ORNL/MD/LTR-182

MOX Average Power Test Fuel Pellet Initial Gallium Content

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March 2000

Fissile Materials Disposition Program

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This report documents the MOX fuel pellet gallium analysis that was undertaken to collect preirradiation gallium data from archival average power test MOX fuel pellets supplied by LANL. This effort was sponsored by the Fissile Materials Disposition Program (FMDP). The work was carried out at the Radioactive Material Analytical Laboratory by the staff of the Chemical Technology Division of the Oak Ridge National Laboratory during the period from December 1999 through February 2000. Assistance for material handling was provided by Metals and Ceramics Division staff. LANL staff arranged the transport of the archive pellets from LANL to ORNL.

This is a Level-2 document as defined in the Fissile Materials Disposition Program Light-Water Reactor Mixed-Oxide Fuel Irradiation Test Project Plan, ORNL/MD/LTR-78.

Sincerely,

Robert N. Morris
Fissile Materials Disposition Program

Enclosure

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CONTENTS

LIST OF FIGURES iv

LIST OF TABLES v

ACRONYMS vi

ABSTRACT vii

1.0 INTRODUCTION 1
1.1 General 1

2.0 ANALYSIS 2
2.1 Method 2

3.0 SUMMARY 6

4.0 ACKNOWLEDGMENTS 7

5.0 REFERENCES 8

6.0 DISTRIBUTION 9

FIGURES

Figure 1 Graphical Representation of Pellet Gallium Analysis 5

TABLES

Table 1 Summary of ORNL Gallium Analysis Results 3

Table 2 Summary of LANL Gallium Analysis Results 4

ACRONYMS

ATR	Advanced Test Reactor
FMDP	Fissile Materials Disposition Program
INEEL	Idaho National Engineering and Environmental Laboratory
LANL	Los Alamos National Laboratory
MOX	Mixed Oxide
ORNL	Oak Ridge National Laboratory
PPM	Parts per Million
TIGR	Thermally Induced Gallium Removal

ABSTRACT

This report summarizes the gallium analysis performed at the ORNL Radioactive Materials Analytical Laboratory on the LANL archival average power test MOX fuel pellets. Ten pellets from each MOX fuel batch (20 total) were analyzed for gallium content. The measured gallium levels range from 1.09 to 2.03 ppm with an average of 1.33 ppm for Batch B (treated batch) and from 1.81 to 4.78 ppm with an average of 2.97 ppm for Batch A (untreated batch). While radical differences in gallium values were not noted between the two batches, the standard deviation of Batch A was considerably larger than that of Batch B.

In both cases the gallium levels found by ORNL were higher than those found by LANL, but the small sample size of the LANL analysis does not allow a straightforward comparison.
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1.0 INTRODUCTION

1.1 General

The Fissile Materials Disposition Program (FMDP) [Ref.1] is investigating the use of weapons derived plutonium in mixed oxide (MOX) fuel for use in light water reactors. The program has developed a set of simple uninstrumented drop-in capsules for the irradiation of candidate MOX fuel and clad in the Advanced Test Reactor (ATR) at Idaho National Environmental and Engineering Laboratory (INEEL). This fuel material contains trace gallium impurities and one of the goals of the testing is to determine if trace gallium has any detrimental effects on fuel system performance.

One of the experimental needs is to document the preirradiation gallium content of the fuel so that an investigative baseline can be established. The purpose of this effort was to analyze 10 pellets from each batch (20 total) to obtain representative gallium impurity information. While only a modest size analysis effort was performed, the net result was that the majority of the archival pellets were consumed. Thus, the overall impurity statistics are limited to the current analyses. A small amount of previous analysis was done by LANL (4 pellets total), which is discussed later.

The fuel was made in two distinct batches, one of which was treated for gallium removal (TIGR process) prior to pellet manufacture, Batch B, and one which was untreated prior to pellet manufacture, Batch A. In the end, the pellet sintering process was quite efficient at driving off the gallium from the green pellets so that the final gallium concentrations in both pellet batches were in the low parts per million range (ppm).

