LA-UR- 98-4981

Approved for public release; distribution is unlimited.

Title: Mixed Oxide Fuel Fabrication in Support of Surplus Plutonium Disposition

Author(s): Angelique Neuman
M.S. E502
LANL/NMT-9
Los Alamos NM 87545

Submitted to: Dr. Cordelia Ontiveros
SHPE National Technical and Career Conference '99
Los Angeles, CA
Feb 3-6, 1999

UNCLASSIFIED NOT UCNI

NOV 09 1998
FOCUS date

S-7

Los Alamos
NATIONAL LABORATORY

DISCLAIMER

MIXED OXIDE FUEL FABRICATION IN SUPPORT OF SURPLUS PLUTONIUM DISPOSITION

Angelique Neuman
Technical Staff Member, NMT-9
Los Alamos National Laboratory
Mail Stop E502
Los Alamos, NM 87545
Phone number: (505) 665-9890
Fax number: (505) 665-4775
e-mail: neuman@lanl.gov

ABSTRACT

There is a present need to reduce the nuclear stockpile and thus reduce the global nuclear danger. The method discussed here is by means of fabricating Mixed Oxide (MOX) Fuel. The PuO_2 in this MOX fuel is obtained by converting weapons grade Pu into PuO_2 via an hydride/nitride/oxidation method. The PuO_2 is then thermally treated to remove gallium, an innate impurity of weapons grade plutonium. This thermally treated PuO_2 is then mixed with UO_2 into a 30-70 wt% primary blend of PuO_2-UO_2, respectively. Part of this master blend is then reduced to a secondary blend of 5 wt% PuO_2. This secondary blend is used to make sintered pellets. The process of pellet fabrication involves, slugging and granulation of the powder, pellet pressing, additive removal, sintering and centerless grinding. The final sintered pellets are inspected and then loaded into pins.

INTRODUCTION

This document summarizes the efforts performed by the Nuclear Fuels Technologies team at Los Alamos National Laboratory (LANL) in support of the Fissile Materials Disposition (FMD) Program under the heading of Feed Materials Baseline Development. It has been determined through previous efforts that development work is necessary when new feed materials are introduced into an established fabrication process (See Figure 1 for a process diagram for the established fabrication process). The FMD Program made the decision to select a new UO_2 source for use in fuel fabrication R&D activities at LANL. The new source of UO_2 is powder derived from the Ammonium Uranyl Carbonate (AUC) process received from Asea Brown Boveri (ABB) in Sweden. Fuel fabrication activities to date have used Cameco UO_2 obtained from Canada. The properties of Cameco UO_2 differ significantly from those of AUC-derived UO_2. Although the AUC-derived UO_2 material has previously been used to fabricate the majority of the European reactor-grade mixed oxide (MOX) fuel, it is important to establish how it will interact with PuO_2 from weapons-grade plutonium in terms of fuel fabricability. Furthermore, plutonium oxide feed materials can be quite different with respect to powder

morphology and impurity levels depending on the conversion process and processing parameters, and it is important to quantify the effects these differences may have on the fuel fabrication process. There were two main tasks included in this effort [1]:

1. Develop baseline MOX fuel fabrication processing parameters for the AUC-derived source of UO_2 feed material using prototypic PuO_2 powders.
2. Fabricate MOX fuel using the baseline fabrication processing parameters, the new source of UO_2 feed material, and an alternative source of PuO_2 feed material.

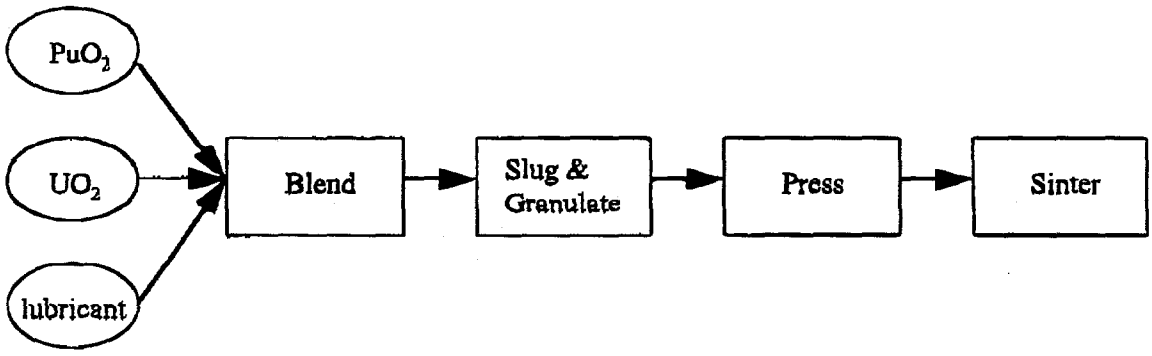

Figure 1. Process diagram for the fabrication process.

