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**ORNL/MD/LTR-146
Level 2**

**Gallium-Cladding Compatibility
Testing Plan
Phase III : Test Plan for
Centrally Heated Surrogate
Rodlet Test**

Revision 2

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July 1998

Fissile Materials Disposition Program

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Phase III : Test Plan for
Centrally Heated Surrogate Rodlet Test**

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July 1998

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ABSTRACT

This is a Level-2 document as defined in the Fissile Materials Disposition Program Light-Water Reactor Mixed-Oxide Fuel Irradiation Test Project Plan. This Plan summarizes the projected Phase III Gallium-Cladding compatibility heating test and the follow-on post test examination (PTE). This work will be performed using centrally-heated surrogate pellets, to avoid unnecessary complexities and costs associated with working with plutonium and an irradiation environment. Two sets of rodlets containing pellets prepared by two different methods will be heated. Both sets will have an initial bulk gallium content of approximately 10 ppm. The major emphasis of the PTE task will be to examine the material interactions, particularly indications of gallium transport from the pellets to the clad.

1.0 INTRODUCTION

1.1 BACKGROUND

The Fissile Materials Disposition Program (FMDP) is investigating the use of weapons grade plutonium in mixed oxide (MOX) fuel for light-water reactors (LWR). Commercial MOX fuel has been successfully used in overseas reactors for many years; however, weapons derived fuel may differ from the previous commercial fuels because of small amounts of gallium impurities. A concern presently exists that the gallium may migrate out of the fuel, react with and weaken the clad, and thereby promote loss of fuel pin integrity. Phases I and II of the gallium task are presently underway to investigate the types of reactions that occur between gallium and clad materials.

Currently, the FMDP intends that most of the gallium will be removed during the plutonia powder conditioning prior to fuel fabrication. A special dry process is to be employed. While this dry process has several advantages over the traditional aqueous process, it is not as effective at removing gallium. Investigations and optimizations of powder conditioning and gallium removed by evolution during fuel fabrication are currently being conducted at Los Alamos National Laboratory (LANL). Nevertheless, it is likely that some residual gallium will be carried through fuel fabrication into the finished fuel, and it is important to understand the potential for cladding interactions. The available safety margins with respect to gallium concentration for licensing and safe reactor operation must also be investigated.

This test plan describes Phase III of the gallium task, which is a gallium-cladding compatibility test aimed at establishing confidence in the stability of residual gallium in fuel rods. A surrogate WG MOX fuel would be heated by an imposed thermal gradient. The gradient will be established by a heater located at the radial center of the fuel rod and cooling of the rod outer surface by a liquid metal bath. Phase III provides a cost-effective way to study the transport of gallium impurities in an environment that is a close thermal approximation to that expected in a reactor. Operation of the test apparatus in a non-radioactive environment offers the advantage of low hazard and limited monitoring requirements.

1.2 OBJECTIVES

The objectives of the Phase III test are to determine whether gallium can migrate from the fuel matrix to the pellet/clad interface under the influence of a thermal gradient and if so, the degree to which the gallium at the interface interacts with the clad.

To achieve these objectives, the following steps will be performed:

1. Fabricate two sets of surrogate fuel rods (6 total). One set of these rods will be made from a "low gallium" powder mix while the other set will be made from a "high gallium" powder mix. Gallium concentrations in the finished pellets are expected to be in the neighborhood of 10 ppm.
2. Design and construct a test apparatus that will hold the rodlet outer clad at the proper temperature while maintaining representative temperature gradients in the fuel and clad.
3. Demonstrate that the fuel rodlets and test apparatus are reliable and can perform the required operations.
4. Carry out the gallium-clad compatibility test.

