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From: Robert T. Jubin

Subject: Completion of ORNL AFCI Level 4 Milestone – CETE – OR0915020301, MS# M4502030137, "Issue MDD status letter report," due 9/15/2009

This letter and attached report document completion of the AFCI Level 4 milestone for the CETE - ORNL work package (OR0915020301), "Issue MDD status letter report" (M4502030137), due 15Sep09.

If you have any questions, please contact me at (865) 574-4934 or Gary Bell at (865) 241-4400.

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Milestone Letter Report
Status of Modified Direct Denitration Development

R. J. Vedder
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September 14, 2009

Work Package OR0915020301 – CETE R&D Rev. 2
Milestone No. M4502030137 – Issue MDD status letter report
Due Date: 15Sep09

1. INTRODUCTION

Current flow sheets for processing used nuclear fuels do not produce separated streams of all of the actinides. These aqueous processing streams must be converted into solid forms suitable for recycle (fuel/target fabrication), storage, or disposal, necessitating co-conversion. A process developed at ORNL in the 1980s to make UO_3 suitable as fuel feedstock was studied for preparation of mixed actinide oxides with similarly favorable ceramic properties. The process, Modified Direct Denitration (MDD), uses ammonium nitrate to alter the thermal decomposition behavior of metal nitrates and improve the ceramic properties of the resulting solid oxide. Since plutonium (IV) and neptunium(IV) form compounds similar to uranium with the ammonium ion [$(NH_4)_2Pu(NO_3)_6$, $(NH_4)_2Np(NO_3)_6$], MDD-conversion of these metals was considered to be applicable.

Co-conversion has advantages for making mixed oxides over individual element conversions that are followed by dry mixing of the oxide powders. Issues associated with preparing a mixture from individual oxides include use of additional equipment, dusting associated with feeding and milling, time requirements for milling, blending to obtain a uniform mixture, and inhomogenity at higher plutonium concentrations. These issues can be partially or wholly avoided by using MDD co-conversion in which the mixing of the individual metals occurs in liquid solution; thus, adjusting relative metal concentrations is simpler and the resulting mixed oxide is more uniform than that produced by blending the individual oxides. Utilizing MDD also eliminates the need for mechanical treatment of the powder to obtain the desired ceramic properties, such as surface area and particle size distribution, since these characteristics are acceptable as-produced.

The original MDD development work[1] established that uranium oxide with good ceramic properties could be made.[2] Following the discovery, a more fundamental understanding of the chemistry of the uranium-ammonium double nitrate salt was developed.[3] Later pilot-scale studies[4] produced kilogram quantities of UO_3 using engineering-scale (1 kg/hour), continuously-operated equipment, while establishing the reliability of the process and equipment. The current work was performed in support of the Advanced Fuel Cycle Initiative (AFCI), utilizing glove-box-contained equipment (100 g/hour) to produce UO_3, PuO_2, and mixed oxides of uranium, plutonium, neptunium, and americium from a nitrate solution of those actinides. Then the MDD glove-box system was utilized in the Coupled-End-To-End (CETE) project to convert the U-Pu-Np and uranium product solutions into oxide powders.

As part of the CETE project, a powder characterization laboratory was established in glove-boxes with instruments required for the determination of: (1) surface area by the BET methodology; (2) tap density by using a Quantachrome AutoTap; (3) flow properties by using a Freeman technology powder rheometer; (4) material composition and crystalline structure by using a powder X-ray diffractometer; (5) particle size distribution by using a laser light-scattering analyzer; and (6) imaging of the powders with a stereomicroscope. These instruments can be used to characterize the products and to determine the effects of MDD operating parameters on product powder morphology. Ultimately, the powder characteristics necessary to produce high-density, sintered MOX pellets can be determined.

2. BACKGROUND

Prior MDD development work focused on the preparation of fuel-grade uranium oxide, although some mixed oxides of uranium and surrogates were prepared, as well as a small amount of U/Pu.[1, 5] The MDD process uses a modifer, ammonium nitrate, to avoid the formation of the sticky, mastic phase that is encountered in conventional thermal denitration. The ammonium nitrate forms a double salt with the actinide metals and the double salt is decomposed simultaneously in the rotary kiln reactor. A relatively free-flowing powder product is produced with characteristics required of nuclear fuel pellet feedstock. Sieving through a 20 or 40-mash screen is required to break up product agglomerates and meet the ASTM requirement for oxide fuel[6, 7].

