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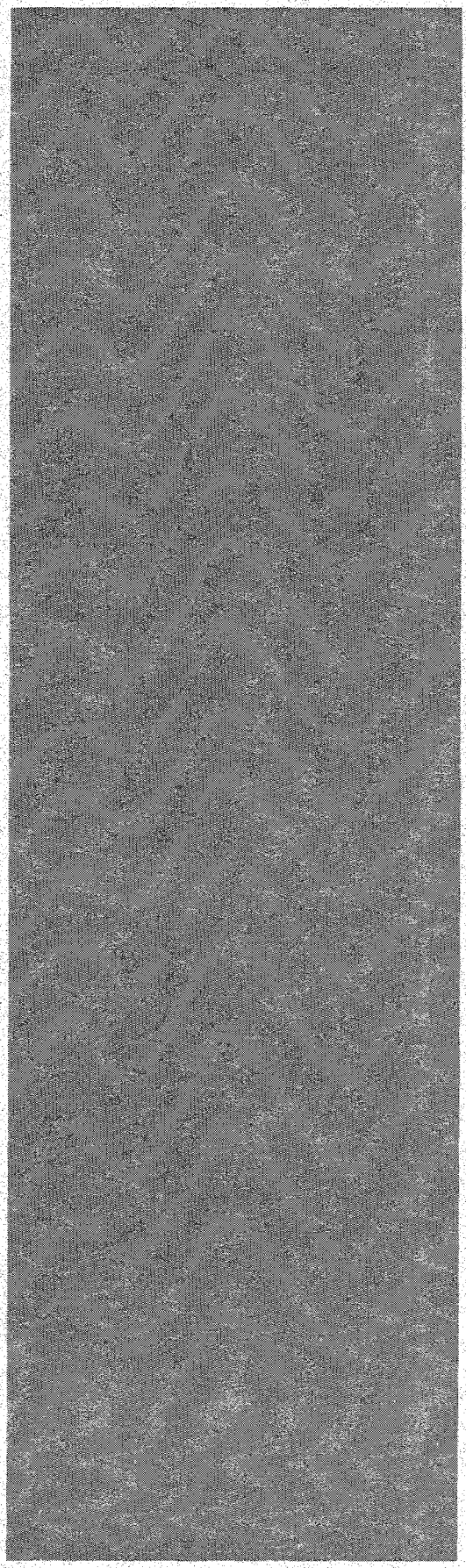

Nuclear Fuels Technologies Fiscal Year 1998 Research and Development Test Plan

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Abstract

A number of research and development (R&D) activities are planned at Los Alamos National Laboratory (LANL) in FY98 in support of the Department of Energy Office of Fissile Materials Disposition (DOE-MD). During the past few years, the ability to fabricate mixed oxide (MOX) nuclear fuel using surplus-weapons plutonium has been researched, and various experiments have been performed. This research effort will be continued in FY98 to support further development of the technology required for MOX fuel fabrication for reactor-based plutonium disposition.

R&D activities for FY98 have been divided into four major areas: (1) feed qualification/supply, (2) fuel fabrication development, (3) analytical methods development, and (4) gallium removal. Feed qualification and supply activities encompass those associated with the production of both PuO_2 and UO_2 feed materials. Fuel fabrication development efforts include studies with a new UO_2 feed material, alternate sources of PuO_2, and determining the effects of gallium on the sintering process. The intent of analytical methods development is to upgrade and improve several analytical measurement techniques in support of other R&D and test fuel fabrication tasks. Finally, the purpose of the gallium removal system activity is to develop and integrate a gallium removal system into the Pit Disassembly and Conversion Facility (PDCF) design and the Phase II Advanced Recovery and Integrated Extraction System (ARIES) demonstration line. These four activities will be coordinated and integrated appropriately so that they benefit the Fissile Materials Disposition Program. This plan describes the activities that will occur in FY98 and presents the schedule and milestones for these activities.