5. Perform a graded post test examination (PTE) on the rodlets to include as appropriate (depending on the end-of-test observations concerning the extent of gallium transport):
 - a) photo-visual and dimensional inspection of the surrogate fuel rods;
 - b) disassembly of the rodlets (as necessary);
 - c) inspection of the inner clad surfaces for evidence of gallium in oxide layers, wall thinning, grain boundary penetration, and transgranular attack;
 - d) estimate of extent of gallium transfer to the clad; and
 - e) inspection of surrogate pellet morphology and microstructure, and a determination of the pellet aggregate gallium content.
6. Prepare a report on the suitability of using the heating apparatus for these tests after approximately 1000 hours of operation.
7. Prepare a comprehensive report on the design and operation of the test apparatus and the results of the PTE on the rodlets.

1.3 SCOPE AND CONDUCT

This Plan describes the generic sequence of steps, procedures, and decision points for the Phase III task. Standard facility procedures and guidelines will be used for the performance of the subtasks as deemed appropriate by the principal investigator. The surrogate pellets will be fabricated at LANL. The test apparatus will be designed, tested, and operated at Texas A&M University Nuclear Science Center (TAMUNSC). The PTE will take place at ORNL. Work will be conducted under the appropriate facility ES&H, and QA guidelines. At each step in the process, the relevance of the remaining work will be assessed so that only examinations with confirmed value to the program will be undertaken.

2.0 TEST DESCRIPTION

The test articles consist of 6 surrogate MOX rodlets, each approximately 30 inches long, with a fueled region approximately 4 inches long. Three of the rodlets will contain pellets from the "high gallium" powder mix and three will contain pellets from the "low gallium" powder mix. After sintering, both sets of rodlets will have a gallium concentration typical of that to be expected in MOX fuel (1 to 10 ppm).

Each rodlet will comprise annular surrogate fuel pellets, alumina insulators, a closed Zircaloy tube, and a small-diameter cylindrical heater. The electrically heated rodlets will be placed in the center of a molten metal bath that will cool the rodlets by natural convection. Heat will be rejected from the system by air cooled fins on the side of the test apparatus assembly.

A schematic of the apparatus is shown in Figure 1. The coolant temperature will increase as it flows up through the heated rod bank and then decrease as the coolant flows down adjacent to the walls of the device. A coaxial construction directs the flow. The power into each heating element will be controlled to maintain a representative temperature gradient across the pellets and a nominal outer clad temperature; auxiliary heaters in the apparatus will be available for start up and testing purposes.

Figure 1. Schematic of centrally heated rodlet test apparatus. Air-cooled fans are not shown.

2.1 RODLETS

The rodlets will be constructed with one of the vendor-supplied clads that has been procured for general program testing [Reference 1]. The current variables in the testing matrix are the two presintering gallium values plus three exposure times (1000, 3000, 5000 h), resulting in a six-test matrix. Specification of two different final gallium levels in the pellet has been considered; fabrication difficulties preclude implementation of this desirable option.

The sintered annular pellets of surrogate MOX fuel will be manufactured by LANL. The surrogate fuel U/Ce/Ga/O is expected to contain approximately 10 ppm gallium. The annular pellets are to have an I.D. of 0.18 inch, a wall thickness of 0.07 inch, and a density greater than 94%. Because of fabrication constraints, the as-produced pellets vary in length from approximately 0.1" to 0.4". The pellet stacks in the rodlets will be composed of random combinations of these pellet lengths to nominal height of 4".

The reference rodlet design diameters (inches) are:

Clad outside diameter	0.382 ± 0.002
Clad inside diameter	0.332 ± 0.0015
Pellet inside diameter	0.185
Pellet outside diameter	0.322 (nominal)

To establish the thermal gradient across the pellets and provide a source for thermally-driven gallium diffusion, a small-diameter (~ 0.18 inch OD) electric heater will be inserted at the center of the surrogate pellet column. The heater will operate within a temperature range of 500 to 1000°C, with a linear power of approximately 15.7 kW/m. The experiment will be configured to produce a representative pellet thermal gradient and a nominal clad-pellet interface temperature of 400°C. A preliminary calculation (TAMUNSC) of the temperatures and heat fluxes is shown in Figure 2.

The pellets, tubing, and heater will be sealed to form the rodlet; once sealed, the rodlet will not be opened until preparation for the PTE. A helium back pressure may be held on the rodlets during the heating to produce an inert test atmosphere.