Figure 1 provides a visual comparison of milled UO_3 powder produced by thermal denitration with MDD-produced powder. The comparative powder characteristics are:

	Thermal Denitration	MDD
Surface area, m^2/g	0.1-1.6	6-12
Avg. particle diam of 50 wt%	43	3
Bulk density, g/cm^3	1.6-2.5	0.7-1.2
Sintered pellet density, %TD	~75	>90

Reference 1 provides detailed information on oxide prepared by MDD in the original development work. Reference 2 provides the results of sinterability studies performed with that material in which pellets of greater than 96% of theoretical density (TD) were made.

The chemistry of the ammonium-uranium double salts was studied by Notz and Haas.[3] The properties and decomposition behavior were determined for three double salts: $NH_4UO_2(NO_3)_3$, $(NH_4)_2UO_3(NO_3)_4$ and $(NH_4)_2UO_2(NO_3)_4{\bullet}2H_2O$. Decomposition of the tetranitrate $[NH_4UO_2(NO_3)_4]$ is accomplished in three distinct steps:

- dehydration at 50°C
- decomposition of the tetranitrate salt at 170-270°C
- decomposition of the trinitrate salt at 270-300°C to UO_3, with crystallization at ~500°C

The current work has employed a metal-to-NH_4 mol ratio of 1:2, which is the tetranitrate. Batch denitrations performed with solutions decomposed in a beaker on a hot plate indicate similar transformations for U/Pu mixed nitrate as those described above..

For comparison, co-precipitation methods for making mixed oxide requires control of valence of the elements, pH of the feed solution, precipitant addition, precipitation temperature, and mixing time. The precipitate must be filtered, washed, and then dried. The UO_2- PuO_2 MOX produced has a relatively wide particle size distribution and requires milling, pre-slugging, and granulation to obtain the desired surface area and particle size distribution for pressing and sintering to the required high theoretical density.

3. EQUIPMENT DESCRIPTION AND OPERATION

Figure 2 is a picture of the rotary kiln and the ancillary equipment used in this work as installed inside of a 6-ft. long glovebox. The equipment is standard labware with a specially designed rotary kiln.

The rotary kiln was fabricated by the Mellen Company and has a 304L stainless steel rotating tube of 2" OD, which is heated over 10" of its length by an electrical single-zone furnace capable of reaching 1200 ^{o}C. The tube is sealed at both ends with Teflon/graphite rotary spring seals. The entrance and exit hoods are sealed to their mounting plates with Viton O-rings. The kiln angle of incline is adjusted using the leveling feet threaded rods. The drive is a remotely adjustable DC motor, which can be separated from the rest of the kiln to reduce the overall width to less than 12 inches. This detachability allows the kiln to be moved through a glove box bag-in port in two easily assembled pieces.

The kiln internals consist of the center tube and roller rods, which are shown in Figure 3. Feed solution is pumped though a 1/8" diameter tube inserted into the center tube which terminates near the tube divider. The feed drips out of the center tube through the bottom holes and onto the surface of the rotating pipe, or onto the roller bars. Off-gas (steam and decomposition gases) exit the heated pipe via holes on the top and sides of the center tube, with vacuum being pulled on the center tube from the condensate pot. Solid rods, which are kept from sliding out of the kiln by disks on the center tube, roll freely to dislodge material which might adhere to the wall. The temperature profile along the inside length of the heated tube is measured by three thermocouples in a single sheath inserted from the product end of the center tube.

The off-gas exits the kiln and enters a condenser. The associated condensate pot is vented to a small scrubber with the scrubbing solution being cooled condensate. Some of the NO_x, and essentially all of the entrained oxide, are removed from the off-gas. The scrubber is vented to a knockout pot and then through a filter to a vacuum pump. Adjustment of the vacuum level is manual, with the criterion to maintain the kiln under vacuum, the exact value not being critical.

A run begins with the heat-up of the furnace concurrent with rotation of the heated pipe. The ancillary systems-vacuum, cooling water, off-gas scrubber, etc. are started. This also includes the air purge, which enters the kiln from the product side and serves to keep off-gas away from the product and removes decomposition gases. When the furnace reaches steady-state operating temperature, water addition is started. The purpose is to cool the feed tube sufficiently to prevent feed from decomposing inside the line and plugging it. After a short while, feed addition begins and then the operating conditions (e.g., feed rate, vacuum, temperatures) are monitored. The product is collected in clear polycarbonate bottles which are exchanged as they fill.

4. RESULTS

The initial phase of the current development work was performed using cerium nitrate, which decomposes similarly to plutonium nitrate[9] and also forms a double salt with NH_4. Operating conditions which served as a starting point for later work were developed. The equipment was then installed in a standard 6-ft glovebox.

