Pacific Northwest National Laboratory
Operated by Battelle for the U.S. Department of Energy

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John Voglewede
Mail Stop T-10K8
U.S. Nuclear Regulatory Commission
Washington DC 20555

Dear John:

In April 2006, Pacific Northwest National Laboratory (PNNL) provided the U.S. Nuclear Regulatory Commission (NRC) with a letter report entitled "FRAPCON-3 and FRAPTRAN Assessment Based on Revised MOX [mixed oxide] Data Comparison"[1]. In that report, PNNL suggested the possibility that out-of-reactor densification of MOX may underestimate in-reactor densification for this fuel type.

PNNL has performed a literature search to collect in-reactor densification data on MOX fuel and data from the associated thermal re-sintering test. These data were compared in order to determine if the standard re-sintering test specified in Regulatory Guide 1.126 is applicable to MOX fuel.

The attached document shows the data comparisons that have been found and contains a discussion on the applicability of the standard re-sintering test to MOX fuel. PNNL has concluded that, despite limited experimental data and opposing views regarding the mechanisms of in- and out-or-reactor densification of MOX, there is evidence that the values obtained from the out-of-reactor tests are equivalent to the maximum densification observed in reactor.

This report completes Task 4.20 under JCN N6326. If you have any questions or comments regarding this report, please contact me at (509)375-6828.

Sincerely,

Walter Luscher

Walter G Luscher

Enclosures
Report on Densification in MOX

cc: KJ Geelhood
CE Beyer
DD Lanning

[1] C.E. Beyer letter to J.C. Voglewede dated April 6, 2006

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Densification in MOX Fuel

Introduction

In-reactor densification of fuel pellets can significantly affect fuel temperature in several ways: (1) gap conductance may be reduced because of the decrease in pellet diameter; (2) the linear heat generation rate will increase because of the decrease in pellet length; and (3) the pellet-length decreases may cause gaps in the fuel column and may produce local power spikes and the potential for cladding collapse[1]. The current method for determining maximum in-reactor densification is the re-sinter test, which assumes that the maximum in-reactor densification is equivalent to the densification obtained by subjecting fuel pellets to a post-sintering thermal treatment at 1700°C for 24 hours[1]. The purpose of this research effort has been to conduct a literature survey of in-reactor and out-of-reactor densification data for mixed oxide (MOX) fuel pellets and determine the applicability of the re-sinter test[1].

Discussion

Review of the literature indicates that few studies have made a concerted effort to characterize or quantify both the in- and out-of reactor densification behavior in MOX fuel pellets. Verifying the applicability of the re-sintering test to include MOX pellets is difficult because studies often neglect to report both in- and out-of-reactor densification and are frequently focused on other fuel performance characteristics such as fission gas release (FGR) [2, 3]. In addition, the methods used to measure densification may be more accurate in some studies than others (e.g. fuel stack elongation sensors vs. normalized rod internal pressures). Furthermore, studies often make qualitative comparisons of in-reactor densification behavior between UO_2 and MOX or focus on specific microstructural aspects of sintering under irradiation [4, 5, 6]. While these studies may present valid observations, they do not invalidate the use of re-sinter tests to predict in-reactor densification. In fact, several studies indicate that the in- and out-of-reactor densification of MOX is equivalent, which supports the application of the re-sinter test described by Regulatory Guide 1.126[1, 7, 8, 9, 10, 11]. The following sections will discuss the results of various test programs.

<u>IFA-651.1</u>

Instrumented fuel assembly (IFA) 651.1 contained MOX fuel pellets in rods 1, 3, and 6. Pellets in rod 1 were fabricated by the short binderless route (SBR) while pellets in rods 3 and 6 were attrition milled (ATT). For cycle 1, rod average ratings were maintained between 20 and 25 kW/m for the majority of the cycle, which was approximately 5 MWd/kg oxide. Cycle 2 extended the average assembly burnup to ~ 10.4 MWd/kg oxide and maintained similar rod average ratings. Densification measurements were based on normalized rod internal pressures, which were used to estimate the changes in free volume within the test rods. Although this methodology provides a measure of densification, it is only valid if there is no fission gas release. Since there was no evidence of FGR in the MOX rods in either cycle, the data obtained was deemed valid. As expected, densification was only observed during cycle 1. A densification increase of 2% was observed in rods 1 and 6 while an increase of only 1% was observed in rod 3. Since rods 3 and 6 were produced by the same manufacturing process, the difference in densification was attributed to lower linear heat rates in rod 3. Although these densification increases are slightly larger than the typical density increase for UO_2 pellets given in NUREG 1754[12] (i.e. 0.9%), no out-of-reactor densification data was provided for comparison. As a result, these studies can not verify the applicability of the re-sinter test to include MOX fuels.

IFA-633

A comparative study between SBR MOX and UO_2 fuel was executed with IFA-633[4]. The assembly was irradiated over ten cycles to an average burnup of 35 MWd/kg oxide at an average linear heat rate of ~20kW/m and contained MOX fuel pellets in rods 2, 4, and 6. In-reactor densification was measured by fuel elongation detectors in rods 2 and 4 while a pressure detector was used in rod 6. Results indicated that densification was greater in the MOX rods than the UO_2 rods, which is a common finding among researchers[7]. However, the extent of densification was never explicitly stated within the report and the difference in densification behavior was attributed to smaller initial grain size in the MOX fuel pellets. Taking grain size into consideration, this study concluded that the UO_2 densification model was applicable to MOX and the same inherent processes responsible for in-reactor densification are present in both materials. Despite the parity between MOX and UO_2 described in this study, this report also does not verify application of the re-sintering test to MOX pellets.