2.0 ANALYSIS

2.1 Method

The pellets were weighed and individually dissolved in an acid solution warmed by a 180°C hotplate. Initially (first 4 pellets) a solution composed of 25 ml of concentrated HNO_3, 15 ml of concentrated HCl, and 4 ml of concentrated HF (per pellet) was used for the dissolution, but subsequent investigation revealed that the HCl was unnecessary. The acid solution was changed to 25 ml of concentrated HNO_3 and 1 ml of HF which allowed a more rapid pellet dissolution. The acid mix was added in steps and the individual solution volume was slowly reduced by continuous evaporation. The overall heated dissolution time was 12 hours.

The free fluorine in the acid solution was tied up by adding 35 ml of saturated H_3BO_3 solution prior to a (heated) final dilution to 100 ml with water. The H_3BO_3 also broke up any precipitated UF_6 formed during the dissolution. The solution then underwent Inductively Coupled Mass Spectrometry for the determination of the gallium content. Both gallium isotope mass lines were clearly evident for these unirradiated pellets. Cross checking was done by both spiking and by confirming the natural abundance ratio of gallium isotopes 69 and 71. No abnormalities were noted. The results are summarized in Table 1.

Previously, LANL had done a minimal analysis of four pellets, two from each batch. Their results are shown in Table 2. In general, ORNL found somewhat higher gallium levels, but the limited LANL sample size makes any comparison of limited value.

A graphical representation of the results is shown in Fig. 1.

TABLE 1 Summary of ORNL Gallium Analysis Results			
Batch	**Pellet ID**	**Gallium Content (ppm mass)**	**Batch Values**
A	A1-1	4.78	
A	A1-2	2.22	
A	A1-3	2.75	
A	A2-1	2.67	*Average* 2.97
A	A2-2	4.44	*Standard Deviation* 1.01
A	A2-3	3.53	*95% Confidence Interval* 1.00 to 4.95
A	A5-1	2.32	
A	A5-2	3.19	
A	A6-1	2.03	
A	A6-2	1.81	
B	B1-1	1.52	
B	B1-2	1.34	
B	B2-1	2.03	
B	B2-2	1.15	*Average* 1.33
B	B3-1	1.15	*Standard Deviation* 0.28
B	B3-2	1.35	*95% Confidence Interval* 0.79 to 1.88
B	B4-1	1.18	
B	B4-2	1.09	
B	B5-1	1.36	
B	B5-2	1.17	

Notes:

Unirradiated pellets are from LANL archives. They consist of pellets that may not have met the QA requirements because of physical damage (chipping), but are otherwise identical to the irradiated pellets.

TABLE 2 Summary of LANL Gallium Analysis Results			
Batch	**Pellet ID**	**Gallium Content (ppm mass)**	**Batch Values**
A	N/A	1.4	*Average* 2.0
A	N/A	2.5	
B	N/A	0.59	*Average* 0.73
B	N/A	0.86	

