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EXPERIENCE MAKING MIXED OXIDE FUEL WITH PLUTONIUM FROM DISMANTLED WEAPONS

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ABSTRACT
Mixed depleted UO_2 and PuO_2 (MOX) pellets prototypic of fuel proposed for use in commercial power reactors were made with plutonium recovered from dismantled weapons. We characterized plutonium dioxide powders that were produced at the Los Alamos and Lawrence Livermore National Laboratories (LANL and LLNL) using various methods to recover the plutonium from weapons parts and to convert it to oxide. The gallium content of the PuO_2 prepared at LANL was the same as in the weapon alloy while the content of that prepared at LLNL was less. The MOX was prepared with a five weight percent plutonium content. We tested various MOX powders milling methods to improve homogeneity and found vibratory milling superior to ball milling. The sintering behavior of pellets made with the PuO_2 from the two laboratories was similar. We evaluated the effects of gallium and of erbium and gadolinium, that are added to the MOX fuel as depletable neutron absorbers, on the pellet fabrication process and on the sintered pellets. The gallium content of the sintered pellets was <10 ppm, suggesting that the gallium will not be an issue in the reactor, but that it will be an issue in the operation of the fuel fabrication processing equipment unless it is removed from the PuO_2 before it is blended with the UO_2.

INTRODUCTION
In June 1994 the Department of Energy (DOE) issued a Notice of Intent to prepare a Programmatic Environmental Impact Statement (PEIS) for the storage of all weapons-usable fissile materials and for the disposition of those materials the President has declared surplus to national defense needs. Among the surplus plutonium disposition alternatives identified for evaluation in the PEIS and for consideration in the Record of Decision (ROD), scheduled for September 1996, are five MOX fuel based reactor alternatives. The DOE's Office of Fissile Materials Disposition funded LANL, beginning in fiscal year 1995, to assist with the preparation of both the PEIS and input to the ROD for the reactor alternatives. Several key issues unique to the use of plutonium from surplus weapons in MOX fuel for commercial power reactors were identified. These issues included:

- how the methods of separating the plutonium from the other weapon materials and of converting it to PuO_2 would affect the fuel fabrication process,
- how the gallium in the weapons plutonium and remaining after the conversion process would affect the fuel fabrication process, and its behavior in the finished fuel pellet, and
- how the addition of depletable neutron absorbers such as gadolinium and erbium oxides to the MOX fuel would affect the fabrication process and the properties of the finished fuel pellets.

New issues were raised as the work progressed. One such issue concerned how rejected sintered MOX pellets could be recycled in the fuel fabrication process. Another involved determining what the effects would be on the suitability of the PuO_2 powder as feed for MOX fuel fabrication when it was heated to over 1000°C to stabilize it for long-term storage. Yet another concerned the possibility of removing gallium from the PuO_2 by a thermal process without adversely affecting the quality of the powder as a fuel feed. The final issue dealt with elimination of the binder from the fabrication process.

METHODOLOGY

Plutonium dioxide produced from a dismantled weapon by the hydride/oxidation process at LANL was obtained, as were two different PuO_2 powders produced from a weapon by the HYDOX process at LLNL. Portions of the LANL oxide were passed through a vibratory mill several times, or heated in flowing Ar-6%H_2 at 400°C and 1000°C to determine the effects on size reduction and Ga content. Samples of each material were submitted for gallium content, particle size, surface area, and loss on ignition analyses.

A test plan was developed that included seven experiments designed to produce empirical data for each of the key MOX fuel issues. The first experiment involved demonstrating the fabrication of MOX fuel pellets using plutonium obtained from a dismantled weapon and converted to PuO_2 by the hydride/oxidation process developed at LANL. This experiment also utilized six sintering time and temperature combinations to evaluate the relationship between sintering variables and the gallium remaining in sintered fuel pellets. The second experiment evaluated an alternative milling/mixing method using a vibratory mill for blending the PuO_2, UO_2, and depletable neutron absorbers to obtain acceptable homogeneity. The third and fourth experiments prepared batches of MOX containing gadolinium oxide and erbium oxide, respectively, and characterized the processing behavior and properties of the finished products. The fifth and sixth tests included fabricating MOX fuel pellets using two PuO_2 powders prepared from a weapon at LLNL by the HYDOX process at different temperatures. The seventh experiment would evaluate another type of vibratory mill for oxide blending.