Initial glovebox runs were made with plutonium nitrate and uranium nitrate solutions to reproduce results from earlier studies. No operational problems were encountered during preparation of plutonium oxide as had been seen in previous MDD work performed by Mailen, et al.[5] Runs were then made using mixed metal nitrate solutions to demonstrate co-conversion capabilities. Early runs used solutions made by mixing nitrate solutions of the individual metals to obtain the following compositions: (a) 90%U/10%Pu, (b) 90.4%U/ 9.1%Pu/ 0.5%Np, and (c) 90.4%U/ 9.0%Pu/ 0.3%Np/ 0.3%Am. The final runs included solutions of mixed nitrates as well as uranyl nitrate from the CETE processing of Dresden used fuel. The large amount of uranium product allowed some examination of the the effects of varied operating conditions.

4.1 GENERAL RESULTS – EQUIPMENT AND OPERATION

The amount of solution fed in a run was sufficient to achieve steady state (constant temperature profile from the middle to the end of the furnace and near-constant powder accumulation rate) for most of the feed time. The steady state product was collected separately from the initial and final material to exit the kiln.

Typical operating conditions as developed after the second metal concentration glovebox uranium run were:

Ammonium to metals mole ratio: 2:1
Counter-current air purge rate: 0.3 cfm
Heated tube rotation speed: ~20 rpm
Furnace controller setpoint: 650 to 675 ^{o}C
Angle of incline: 6-1/2 degrees
Average feed rate: 5 mL/min

Typical internal centerline temperatures are given in Figure 3. The temperature nearest the feed location was observed to fluctuate. A steady drop in this temperature was a good indicator of excessive accumulation of material on the wall of heated pipe.

The throughput was ~100 g of metal per hour with this 2” dia. x 10” heated length rotary kiln. The limiting factor appeared to be conveyance of solids through the area between the retainer disk and heated pipe, and not heat transfer.

No significant operational issues were encountered until one of the parametric studies with the bulk uranium. That run tested the maximum feed rate and resulted in the center tube becoming bent which allowed the roller rods to slide out of the heated zone. Without the roller rods to remove adhering material from the wall of the heated pipe and breakup agglomerates, material accumulated. A large decrease in the feed zone temperature was observed.

4.2 PRODUCT UNIFORMITY

Product uniformity from one of the early co-conversion runs was examined. This quality is normally determined by examining a sintered or irradiated fuel pellet[10] – irradiation improving conversion of the mixture to a solid solution, which aids PuO_2 solubility. Because no fully-sintered pellets have been produced in the current work, this evaluation was performed by sieving the product to three size fractions, sampling, dissolving the sample in refluxed concentrated nitric acid, and analyzing the dissolvent. The dissolutions were performed in a Teflon round-bottom flask surrounded by a heating mantle with the solution agitated by a lab mixer. Concentrated nitric acid heated to the atmospheric reflux temperature was the solvent. The samples ranged from 5 to 13 mg total oxide per ml of solvent. The results of this are given in Table 1.

Table 1. Pre-CETE co-conversion product: weight fractions in dissolvent

MDD run feed composition	Size fraction	U (wt%)	Pu (wt%)	Np (wt%)	Am (wt%)
92.3%U/ 7.15%Pu/ 0.31%Np/ 0.28%Am	-40 +70 mesh	92.5	7.02	0.31	0.17
	-70 +170 mesh	92.5	7.03	0.31	0.16
	-170 mesh	91.0	8.50	0.35	0.15

The -170 mesh fraction comprises a small amount of the total product. For this size fraction, the ratio of plutonium to uranium is greater than in the larger size fractions and also greater than in the feed. A possible explanation is contamination from material from the previous run which had a U:Pu ratio of 10:1 compared to 13:1 for the most recent run. The smallest material would coat the internals of the equipment more than any of the large particles and would take a longer time to exit the kiln. Since the kiln was not flushed at the completion of a run, the fines would not exit the kiln until the next batch of feed was run through the equipment, in this case, resulting in a higher Pu concentration in the smallest size fraction product.

4.3 MATERIAL BALANCE

Extended runs in which 150+ g of uranium averaged losses of <0.3% of feed while losses for shorter duration runs averaged 1%. Loss of product is caused by entrainment of some of the finer oxide in the off-gas and appears to occur more at the beginning of a run, thus runs of longer duration resulted in lower losses. Additionally, the most concentrated feed had a significantly lower loss than the most dilute feed: 0.02 vs. 0.5%. This material ends up in the condensate either with condensed off-gas or with condensate used for the scrubber.