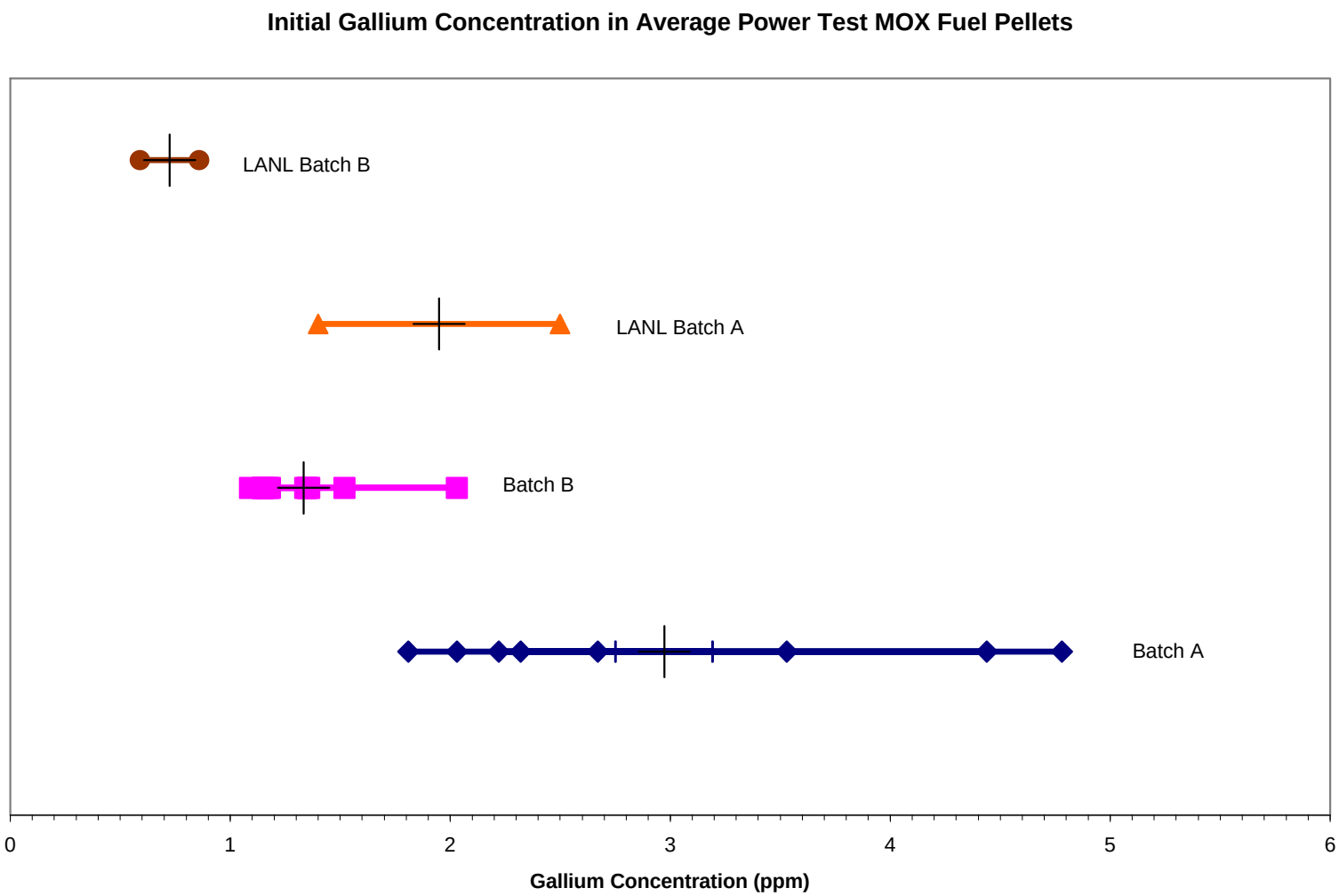

Figure 1 Graphical representation of pellet gallium analysis.

3.0 SUMMARY

Ten pellets from each average power test MOX fuel batch (20 total) were analyzed for gallium content. The measured gallium levels range from 1.09 to 2.03 ppm with an average of 1.33 ppm for Batch B (treated batch) and from 1.81 to 4.78 ppm with an average of 2.97 ppm for Batch A (untreated batch). While radical differences in gallium values were not noted between the two batches, the standard deviation of Batch A was considerably larger than that of Batch B.

In both cases the gallium levels found by ORNL were higher than those found by LANL, but the small sample size of the LANL analysis does not allow a straightforward comparison.

4.0 ACKNOWLEDGMENTS

The authors wish to thank Deborah Figg of LANL for consultations on gallium analysis techniques and methods and both Dave Alberstein and Tom Blair of LANL for making the arrangements to ship the archive MOX pellets to ORNL.

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