2.2 TEST APPARATUS

The TAMUNSC will design and construct a non-pressurized system for performing the centrally heated pellet tests. TAMUNSC will build and evaluate the test system using the surrogate rodlets. Computations, scoping tests and a subscale apparatus may be constructed as necessary to prove out the concept.

Using water as a coolant in a prototypic fashion for this task would require the design and construction of a high-temperature pressurized (greater than 2000 psig) cooling loop with a complicated pumping system. To simplify the test apparatus and reduce costs, liquid metal cooling was selected as a means to achieve the test goals while avoiding a high-temperature pressurized system. (A combination gas gap/water jacket is another possibility offering low pressure operation, but the small gaps, high flow rates, and tight mechanical tolerances make a system of this nature less attractive for the Phase III task.)

Lead-Bismuth (Pb-Bi) Eutectic (44.5 wt-% Pb, 55.5 wt-% Bi) has been selected as a coolant because of its relative chemical inertness and low melting temperature. Table I summarizes the properties of the selected Pb-Bi eutectic alloy.

TABLE I
PB-BI COOLANT PROPERTIES

Calculated Atomic Mass	Melting Point (°C)	Boiling Point (°C)	Density at 400°C (gm/cc)	Specific Heat 144-358°C (cal/g-°C)	Thermal Conductivity at 300°C (cal/sec-cm-°C)
208	125	1670	10.19	0.035	0.026

External corrosion of the test specimens and/or the materials of construction of the test system by the lead-bismuth due to thermal-gradient transfer (TGT) is not expected with a steady-state average coolant temperature less than 400°C. A successful compatibility test between the clad material and the coolant will be performed prior to beginning the test. In addition, a compatibility test between the Inconel heater sheath material and gallium oxide was also successful.

The cooling system will consist of an internal flow guide section surrounding the heated rodlet group and a return section concentric with the flow guide (Figure 1). The rodlets will be placed low in the flow guide for maximum thermal driving head. The coolant will flow from the flow guide into the upper heat exchanger. The Pb-Bi coolant will dissipate heat through the air-cooled heat exchanger, sink to the lower insulated section along the wall, and reenter the lower flow guide. The efficiency of the air-cooling will determine the steady-state coolant operating temperature. Modeling reinforced by testing will be used to certify the thermal behavior of the test apparatus.

Considerations for the cooling system concept are based on simplicity and low cost. Using the natural circulation of the coolant avoids design of a mechanical pumping system. Design calculations indicate that natural flow rate will be sufficient, but the high density of the Pb-Bi and system friction may require an inert-gas bubbler to enhance the natural flow in the riser section of the cooling system. Heat will be removed by forced air flowing through fins or channels on the upper section of the main coolant tank. Calculations (TAMUNSC) show that the temperatures produced by a small-diameter heater in the surrogate pellet will closely match the thermal gradient in operating fuel pellets (Fig 2).

System components will be fabricated from ferritic stainless steel or other compatible materials that have good corrosion resistance to Pb-Bi. The cladding samples will be immersed directly into the Pb-Bi coolant and held in place by the cover over the bath. The thermal and hydraulic parameters of the system have been calculated and indicate that the designed system will be able to remove the 20-30 kW of heat generated. A half-size prototype was built at the TAMUNSC and was used to benchmark the calculations and test the data acquisition and protection systems.

The test system will be operated for approximately 5000 hours. After approximately 1000 hours of operating time, an evaluation of the systems performance will be made by TAMUNSC. The primary goal of this evaluation is to determine the suitability of the design for the intended purpose and any operational problems.

Figure 2. Energy flux and temperature profiles for simulated and actual PWR fuel.

2.3 INSTALLATION AND OPERATION

The test apparatus will be installed and operated at the TAMUNSC. The test will be conducted by TAMUNSC; overall program coordination and PTE will be provided by ORNL.