1.0 Introduction

This document details the research and development (R&D) activities that will be conducted in Fiscal Year 1998 (FY98) by the Nuclear Fuels Technologies Program team at Los Alamos National Laboratory (LANL) for the Department of Energy Office of Fissile Materials Disposition (DOE-MD). This work is a continuation and extension of experimental activities that are conducted in support of the disposition program and are based on the use of surplus-weapons plutonium in the fabrication of mixed oxide (MOX) nuclear fuel for reactor-based disposition. The purpose of this work is to provide information leading toward the resolution of technical issues associated with the use of surplus-weapons plutonium in the fabrication and use of MOX fuel in commercial light-water reactors (LWRs). Not only do the activities described in this plan directly support plutonium oxide preparation and analytical improvements, as well as other ongoing Nuclear Fuels Technology Program efforts, but the results are expected to support the procurement process for MOX fuel fabrication and irradiation services and the subsequent contract negotiations. In addition, these results will be of significant value to the selected

commercial fuel fabricator by providing a technical basis upon which to build, thereby reducing the amount of development effort and time required for implementation of the MOX disposition mission.

Although fabrication of MOX fuel using reactor-grade plutonium is a well-developed, industrialized process, several differences exist between the reactor-grade and surplus-weapons plutonium which generate technical issues that must be resolved. These differences include: variation in powder characteristics because the weapons material is to be converted using a dry pyrochemical process as opposed to a chemical dissolution and precipitation process as used in spent-fuel reprocessing facilities; the presence of gallium in the weapons material; and the variation in plutonium isotopics between the reactor-grade and surplus-weapons material. All of the experiments outlined in this report address one or more of these issues, and Table I summarizes which issues each project addresses.

The R&D activities proposed for this fiscal year are divided into four major areas: (1) feed qualification/supply, (2) fuel fabrication development, (3) analytical methods development, and (4) gallium removal. The feed qualification and supply activity (Section 2.0) is intended to cover all issues associated with the identification, acquisition, and characterization of both PuO_2 and UO_2 feed materials. This includes the creation and utilization of a feed characterization database, the acquisition of required feed materials, publication of a draft PuO_2 feed specification, and all additional interactions with the pit disassembly and conversion activities.

The fuel fabrication development activity (Section 3.0) will focus on baseline development work necessary for the use of a new UO_2 feed material. This includes establishing process parameter ranges for powder preparation, pressing, and sintering to fabricate high density cylindrical pellets. The process parameter ranges will then be used to develop baseline parameters to fabricate MOX fuel pellets. In addition, fabrication studies will be performed with alternate sources of PuO_2 to study their effects on processing and final pellet characteristics. The effects of gallium on the sintering process will be studied first with CeO_2 as a surrogate for PuO_2 in a blend with UO_2, and then a few select tests will be performed with PuO_2 in the blend.

The analytical methods development activity (Section 4.0) will serve to upgrade and improve several analytical measurement techniques needed to support other R&D and test fuel fabrication tasks and will provide valuable information for the selected commercial fuel fabricator. The methods being developed include micro x-ray fluorescence (MXRF) to measure spatial distribution and concentration of gallium and an existing Laser Induced Breakdown Spectroscopy (LIBS) system to measure gallium on-line during processing. A small effort will be applied to improving the autoradiograph for measuring homogeneity of plutonium in a MOX fuel matrix and to study alternate oxygen-to-metal (O/M) ratio measurement techniques with the goal of improving efficiency and accuracy.

The gallium removal system activity (Section 5.0) describes the tasks in the first year of a two-year effort designed to develop and integrate a gallium removal system into the Pit Disassembly and Conversion Facility (PDCF) design and the Phase II Advanced Recovery and Integrated Extraction System (ARIES) demonstration line. The primary activities for this year include process development to establish the gallium removal system design requirements, completion of a study with PuO_2 to validate surrogate gallium removal studies, and completion of a system design.

A fifth activity of project coordination and integration (Section 6.0) was added to ensure the timely completion of activities, reporting of results, and coordination of the R&D activities with PDCF and other MOX fuel activities. Finally, an overall R&D schedule and a

summary of the major milestones for the activities described herein are provided in Section 7.0.

