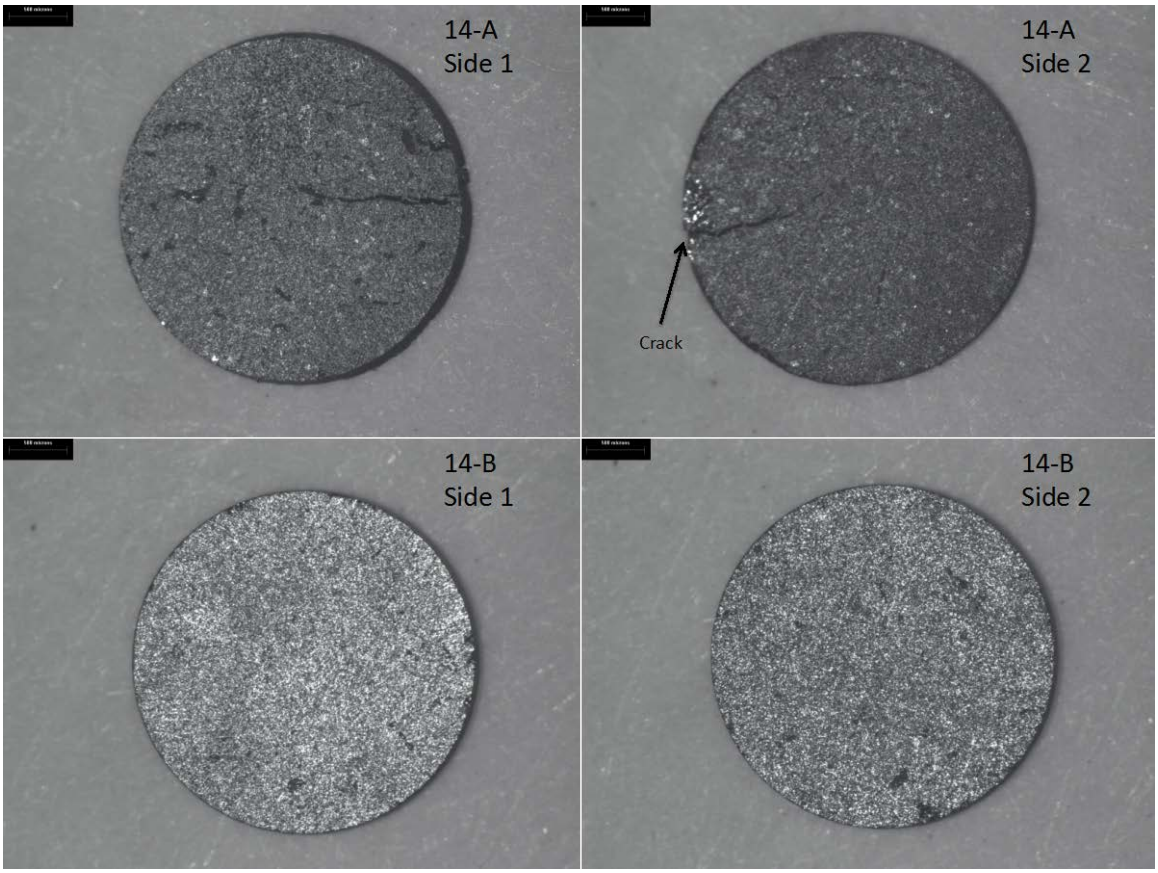

Figure 6: Appearance of natural UO_2 disks cored and polished from pellet 14.

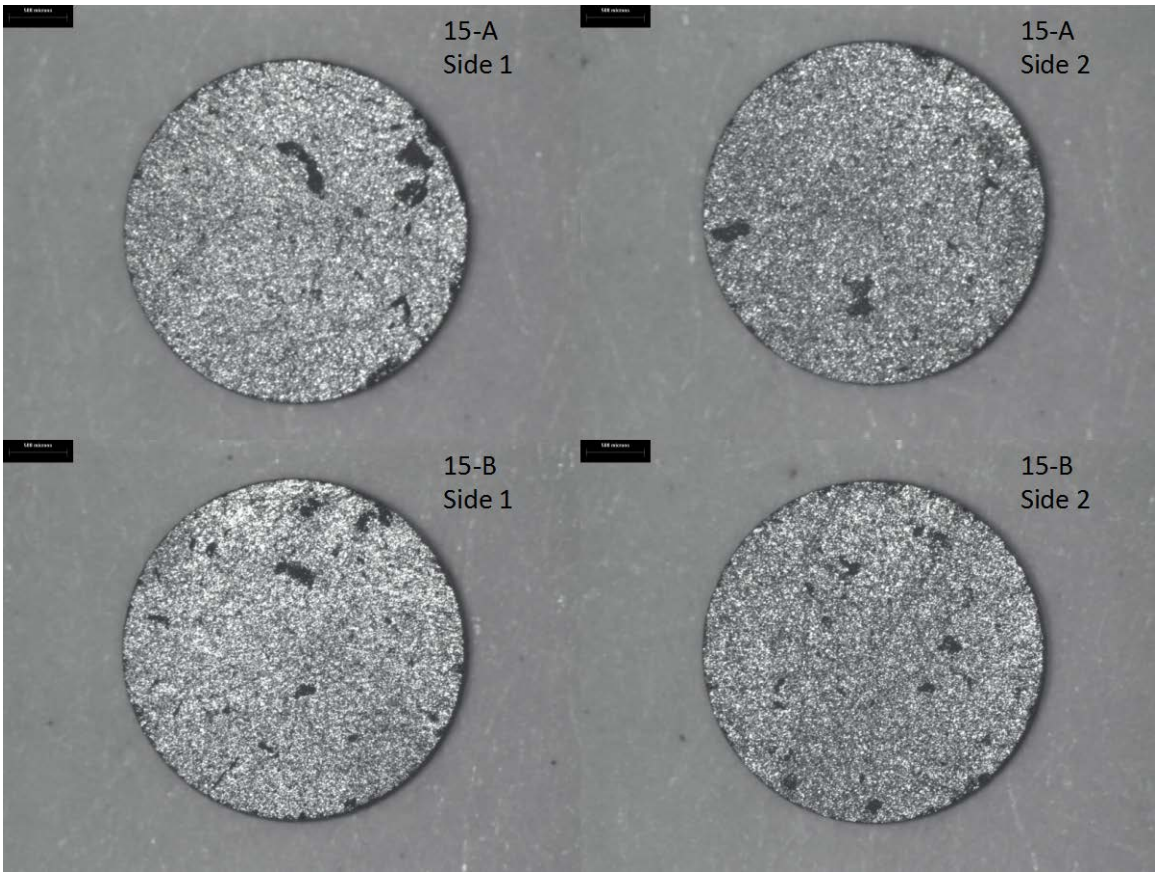

Figure 7: Appearance of natural UO_2 disks cored and polished from pellet 15.

4. SUMMARY

Natural UO_2 disks were fabricated from parent natural U_3O_8 powder as part of a work for other agreement with Texas A&M University. A series of exploratory pellet fabrication conditions were investigated to refine the properties of the bulk pellets used for disk fabrication. The conditions that were explored included the use of binder additives, the starting powder blend (stoichiometry and particle size), pressing pressure, and sintering temperature. The final identified pellet processing conditions resulted in fabrication of natural UO_2 disks that met the specified dimensional tolerances and density criteria.