Each of the furnished plutonium oxide powders was used to prepare batches of MOX by blending approximately 5wt% PuO_2 with 95wt% depleted UO_2 and 0.2wt% stearic acid as a lubricant and 0.2wt% polyethylene glycol as a binder. These batches were mixed by ball milling them with tungsten carbide media for 16h or passing them through a vibratory mill containing steel balls four times. The MOX batches were then slugged, granulated, and pressed into pellets about 0.37in diameter by 0.40in long using a uniaxial hydraulic press. Subbatches of ten pellets each were heated to 450°C in flowing argon in a retort furnace to remove the organic additives. These pellets were then sintered in flowing Ar-6%H_2 at temperatures ranging between 1500°C and 1700°C and times ranging between 4h and 16h. The reference sintering conditions used when other processing variables were being studied were 4h at 1600°C. A control batch of UO_2 pellets was fabricated. Sintered MOX pellets were also recycled by crushing and milling them and forming pellets with the resulting powder without the use of a binder. During the fabrication samples were taken after the mixing, the additive removal, and the sintering process steps and submitted for gallium analysis. Following sintering the pellet dimensions were measured and shrinkage and density were calculated. Samples were also submitted for ICP, O/M, Pu and U content, microstructural, homogeneity, SEM. porosity, and microprobe analyses.

RESULTS

The results of the tests and analyses performed on the three PuO_2 powers are presented in Table I. These results show the gallium content of the oxide produced at LANL is the same as it was in the metal before the conversion to oxide while the gallium content of the oxides from LLNL are much lower than what was in the plutonium metal before the conversion process. The PuO_2 produced at LANL has a mean spherical equivalent particle size less than 15μ that can be further reduced by several passes through a vibratory mill. Heat treating the LANL-produced oxide in flowing Ar-6%H_2 had little effect on the particle size but heating to 1000°C did reduce the surface area.

Table 1. Plutonium Dioxide Processing Variables and Results

Powder Identification	Process Variable	Gallium Content (ppm)	Particle Size (μ)	Surface Area (m^2/g)	Loss On Ignition (wt%)
LANL-1	As Received	5100	14.7	5.5	0.29
LANL-2	Vibratory Milled		6.5	11.6	
LANL-3	2h@400°C	TBD	12.3	5.1	0.04
LANL-4	2h@ 1000°C	TBD	13.6	0.7	-0.19
LLNL-1	400°C & Ground	744	16.5	0.8	-0.04
LLNL-2	125°C	239	39.3	3.0	0.02

The results of the MOX fabrication studies are presented in Table II. Although the results of these tests are not all available yet, some interesting observations can be made. First, the addition of only 5wt% PuO_2 to the UO_2 has a significant effect on the sintering behavior of the pellets by lowering the density. Second, most, if not all, of the gallium escapes from the MOX during the pellet fabrication process. Third, vibratory milling when compared to ball milling greatly enhances the sintered density of the pellets. Forth, MOX pellets can be successfully made with recycled material and no binder. And, fifth, the sintering behavior of the oxides from LANL and LLNL are much the same although there is an obvious difference between the two materials from LLNL. It seams acceptable fuel could be made with any of the three oxides. It also looks like the gallium will not be an issue in the reactor core, but that it will be an issue to be dealt with in the fuel fabrication processing equipment unless it is removed from the PuO_2 before it is blended with the UO_2.

Table II. MOX Fuel Fabrication Studies Variables and Results

Batch Identification	First Variable	Second Variable	Density (%TD)	Gallium Content (ppm)
LANL-CONTROL	Ball Milled	UO_2 Only	93.37	
LANL-1	Ball Milled	4h @ 1500°C	72.10	
LANL-2	Ball Milled	4h @ 1600°C	81.64	130
LANL-3	Ball Milled	4h @ 1600°C	79.32	
LANL-4	Ball Milled	16h @ 1500°C	74.98	130
LANL-5	Ball Milled	16h @ 1600°C	85.74	
LANL-6	Ball Milled	16h @ 1700°C	93.17	130
LANL-2&3	Recycled	No Binder	90.16	
LANL-7	Vibratory Mill	Reduced Additives	96.83	<10
LLNL-1	HYDOX @ 400°C	PuO_2 Ground	89.28	10-20
LLNL-2	HYDOX @ 125°C		93.95	<10

CONCLUSIONS

- The gallium content of the PuO_2 produced at LANL by the hydride/oxidation process is the same as in the metal before the conversion.

- The gallium content of the PuO_2 produced at LLNL by the HYDOX process is much lower than that in the metal before the conversion.

- The oxide produced at LANL has a mean spherical equivalent particle size of 14.7μ that can be halved by vibratory milling.

- Heating the LANL oxide to 1000°C had little effect on the particle size, but it did reduce the surface area.

- The addition of 5wt% PuO_2 to the UO_2 has a significant effect on the sintered density.

- Most, if not all, of the gallium escapes from the MOX during the pellet fabrication process which suggests it will not be an issue in the reactor, but it will have to be dealt with in the fuel fabrication processing unless it is removed from the PuO_2 before blending with the UO_2.

- Vibratory milling results in higher sintered density than ball milling.

- MOX pellets can be successfully made with recycled material and no binder.

- The sintering behavior of the oxides from LANL and LLNL are much the same, but there is an obvious difference between the results with the two oxides from LLNL.