Table I. Relationship Between Proposed Tasks and Technical Issues

Applicable Section	Task	Difference in PuO_2 Characteristics	Presence of Gallium	Isotopic Differences
2.1	Identification of Disassembly and Conversion Activities	X	X	
2.2	Feed Characterization Database	X	X	X
2.3	Feed Acquisition	X	X	
2.4	PuO_2 Feed Specification	X	X	X
3.1	Feed Materials Baseline Development	X		
3.2	Alternate PuO_2 Feed Test	X		
3.3	Gallium Sintering Study		X	
4.1	MXRF Development		X	
4.2	O/M Measurement Technique Evaluation	X		
4.3	LIBS Capability Implementation		X	
4.4	Autoradiography Development			X
5.1	Gallium Removal Process Development		X	
5.2	Gallium Removal Prototype Design and Testing		X	

2.0 Feed Qualification/Supply

The intent of this activity is to define and develop the processes, equipment, and specifications for producing PuO_2 and UO_2 feeds needed to qualify MOX fuel. These efforts are ultimately intended to support the award of a contract for private industry to perform the plutonium disposition mission. Information is needed by the winning fabricator on the characteristics of the PuO_2 feed materials and how these characteristics affect MOX fuel fabrication. Programmatically, it is also desirable to understand the potential classification issues associated with the surplus-weapons PuO_2 feed, if future experimental results indicate this to be a problem. This activity will be performed in close conjunction with the pit disassembly and conversion activities also currently in progress. In addition, this activity includes procurement and characterization of UO_2 feedstocks, which will be blended with PuO_2 for MOX fuel development and testing. The intent is to acquire UO_2 feed powder which is representative of the powders used by the current commercial MOX fuel industry.

There are several specific tasks included in this activity:

- Identify plutonium components planned for disassembly and conversion, their schedule of availability, and the processing parameters associated with their conversion to feed material.
- Establish a PuO_2 feed characterization database, including computerized record keeping.
- Obtain PuO_2 and UO_2 required for research, development, and testing activities.
- Create a draft DOE-MD PuO_2 feed specification for use by the government and contractor during procurement negotiations.

As well as supporting the procurement process, each of these activities also supports several other R&D and MD programmatic tasks occurring this year, including both Fuel Fabrication and Thermally Induced Gallium Removal (TIGR) R&D (see Sections 3.0 and 5.0, respectively) and fabrication of test fuel to be irradiated in the Advanced Test Reactor (ATR) at the Idaho National Engineering and Environmental Laboratory. In addition to the specific tasks outlined above, this activity will include the overall interactions between the Nuclear Fuels Technologies and Pit Disassembly and Conversion projects, as well as participation in material declassification and inventory assessment and management activities as necessary.

2.1 Identification of Disassembly and Conversion Activities

This task will cover all efforts to coordinate MOX fabrication activities with pit disassembly and conversion activities. In general, plutonium components that are planned for disassembly and conversion will be identified. Their schedule of availability will be determined, and their associated processing parameters will be tracked. This will include components scheduled to undergo disassembly and conversion at both LANL and Lawrence Livermore National Laboratory (LLNL). Information on the characterization of the precursor (metal) and product (oxide) materials will be obtained and entered into a database. The funding for the characterization effort will originate from a source to be defined by DOE-MD. In addition, the finger ingot data from post-1979 component production and other sources of applicable data will also be reviewed and incorporated as appropriate.

2.2 Feed Characterization Database

MOX fuel fabrication with PuO_2 derived from weapons differs from standard MOX fabrication with reactor-grade plutonium in three significant ways: (1) surplus-weapons PuO_2 will be derived through dry conversion of metal to an oxide and will have a different powder morphology than reactor-grade PuO_2 which is produced from a aqueous process, (2) surplus-weapons metal contains impurities not commonly found in reactor-grade material, and (3) the isotopics of surplus-weapons PuO_2 differ significantly from those of reactor-grade. These differences are significant enough to warrant development of a surplus-weapons MOX fabrication database. This database is intended for comparison to the existing commercial reactor-grade MOX fuel database to provide assurance of the suitability of surplus-weapons MOX for light-water reactor irradiation. In addition to furthering the development of a licensing basis, this database can also serve as an R&D tool. Central collection of R&D data will facilitate correlation of results and identification of additional data requirements, thereby helping to guide the direction of ongoing R&D studies.