The operation of the test apparatus will be largely automatic with the controller and the data logger commanding and monitoring the test. At specified times, the test engineer may retrieve and review the data to confirm the proper operation of the test apparatus. The only significant activity associated with the heating phase of the test will be the periodic removal of the rodlets at their specified time intervals (1000 h, 3000 h, and 5000 h). Once permanently removed from the apparatus, the rodlets will be prepared for PTE and shipped to ORNL for performance of the PTE.

To economize on the use of test specimens, the program may elect to remove a set of rodlets at the appropriate time, but examine just one. If the selected rodlet shows little or no signs of gallium transport, the sister rodlet may be reinserted into the test apparatus for continued heating. The extra exposure time will be based on experimental knowledge accumulated at that time. Figure 3 shows the PTE decision flowchart.

Figure 3. Rodlet PTE decision flowchart

3.0 POST TEST EXAMINATION

PTE of the rodlets will be performed. Evidence of gallium transport across the pellet and into the clad, wall thinning, grain boundary penetration, and other changes, when present, will be documented. The morphology, microstructure, and gallium content of the surrogate fuel pellets will be evaluated and compared to the as received material. The primary interest is in the gallium that leaves the pellet and deposits on or enters the cladding material. As the gallium program evolves, the degree to which this task is performed will be determined at the start of the PTE effort. An important goal is to identify analytic techniques for this task.

The fuel and clad sampling matrix is shown in Table 2. Samples will be taken as needed from the available rodlets. Samples can be obtained by sectioning the fuel pin either in a transverse or an axial manner depending on the sample shape desired. The PTE will be conducted in a graded fashion to focus resources on the areas of interest. Primary interest will be on the transport of gallium to the clad and whether this gallium has penetrated the surface oxide layer and reacted with the clad. Investigations of gallium transport through the pellet are also important and will be made as resources and schedule permit.

TABLE 2
RODLET SAMPLE MATRIX

Sample Type	Sample Location	Analysis
Rodlet	N/A	Photo-Visual Examination
		Dimensional Measurement
Rod Segment (Segment may be either an axial or transverse cut through the rodlet)	Various axial regions along the fuel pin	Photo-Visual Examination
		Ceramography/Metallography
		Aggregate Gallium Analysis
		Elemental Analysis and Mapping

The planned analysis for the 6 rodlets is shown in Table 3. This work will be done in a hood, glovebox, or hot cell as equipment and resources permit. Minimal hazards are associated with the surrogate material.

3.1 INITIAL RODLET INSPECTION

Before any PTE work is done, the test engineer will locate and record the identifying marks on each rodlet; if none are present, the rodlets will be marked so that their identity can be maintained. The project will document the ID before the PTE begins. In addition, each rodlet will be radiographed prior to any destructive PTE to determine its internal state. Internal damage such as cracks or voids will be noted and may be used to guide the later segmenting of the rodlet.

TABLE 3
SURROGATE FUEL AND CLAD ANALYSIS

Planned Analysis - Six Rodlets total, 1000 h (2), 3000 h (2), and 5000 h (2) heating times. Archives will be examined as appropriate.		
Examination	Approximate Number per Rodlet	Estimated Total Number
Rodlet Photo-Visual and Dimensional Inspection	1	6
Rodlet Segment Photo-Visual Inspection (Rodlets and/or archives)	2	12
Rodlet Segment Ceramography/Metallography (Rodlet and/or archives)	2	12
Selected Clad Specimen Hardness Test (if applicable)	1	6
Clad Segment Elemental Mapping (Tentative)	1	6
Pellet Elemental Mapping (Tentative)	1	6

The PTE will begin by visually inspecting the rodlets for physical abnormalities such as blisters, cracks, unusual or heavy corrosion layers, weld or seal failures, or distortions. Any abnormality and its location on the rodlet assembly will be recorded by photographs and/or video recordings. Should any major problem be noted such as breeched containment, broken welds or seals, liquid metal intrusion, or large physical distortions, the ORNL project manager will be consulted before proceeding with further PTE tasks.