A run was made with feed prepared from the condensate collected from all previous co-conversion runs. This recycle feed was prepared by ammonium hydroxide precipitation followed by decanting the solids, which were then fed to the kiln as a slurry. No operational difficulties were encountered and the decantate was sufficiently low in activity for disposal without any further processing.

Approximately one-half of the nitrate in the feed was recovered in the condensate using a scrubber of 3” dia. x 4” long with Teflon Raschig rings and cooled condensate as the scrubbant. Nitric acid in the condensate can be reacted with ammonium hydroxide to make ammonium nitrate for subsequent feed makeup.

A small amount of technicium followed the uranium into the U/Pu/Np stream from CETE. Analysis was performed to determine whether the Tc was converted to oxide form with the uranium, plutonium and neptunium, or volatilized into the condensate. Results are shown in Table 2.

Table 2. ICP-MS Tc results for mixed nitrate and mixed oxide MDD product

Material sampled	**Tc concentration**	**Tc concentration: ratio to total U, ug/g U**
dissolved product	785 ug/g oxide	1170
mixed nitrate initial concentrate	300 ug/ml	1822
mixed nitrate final concentrate- acid reduced and filtered	582 ug/ml	1925

4.4 X-RAY DIFFRACTION ANALYSIS OF PRODUCTS

X-ray diffraction analysis of the uranium products indicates that the UO_3 product is amorphous while the reduced product has a crystalline structure characteristic of CaF_2. Complete conversion of UO_3 to UO_2 under conditions of 4 hours at either 550 or 600 ^{o}C in flowing 4% hydrogen/argon was confirmed. The Rietveld refined lattice parameter for the CETE mixed oxide product was determined to be 5.4551.

4.5 PRODUCT FLOW PROPERTIES

An instrument which is used to study the fundamental and bulk properties of powders was installed in a glovebox as part of the CETE project. The instrument is a Freeman Technology FT4 Powder Rheometer. The instrument allows measurement of powder properties, such as:

- Flow energy when consolidated and unconsolidated and aerated and un-aerated
- Cohesiveness
- Permeability
- Compressibility
- Sensitivity to flow
- Stability

Test results are utilized to design powder handling equipment and evaluate powders in operations such as pelletization/tablet making. The bulk properties of powders depend upon properties of the powder such as surface area, particle size distribution, shape, and texture – the same properties that are important in fuel pellet fabrication.

Fresh material should be used in each test because the mechanical nature of the tests breaks apart agglomerates; however, the product sieved to -40 mesh was less affected by testing than the product directly from the kiln. Results for the sieved materials tested are given in Table 3.

Conclusions from the test results on sieved MDD products:

- Average sensitivity to aeration;
- In the lower part of the stable region with slight de-agglomeration resulting from being made to flow;
- Average flow rate sensitivity;
- Flow functions were between 10 and 4, which characterizes the powders as easy-flowing; and
- Typical to readily aerates, which is a positive property for powder to be pressed into pellets.

Table 3. Results of measurements on sieved MDD CETE products using a Freeman Technology FT-4 powder rheometer

MDD CETE Product	**Aeration ratio**	**Aeration energy, mJ**	**Compressibility**	**Stability index**	**Flow rate index**	**De-aeration**	**ff_c[1]**	**AIF(SS)[2]**	**Wall friction angle- 304 SS/ 0.05um finish**	**Wall friction angle- 304SS/ 1.2um finish**
Clean cut bulk U – (standard operating conditions)	5.5	30	6.8% @ 1kPa 15.2% @14.9 kPa	0.7	1.5	Readily de-aerates	5.1	40.3°	32°	37°
Higher NH_4:U ratio (3 mol:1 mol) bulk U	4.7	51	8.8% @ 1kPa 21.0 @ 14.9kPa	0.8	2.3	Typical	6.6	40.4°	26°	34°
Higher acid bulk U	7.1	56	8.8 @ 1kPa 18.4 @ 14.9 kPa	0.8	1.4	Readily de-aerates	6.9	40.6°	28°	36°
CETE mixed oxide	7.4	60	8.8 @ 1kPa 20.0 @ 14.9 kPa	0.6	2.2	Readily de-aerates	4.4	37°	29°	36°

[1] Flow function or flowability: major principle stress ÷ unconfined yield strength

[2] Angle of internal friction at steady-state flow

4.6 PELLETS PRESSED WITH MDD-PRODUCED MIXED OXIDE

Pellets were pressed with pre-CETE U/Pu/Np/Am (see Table 1) and the CETE U/Pu/N mixed oxides. Table 4 gives the significant constituents in the CETE MDD feed solution. Characterization of the CETE U/Pu/Np mixed oxide, directly from the MDD process and reduc given in Table 5. The pressing equipment is inside a glovebox and utilizes a drill bushing of 0.2 inch ID with a removable plug in the bottom into which the powder is poured. A piston is then inserted into the drill bushing and this placed into a holder (with a blind hole for the pressing an through hole for the pellet removal). The piston is forced vertically down onto the powder.