The feed characterization database will provide a means of tracking surplus-weapons plutonium from its weapons source through any processing steps to post irradiation examination (PIE) of the MOX fuel. The specific parameters used for each process and the

sample data generated from each material product form will be stored electronically. The intent is to enable correlation of material properties and process parameters with fuel characteristics and performance. Material at any point in the fabrication process should be clearly traceable back to its weapon source as well as its UO_2 source for blended materials. To provide this traceability, a record will be developed for each unique batch of fuel. A single record will contain all the data associated with the characterization and processing of a unique batch of fuel. This may involve the development of several different records from a single plutonium metal source. For example, a single record for PuO_2 powder produced from the same weapon source and undergoing the same metal to powder conversion process will be split into two or more different records if the subsequent PuO_2 gallium removal processing parameters are varied for different portions of the original unique powder batch. For this example, the different records would be identical up to the gallium removal step.

A separate document will be issued detailing the structure of the database. The records will be filled in with historical data, as well as new data as it is collected. Some material will be used in fabrication R&D efforts and will never be completely processed to produce fuel, and not all fuel will be irradiated or undergo PIE. Thus, some records will be incomplete while awaiting further processing data, and some will never be completed. Overall, however, all feed materials and MOX fuel processing information will be available in some form to the selected vendor.

Milestones:

- Complete MD MOX Feed Database Architecture (November 1997)

2.3 Feed Acquisition

This task will cover all efforts associated with the acquisition of PuO_2 and UO_2 feed materials for planned research, development, and testing activities. First, the quantities and extent of characterization performed on materials already on hand will be evaluated. The availability of these existing sources will be integrated with the schedules and requirements for other planned R&D and test fuel fabrication tasks to determine additional feed material needs.

Sources of PuO_2 powder will be needed for fuel fabrication and TIGR R&D activities (see Sections 3.0 and 5.0, respectively), as well as for ATR test fuel fabrication. PuO_2 powder created at LLNL by the 3-step (hydride-nitride-oxidation) method is already available at LANL. Although some of this powder has been used in previous R&D and test fuel fabrication experiments, a significant portion is still available that has been fully characterized. LLNL 2-step (hydride-oxidation) powder is also available and characterized, but is not considered to be prototypic of the ultimate HYDOX process product. The possibility exists for obtaining more 3-step material from LLNL disassembly and conversion activities, but this availability will need to be evaluated as their work progresses. Finally, although LANL does not currently have PuO_2 directly oxidized from metal ingots on hand, it can be produced as necessary and is considered a potential source of feed material.

The MD Program has made the decision to select a single UO_2 source for use in FY98 fuel fabrication R&D activities. As part of this activity, therefore, LANL will acquire a new source of depleted UO_2 feed material derived through the ammonium uranyl carbonate (AUC) process. This material is available commercially through ABB in Sweden, and was converted from UF_6 to UO_2 by the AUC process. This conversion process has been used to supply feed for fabrication of more than 90% of the world's MOX fuel supply. The AUC process produces an oxide with superior ceramic characteristics, including particle

size, surface area, and resistance to stoichiometry (O/M ratio) shifts under storage conditions. It is important for the MD Program's success to mimic the previous MOX experience as closely as possible through use of similar feed materials. Hence, this material will be used for such activities as the fabrication of MOX fuel containing surplus-weapons plutonium to tie the program's disposition efforts closely to the extensive European MOX database.

Milestones:

- Obtain PuO_2 for R&D and Test Fuel Fabrication Activities (January 1998)
- Obtain AUC UO_2 (January 1998)

2.4 PuO_2 Feed Specification

The main interface between pit disassembly and conversion and MOX fuel fabrication efforts for the MD program will be by means of a PuO_2 feed specification. This specification will detail the physical characteristics of the material, including maximum acceptable impurity levels. This specification will serve as a major negotiating point between the government (DOE) and the selected fuel fabricator. The government will agree to a specification as its commitment of what the PDCF will produce. The selected vendor will agree to the specification knowing that feed material that meets the specification will need to be used to fabricate acceptable MOX fuel. As such, a draft specification will be needed by both sides as a starting point for negotiations. This activity will produce such a draft for the DOE, using all available resources of feed material characterization to determine individual limits.