In the Test Plan [2] written for this project, UO_2 development activities were described that would establish a process by which MOX fuel could be successfully fabricated using the AUC-derived material. Samples were to be taken of the powder and characterized for particle size, surface area, O/M ratio, and powder morphology. Sintering tests were to be performed on UO_2 powder alone and MOX simultaneously to utilize resources most effectively. Sintered pellets were then to be characterized for such properties as density, microstructure, and shrinkage. Further sintering tests would be continued until the sintered pellets met a predetermined standard. The immediate goal of these efforts was to obtain sufficient processing information to allow other FMD fabrication activities to begin. Completion of the remainder of the UO_2 development work would then be performed in parallel with the other activities, as schedule and resources permitted.

The second task under the Baseline Development effort is the alternative PuO_2 fabrication task. The Test Plan specified that the UO_2 development activities involving MOX powders described above were to be performed using PuO_2 powder created by Lawrence Livermore National Laboratory (LLNL) by the 3-step (hydride-nitride-oxidation) method. For the alternative PuO_2 fabrication task, two different sources of PuO_2 powder were identified. The two sources of PuO_2 material for this task were identified as LLNL 2-step (hydride-oxidation) and an aqueously-derived PuO_2.

DISCUSSION AND RESULTS

The first experiment, labeled T1, was performed to determine the powder's flow properties and pellet shrinkage information and was performed by pressing the as-received UO_2

powder without pre-compaction or granulation. The order of the pellets was inadvertently mixed up after sintering of the fuel, so only flow properties were determined from this test and were found to be good. No immersion density data was obtained, however, due to the pellet mix-up.

The second experiment, labeled T2, was performed to obtain the shrinkage data not available from the first experiment. Again the powder was pressed as-received. sintered density obtained was found to be within the predetermined specification of 95 ± 1% of theoretical density. The density of one pellet (pressed at 52,000 psi) actually exceeded the specification at 96.8% of theoretical density. The shrinkage exhibited a nearly linear behavior, decreasing slightly with increasing pressing pressure and ranging from ~19-23%.

The third experiment, labeled T3, was performed to determine the effects of the addition of a die lubricant. Zinc stearate (0.2 wt%) was added to the UO_2 powder before pressing. The remaining processing parameters were held constant. The pellets pressed at less than ~50,000 psi were within the predetermined specification of 95 ± 1% of theoretical density.

The fourth experiment, labeled T4, was performed to determine the effects of making a primary blend with a Turbula® mill, but only using UO_2. The master blend consisted of 95 wt% UO_2 and 5 wt% UO_2 (as a substitute for PuO_2) to simulate a MOX fuel master blend. The powder was blended with the Turbula® for 15 minutes and was pressed as-blended without any additives. Only the pellets pressed at greater than ~52,000 psi were within the predetermined specification of 95 ± 1% of theoretical density. The pellets exhibited a rough, "blistered" diametral surface and some pellets were "hourglassed". This is likely due to insufficient die lubrication and could be an indication of a rough die surface.

The fifth experiment, labeled T5, was the first MOX batch made using the AUC-derived UO_2 and PuO_2 powders. The 3-step PuO_2 powder processed by LLNL was used for this experiment. The experiment was performed exactly as the fourth (T4), except 5 wt% PuO_2 powder was used to create a true MOX blend. The MOX powder showed good flow properties and pressed without problems, indicating that the addition of the PuO_2 had no detrimental effect on the blend, however none of the pellets were within the predetermined specification of 95 ± 1% of theoretical density. All the pellet densities were between 91-93% of theoretical density. Again, the pellets exhibited a rough, "blistered" diametral surface and some pellets were "hourglassed". Figure 2 shows a photograph of MOX pellets from test T5 and Table 1 indicates the processing parameters used for the five above-mentioned experiments. All the batches were sintered at 1750 °C for 7hrs and none underwent the slugging (pre-compaction) and granulation step.

Figure 2. MOX fuel pellets after sintering for 7 hours at 1750 C in an Ar/6% H_2 atmosphere.