Table 4. CETE mixed nitrate feed to the MDD process

Total Pu, g By alpha/PHA	Total U, g By ICP-MS	Total Tc, g By ICP-MS	Total Np, g By ICP-MS	U:Pu ratio, g
13.2	164	0.31	0.72	12.4

Table 5. CETE mixed oxide properties before and after reduction

CETE MDD product			CETE reduced oxide		
Tap density, g/mL	BET specific surface area[1], m^2/g	Particle size (dia.), µm	Tap density, g/mL	BET specific surface area[1], m^2/g	Particl (dia.),
1.3	10.6	Mean 5.51 50% passing 3.26	1.4	7.5	Mean 50% p 4.61

[1] After calcination to 300 °C

The MDD product was passed through a 40 mesh sieve by shaking the sieve containing ½” stainless steel ball bearings and the powder. The reduction of the UO_3 constituent to UO_2 was d flowing 4% H_2/Ar with the powder contained in a quartz tube heated to either 550 °C or 600 °C. slugging/granulation was performed prior to pressing. Stearic acid was mixed with the reduced to ~0.3 wt%. The pressing pressure was ~43,200 psi. Pellets were pressed at length to diameter up to 1.6 and average density of 50% of theoretical. The green pellets are shown in Figure 4 wi those made from the CETE mixed oxide on the right.

Two of the pellets prepared from the CETE mixed oxide were heated in a glovebox-contain furnace capable of reaching ~1340 °C. This is significantly less than the typical temperature at mixed oxide pellets are sintered. The pellets reached 86.3% and 88.7% of theoretical density.

4.7 CONVERSION OF CETE URANIUM PRODUCT

The 2.8 kg of U in the uranyl nitrate product solution obtained from processing Dresde nuclear fuel was converted to UO_3 by MDD. Some product solution was bottled and transferred directly to a glove box rather than concentrating using hot cell evaporators: this material is refe as the “clean cut”. Other than concentration of a dilute flush solution, no other processing was performed out-of-cell prior to addition of ammonium nitrate and conversion. The processing rat ~100 g U/h, and the standard operating conditions given in Section 4.1 were used.

The large amount of uranium provided an opportunity to make products under varying characteristics of feed and operation. Tables 6 and 7 provide run conditions and characterizatio the products directly from the rotary kiln.

Table 6. Run conditions and products made in conversion of the CETE bulk uranium

Run description	**Feed conc., g U/L**	**NH4:U ratio mol/mol**	**Steady state temperature profile[1], °C**	**Weight loss on calcination[2]**	**Tap density, g/ml**
Standard conditions with *Clean cut* feed	214	2	240/545/505	4.5%	1.01
Lower temperature (625°C)	288	2	150/445/430	6.6%	0.75
Dilute feed (180g U/L)	180	2	195/530/490	7.2%	0.81
Higher ratio of NH_4 to U	280	3	295/555/495	1.1%	1.18
Higher feed rate	300	2	135/455/480	3.3%	0.71
Higher feed acidity (~2N H^+)[3]	---	2	150/480/480	4.7%	---
Concentrated feed	330	2	130/430/470	4.4%	0.77

[1] Internal heated pipe centerline temperature: feed end/middle/product end

[2] Samples heated in air to 300°C for 2 hours

[3] Other solutions were ~0.3 N HNO_3

Table 7. Characterization of products made in conversion of the CETE bulk uranium

	MDD product		**Reduced MDD product[1]**	
Run description	**BET specific surface area[1], m^2/g**	**Particle size (dia.), µm**	**BET specific surface area[1], m^2/g**	**Particle size (dia.), µm**
Standard conditions with *Clean cut* feed	7.4	Mean 3.95 50% passing 2.54	4.1	Mean 3.12 50% passing 2.34
Lower temperature (625°C)	7.6	Mean 8.94 50% passing 3.68	6.6	Mean 4.67 50% passing 2.73
Higher feed acidity (~2N H^+)	7.3	Mean 4.29 50% passing 3.87	6.7	Mean 3.43 50% passing 1.87

[1]Reduction at 550 °C in 4% H_2/Ar confirmed by XRD

4.8 CETE MDD-PRODUCED HYDRATED URANIUM OXIDE

The orange-colored UO_3 MDD product turns orange-yellow or yellow (see Figure 5) in air. XRD results identify the yellow material as $UO_3 \cdot 0.8\ H_2O$; which concurs with the weight loss determination. From XRD analysis, the orange-colored UO_3 is amorphous and the hydrate is crystalline. The hydrated UO_3 reverts back to the original orange color when heated to ~250 °C; it's a fairly stable hydrate. However, the surface area is permanently reduced.