The following measurements will be performed on the fueled region (the lower 4" or so) of the rodlet as conditions permit:

- 1) The diameter of the rodlet will be measured at a minimum of four equally spaced longitudinal locations.
- 2) Each diameter measurement will consist of at least two measurements 90° apart so that any out-of-round condition will be detected.
- 3) The outside of the rodlet will be checked for variations of diameter along its length ("bambooing"). All dimensional measurements will be recorded in a logbook.

3.2 SEGMENTING OF THE RODLET

After a rodlet has been visually and dimensionally inspected, it may be segmented for examination either in a transverse or an axial manner. A schematic of the likely segmenting is shown in Figure 4. It is likely that the fuel will be loose within the clad, and special techniques may be used prior to segmentation to preserve the geometrical arrangement of the fuel and clad. Before this task is started, the current goals of the FMDP should be revisited as this work is destructive.

Once the segments have been cut, they will be visually examined and prepared for mounting and polishing. Additional small samples may also be cut from the segments or rodlets, depending on the methods of elemental analysis to be employed.

Figure 4. Rodlet sectioning specimens

3.3 CERAMOGRAPHIC/METALLOGRAPHIC MOUNTS

Selected rodlet segments will be mounted and polished for ceramographic/metallographic examination. These specimens will be visually examined and photographed under high magnification, paying special attention to heater-pellet and pellet-clad interfaces. Selected mounts may undergo additional preparation for high-resolution scanning electron microscope examination (SEM) and elemental surface distribution mapping using the integrated electron microprobe capability of the SEM.

3.4 GALLIUM ANALYSIS

For the PTE, aggregate gallium measurements may be attempted on intact pellets or large sections of an original pellet. Radial analysis of the gallium profiles will require either scanning the pellet cross section with special instruments or cutting the pellet into sections, performing an aggregate gallium analysis on each small piece, and assembling the results to produce a profile.

The gallium transport investigation may begin by the analysis of a couple of samples in a SEM to find the disposition of the gallium; it might be evenly distributed throughout the sample, segregated at the surface of the pellet, or concentrated in grain boundaries. Once the gallium disposition is known, the feasibility of doing micro-X-ray fluorescence (MXRF) can be evaluated. Radial gallium scans are most easily done using MXRF and it is the preferred method if suitable. Additional measurements using comparative neutron activation analysis (CNAA) may be attempted on the cut up pieces of a pellet or a clad section. CNAA will serve as a back up method, as it has been used by other programs to determine trace gallium levels. Since trace gallium detection methods are still under study, this task will evaluate and select the best method for gallium detection at the time that the PTE is to be performed.

4.0 STORAGE OF RODLET SEGMENTS

All of the fuel pellets, cladding, and specified pieces of the fuel pins will be stored in suitably numbered containers unless the project requests that they be discarded during the PTE task. A list of the PTE objects, their location, and the personnel preparing these records will be kept. The identification and contents of each container will be recorded in an appropriate logbook.

After the completion of Phase III, the test apparatus and all remaining rodlets, mounts, other specimens, components, etc., will be discarded.

5.0 DOCUMENTATION

Observations, photographs, videotapes, digital files, and other data will be recorded on the appropriate medium and documented in UTAMNSC and ORNL laboratory notebooks as the testing and examination proceeds. Progress of both the heating and the PTE effort will be described in the project monthly reports. The final results of the UTAMNSC heating will be documented in a final report in addition to the 1000 hour interim report. The final results of PTE examination will be documented in an ORNL topical report.

6.0 SCHEDULES AND MILESTONES

The schedule for the task and milestones are shown in Table 4 and Figure 5. The PTE task items are subject to change and/or modification as the program proceeds and new information becomes available.

TABLE 4
Phase III Milestones

Item	Date
Complete Fabrication of Annular Pellets and Rodlet Assembly (LANL & TAMU)	June 26, 1998
Start-up 5000 Hour Testing (Texas A&M Nuclear Science Center)	July 31, 1998
Letter Report Documenting Suitability of Test System for Intended Purpose (Texas A&M/ORNL)	September 30, 1998
Final Report on Phase III Tests (ORNL & TAMU)	July 30, 1999

Figure 5 Phase III Schedule

7.0 REFERENCES

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8.0 DISTRIBUTION

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