Table I. Baseline Development Experimental Variables and Processing Parameters

Experiment	PuO_2 (g)	UO_2 (g)	Lubricant	Blending	Pressing (x 1000 psi)
T1	none	200	none	none	28.8 - 86.5
T2	none	200	none	none	28.8 - 57.7
T3	none	199.6	0.4g zinc stearate	Turbula 5 min.	40.4 - 69.2
T4	none	190+10	none	Turbula 15 min.	40.2 - 54.8
T5	10g 3-step	190	none	Turbula 15 min.	40.4 - 57.7

The second task studied was the alternative PuO_2 fabrication task. Two additional sources of PuO_2 powder were used for this task: a 2-step hydride-oxidation material created by LLNL and an aqueously-derived source processed at LANL via oxalate precipitation. Although it is not currently anticipated that the 2-step material will be selected as the method for PuO_2 conversion, it was hoped that the material would behave similar to the 3-step material. This would allow its use as an additional source of feed for future experiments, since there is little inventory of 3-step left available. MOX pellets were fabricated using these two sources and the same parameters used for experiment T5. For each batch, the AUC powder was blended with 5 wt% PuO_2 (with no additives) in a Turbula® mill for 15 minutes. Table II indicates the results of these batches. Batch number T7 refers to the pellets made with the 2-step material, and batch number AT1 refers to the pellets made with the aqueously-derived source.

The intention of this activity was to compare directly the results to those obtained using the 3-step PuO_2 (experiment T5). None of these sintered densities for the 3-step and 2-step batches meet the 95 ±1% theoretical density specification. The aqueous batch, however, met the density specification at ~52,000 and 55,000 psi pressing pressures.

Since so few of the pellets actually met the predetermined specification, new batches using the three sources of PuO_2 powder were fabricated in an attempt to increase the sintered density results. The new batches were designated as T9 for the 3-step material, T10 for the 2-step material, and AT3 for the aqueously-derived material. A single pressing pressure of 52,000 psi was used for all pellets in all batches, as that pressure created higher quality pellets based on the initial batches. The main difference between the original and new batches is the addition of zinc stearate to the new batches. Table II provides a summary of the green and sintered density and shrinkage data from the original batches and compares that summary to the results obtained from the new batches. The new 3-step batch had approximately the same green density as the original 3-step batch, but lower sintered densities and shrinkage values. The same trends were seen with the 2-step and aqueously-derived batches as well. Overall, the aqueous batches have the highest sintered densities, although they are still for the most part below the desired specification. The addition of the zinc stearate, therefore, did not have the desired effect of increasing the sintered densities.

Table II. Comparison of Original and New PuO_2 Variability Study Results.

PuO_2 Source	Parameter	Batch	Original Values (varying pressures)	Batch	New Values (52,000 psi)
3-step	Green density (% of TD)	T5	45.9-49.7	T9	48.3, 48.8
	Sintered density (% of TD)		91.4-93.9		93.1, 93.3
	Shrinkage (%)		18.8-21.4		19.5, 19.0
2-step	Green density (% of TD)	T7	46.1-50.1	T10	48.9, 49.0
	Sintered density (% of TD)		90.9-93.2		92.4, 92.6
	Shrinkage (%)		18.0-20.6		19.6, 19.3
Aqueous	Green density (% of TD)	AT1	47.6-50.6	AT3	48.7, 49.0
	Sintered density (% of TD)		93.8-96.2		94.6, 94.1
	Shrinkage (%)		18.5-19.9		19.5, 19.0

SUMMARY

The experiments performed to date have completed much of the development work needed to perform other FMD activities, including providing shrinkage data to determine appropriate punch and die sizes for irradiation test fuel fabrication. Samples were taken of the as-received powder for characterization of particle size and surface area analyses, but are not available at this time. O/M analysis capabilities are currently unavailable at LANL, and the morphology data as well as the microstructure results have not been completed. Sinterability tests were performed with both UO_2 and MOX powders, and the sintered pellets were characterized for density and shrinkage.

For the PuO_2 variability studies, two different sources of PuO_2 feed material were identified: LLNL 2-step and aqueously-derived. Pellets were fabricated, sintered, and characterized for density and shrinkage. The results were directly compared to those obtained in the baseline development efforts.

Overall, from these experiments, the AUC powder was found to have good flow properties. Pellets were fabricated to almost 94% of theoretical density, just outside of the required specification. Future experiments will be conducted to more fully develop LANL's ability to make quality MOX fuel using the AUC-derived powder. These experiments will be geared towards obtaining specification densities and examining other variables and parameters. Further PuO_2 variability experiments will also be performed as new sources of PuO_2 powders become available such as direct metal oxidation.

REFERENCES

1 S.L. Eaton, A.D. Neuman, J.G. Teague, S.A. Talachy, K.Chidester, "Nuclear Fuels Technologies Fiscal Year 1998 Fuel Fabrication Process Development Feed Materials Baseline Development Summary Report". Los Alamos National Laboratory, August 28, 1998.

2 S.L. Eaton, et al., "Nuclear Fuels Technologies Fiscal Year 1998 Fuel Fabrication Development Feed Materials Baseline Development Plan," letter report from D. Alberstein to J. Johnson (DOE-MD), NMD-98-003, October 31, 1997.