The surface area of one of the CETE UO_3 products (from the high acid run) in the following forms were measured: material from the product storage bottle calcined at 300 °C to remove residual volatiles; the product reduced at 600 °C in 4% H_2; the hydrate reduced under the same conditions; and the hydrate first converted back to UO_2 by heating to 300 °C and then reduced under the same conditions. The results are presented in Table 8.

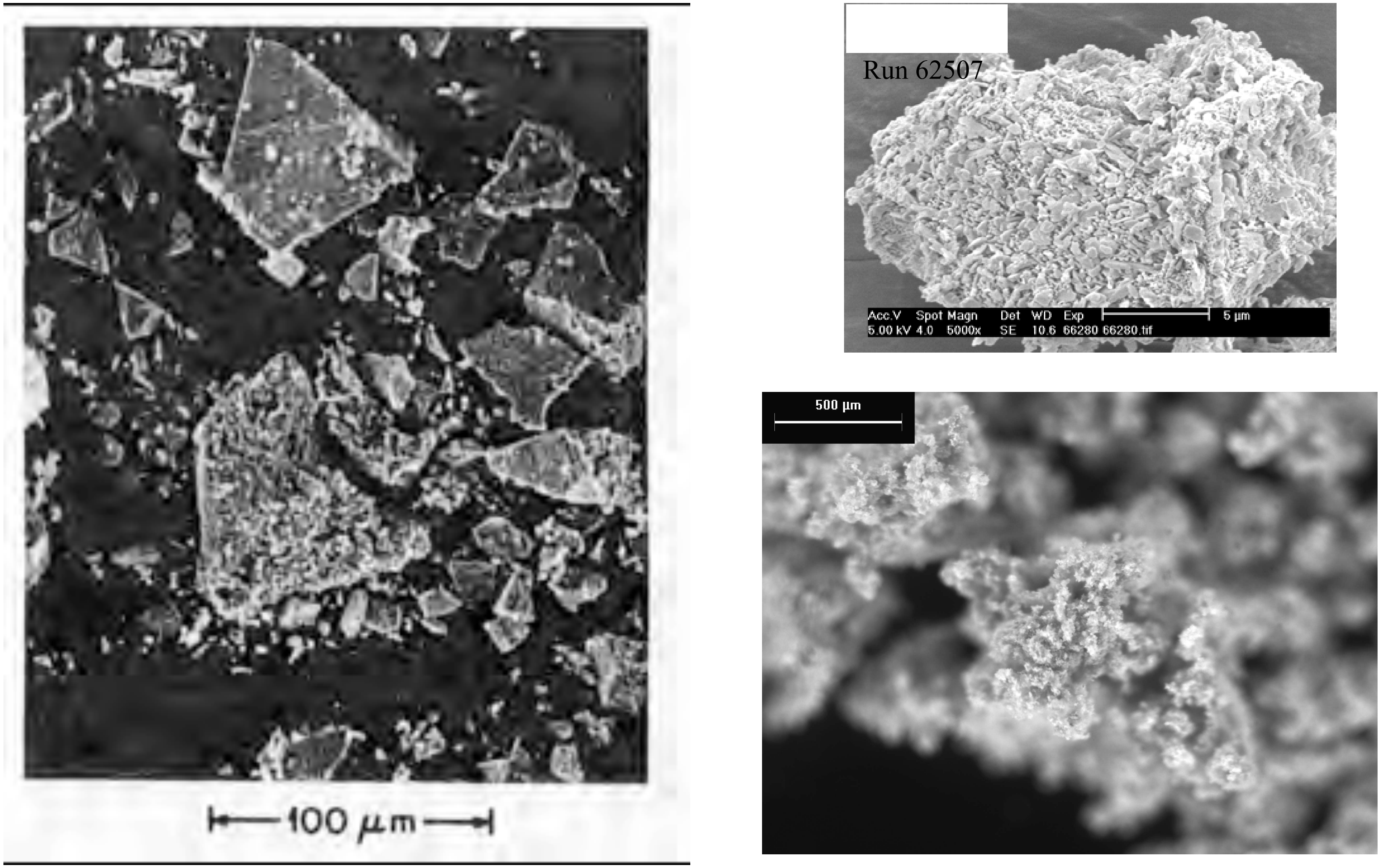
Table 8. Surface area measurements of an MDD uranium oxide to examine the effect of hydration