IFA-655.1

Results of irradiating IFA-655.1 to ~53 MWd/kg oxide provided additional densification data regarding MOX fuel pellets[6]. Two MOX variants were examined in this study. Both types had similar grain size, but one exhibited a homogenous microstructure (well distributed plutonium) while the other exhibited a heterogeneous microstructure. Fuel extensometers were used to detected stack elongation of these variants in rods 5 and 6, respectively. Results indicate that densification increased by 0.7% and 1.6% in the homogeneous and heterogeneous variants, respectively. Although a difference is expected based on the relative grain size (11 vs. 8 μm, respectively), it was larger than expected by the authors. It was also noted that the homogenous microstructure exhibited dissimilar in-reactor and out-of reactor densification. However, this was not the case for the heterogeneous MOX, which exhibited comparable in- and out-of-reactor densification and offered some support in verifying the use of a re-sinter test for MOX pellets.

IFA-597.4/.5/.6/.7

Mixed oxide fuel pellet performance was also studied in rods 10 and 11 in IFA 597.4/.5/.6/.7 [8, 9]. The average burnup was ~33MWd/kgMOX and the linear power rating was varied to study fission gas release. In these studies, both solid and hollow pellet geometries were examined and densification results were obtained from beginning-of-life pressure data for each pellet type. Densification data indicated an increase of 0.9% and 1.7% in rods 10 and 11, respectively. However, after applying a correction factor for He absorption, the densification data indicated increases of 0.5 and 0.6% for rods 10 and 11, respectively. The persistence of the larger increase in densification observed in rod 11 was attributed to the greater extent of He absorption expected from the hollow pellet geometry. Regardless, the corrected densification data was in agreement with densification data collected out-of-reactor (~0.46%). Results such as these indicate that the re-sinter test discussed in Regulatory Guide 1.126[1] is applicable MOX pellets.

IFA-626

Densification data was also collected from IFA-626 [10, 11]. This test assembly contained 4 SBR and 4 Micronized Master Blend (MIMAS) MOX rods as well as 4 UO_2 reference rods. Initially started in 1995 as IFA-609, signal cable troubles halted testing and the rods were transferred to IFA-626. The test assembly was irradiated to 72GWd/tM at an average power of 15-20 kW/m. In-reactor densification was measured with fuel stack elongation detectors and revealed a 0.4% and 0.2-0.3% for the SBR and MIMAS MOX rods, respectively. These results were in agreement with out-of-reactor densification measurements indicating that the re-sintering test is applicable to MOX pellets.

Other Research Efforts

Microstructural effects on in-reactor densification have been examined by Garcia et al[5]. Densification, both in- and out-of reactor, was defined as a decrease in the as-fabricated pore fraction. However, Garcia stated that in-reactor densification occurred at temperatures too low to be dominated by typical thermal mechanisms. Athermal in-reactor densification was attributed to the radiation field, which generates larger point-defect concentrations than those observed out-of reactor at similar temperatures. Since the sintering mechanisms proposed for in- and out-of-reactor conditions were significantly different, it was concluded that out-of-reactor re-sintering tests could not be considered predictive with regard to in-reactor densification tests on the basis of microstructural evolution.

Despite differences in microstructural densification mechanisms cited by Garcia[5], other researchers have found the in-reactor densification to be equivalent to the out-of-reactor densification observed during the re-sinter test. For instance, a particular study subjected fuel rods containing SBR MOX to a burnup of 45GWd/t at an average power of 20kW/m[7]. Although the SBR MOX exhibited greater densification than the reference UO_2 fuel rods, the overall in-reactor densification of the SBR MOX fuel was equivalent to that observed during the re-sinter test (~0.6%).

Conclusions

Despite limited experimental data and opposing views regarding the mechanisms of in- and out-of reactor densification of MOX, there is evidence that the values obtained from the out-of-reactor tests are equivalent to the maximum densification observed in reactor. Although the re-sinter test is an empirical approximation of in-reactor conditions, prior experience has shown its applicability. Based on literature review presented here, which is summarized in Table 1, it appears that the re-sinter test is applicable to MOX fuel pellets. However, additional studies focused on gathering in- and out-of-reactor MOX fuel pellet densification data would be useful in further validating the applicability of the re-sinter test to MOX fuels.

Table 1. Summary of MOX fuel pellet densification data.

	MOX Description	In-Reactor	Out-of-Reactor
IFA 651.1	SBR Rod 1	2%	Not Measured
	ATT Rods 3 and 6	1% and 2%,respectively	Not Measured
IFA-633	SBR	Not Stated	Not Stated
IFA-655.1	Homogenous Microstructure	0.7%	Not in Aggrement with In-Reactor
	Heterogeneous Microstructure	1.6%	1.6%
IFA-597.4/.5/.6/.7	Solid Pellet	0.5%	0.5%
	Hollow Pellet	0.6%	0.5%
IFA-626	SBR	0.4%	0.4%
	MIMAS	0.2-0.3%	0.2-0.3%
Doi	SBR	0.6%	0.6%

References

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