Material	B.E.T. specific surface area, m^2/g
Product from MDD calcined to 300 ^oC to remove residual volatiles	7.3
Product from MDD run reduced to UO_2 at 600 ^oC in 4% H_2	6.9
Hydrated product from MDD run directly reduced to UO_2 at 600 ^oC in 4% H_2	2.9
Hydrated product from MDD run first calcined at 300 oC and then reduced to UO_2 at 600 ^oC in 4% H_2	3.7

As expected, surface area decreased slightly when the UO_3 was reduced. Also observed was a much greater decrease in surface area when the hydrate of the same material compared to the non-hydrated UO_3 is reduced. When the hydrate is directly reduced, the specific surface area decreased by ~60% while if the hydrate is first calcined the decrease was somewhat smaller. Measurements on pellets pressed and sintered from depleted uranium powders found that pellets made from the reduced hydrated trioxide sinter to >90% theoretical density (correspondence from Jim Kiggans).

5. CONCLUSION

Nitrate solutions of mixed actinides of U, Pu, Np, and Am and uranyl nitrate solutions have been successfully converted to mixed oxides or uranium oxide by Modified Direct Denitration. Feed solutions initially originated from mixing individual nitrates and later from the CETE processing of Dresden used nuclear fuel. The development effort used equipment contained in a glovebox with a process throughput of ~100 g metal/hour. The CETE products were measured for standard characteristics of nuclear fuel feedstock and found to be acceptable, but somewhat low in density. A test for uniformity of a reduced mixed oxide found the powder to be uniform across varying size fractions, which should be beneficial for making uniform sintered fuel pellets. Testing with a powder rheometer found the powder flow and de-aeration properties to be good. Varying the operating conditions did not negatively impact the product properties or operation, indicating a forgiving process. Pellets were successfully pressed to L/d of up to 1.6 with mixed oxides powders but not sintered due to the unavailability of a furnace capable of reaching the required temperature. The pellets reached ~86% of TD in heating to ~1340^{o}C, and thus should reach the desired >90% TD when sintered at 1600 to 1800^{o}C.



a) Milled UO_3 prepared by traditional direct denitration (taken from Haas, Ref. 1).

b) MDD-produced UO_3 directly from the kiln, before sieving (photos provided by John Hunn).

Fig. 1. Comparison of powders produced using traditional direct thermal denitration and modified thermal denitration processes.

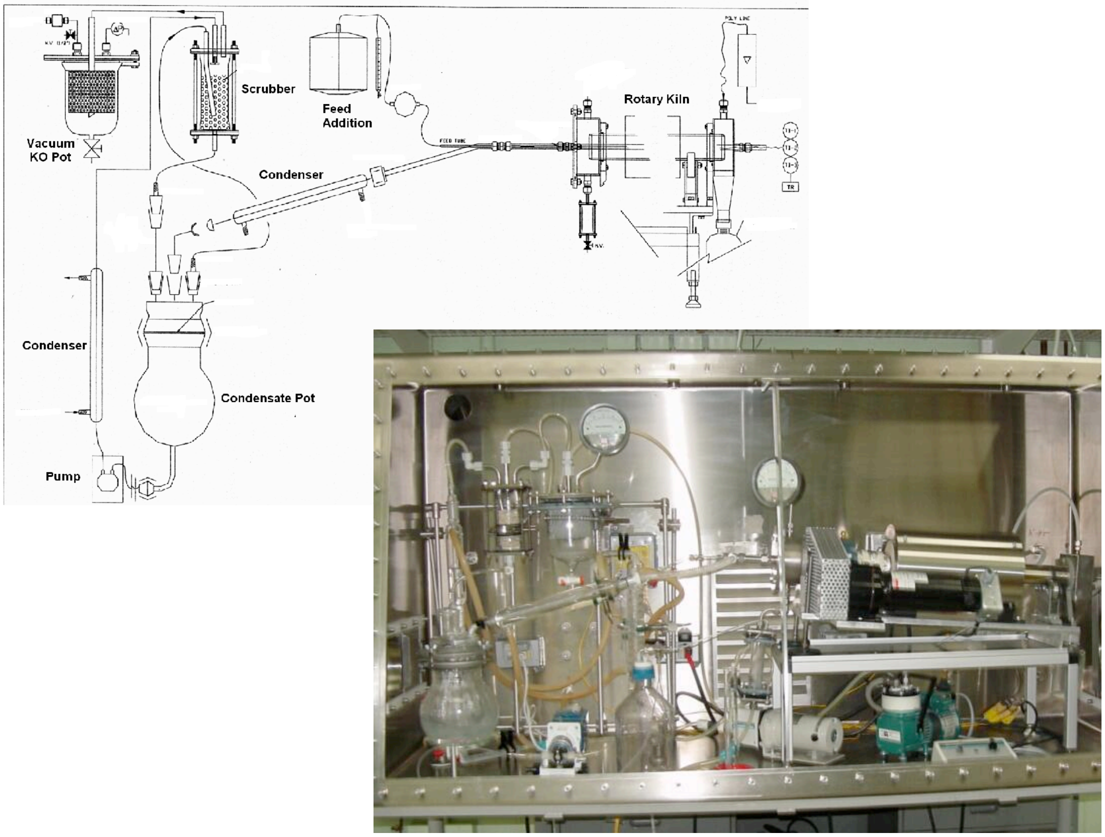


Fig. 2. Glovebox-contained MDD process equipment.

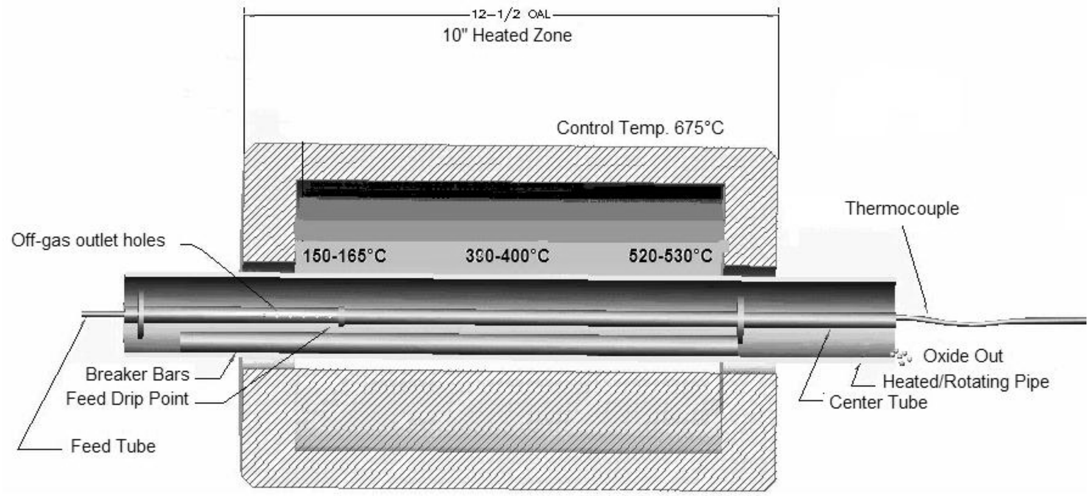


Fig.3. Section through furnace showing internals and typical internal centerline tempera profile.

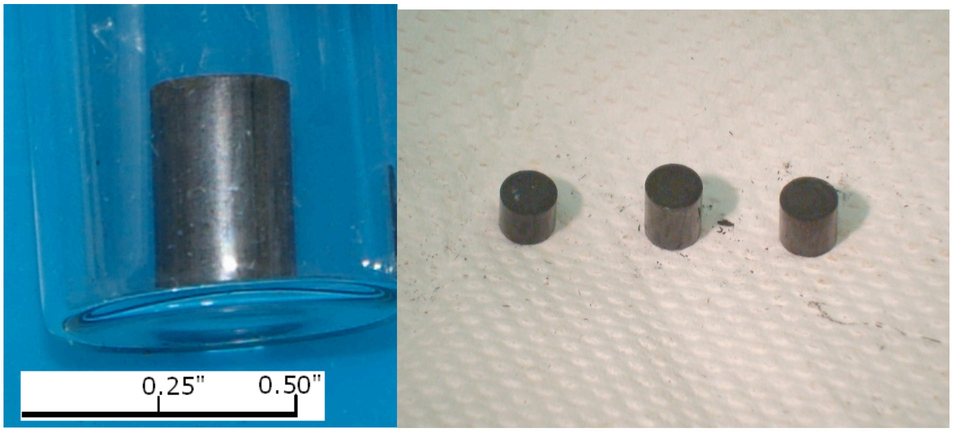


Fig.4. Green pellets pressed with reduced U/Pu/Np/Am MDD product (left) and CETE U/Pu/Np.

Fig.5. Hydrated MDD-produced UO_3 on left; UO_3 from kiln on right.

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