Milestones:

- Issue Draft MD PuO_2 Feed Specification (June 1998)

3.0 Fuel Fabrication Development

The purpose of the fuel fabrication development activities is to identify and, if possible, resolve technical issues associated with applying the large experience base (existing mainly in Europe) of making MOX fuel with recycled reactor-grade plutonium to the fabrication of MOX using surplus-weapons plutonium. More specifically, the fabrication of MOX fuel using a new baseline of AUC-derived UO_2 and various PuO_2 sources and processing parameters is required to establish a database useful in support of the selection of a private firm to carry out fuel fabrication activities of the disposition mission. Furthermore, in support of contract negotiations, gallium sintering studies will help determine the impact of residual gallium levels on fuel fabrication process parameters and equipment, with specific attention being paid to the impact of residual gallium levels on the sintering process.

The first two tasks in this activity involve the development of fabrication processing parameters for use with new feed materials. It has been determined through previous efforts that a certain amount of development work is necessary when new feed materials are introduced into an established fabrication process. MOX fuel fabrication activities to date in support of DOE-MD 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 been used to fabricate the majority of the European reactor-grade MOX fuel, it is important to establish how it will interact with surplus-weapons plutonium in terms of fuel fabricability. Furthermore, plutonium feed materials from the same weapons plutonium conversion process (HYDOX) can appear quite different (from a ceramics perspective) depending on the processing parameters, and it is important to quantify the effect these differences may have on the fuel fabrication process.

The third main task is the gallium sintering studies. Previous R&D experiments performed at LANL have demonstrated that the gallium found in surplus-weapons plutonium volatilizes in a reducing atmosphere. Since the sintering of MOX fuel occurs in a reducing atmosphere, this behavior could lead to significant sintering furnace degradation especially in larger-scale processing of surplus-weapons MOX fuel. Although concurrent activities are underway this fiscal year to develop a process by which the gallium is removed prior to sintering (see Section 5.0), it is likely that some residual amount of gallium will still remain in the feed material. It is important, therefore, to learn about the behavior of gallium during sintering and what impact gallium will have on the fuel fabrication process and equipment. The sintering studies will be performed using both cerium (as a surrogate for plutonium) and actual plutonium.

As a separate task, work will continue on the optimization of related phase diagrams involving UO_2, PuO_2 (or CeO_2), and Ga_2O_3. A fundamental understanding of the phase relationships and interaction between the components of this material is necessary in order to predict behavior under various conditions. This information will enable process engineers to set optimum sintering parameters and help fuel performance engineers model in-reactor behavior.

3.1 Feed Materials Baseline Development

This task will focus on the development of new baseline MOX fuel fabrication processing parameters. The MD Program has made the decision to select a single UO_2 source for use in FY98 fuel fabrication R&D activities. Previous experience has shown that a certain amount of development time is needed with the introduction of any new material into an established process. This activity, therefore, will help to ensure the success of future fuel fabrication R&D activities by establishing processing parameters by which the baseline UO_2 source can be combined with surplus-weapons PuO_2 to fabricate a useable MOX fuel form.

As part of a separate task this fiscal year, LANL will acquire a new source of depleted, AUC-derived UO_2 feed material (see Section 2.3). After the new feed material has been obtained and is available for use, development activities will begin to establish a process by which MOX fuel can successfully be fabricated using the AUC-derived material. Sinterability experiments will be performed using a wide matrix of processing parameters (for milling, additive, pressing, and sintering processes). Initial experiments will be performed using solely the UO_2 powder until a predetermined sinterability standard can successfully be achieved.

Development work incorporating the PuO_2 feed source will be performed simultaneously with the UO_2-only work to utilize resources most effectively. For this effort PuO_2 powder created by LLNL by the 3-step (hydride-nitride-oxidation) method will be utilized. This material is already available at LANL, has been fully characterized, and will provide a link to previous LANL MOX fuel fabrication efforts. Small batches of MOX fuel will be fabricated concurrently with UO_2 fuel, initially using the same processing parameters. The two types of fuel will be compared directly in terms of pressability, green density, and final sintered density. Processing parameters will be established by which a quality MOX fuel form can be fabricated using the AUC-derived UO_2 and the 3-step PuO_2 feed materials. In parallel, similar experiments will be performed in the cold laboratory using CeO_2 as a surrogate material for PuO_2. This process will then serve as a baseline for future surrogate MOX fuel fabrication activities.

The results of this task will directly support the remainder of the fuel fabrication development tasks planned for this fiscal year. The processing parameters developed will

be used to fabricate fuel with an alternate source of PuO_2, as described in Section 3.2. These parameters will also be used in the study of how gallium impacts the sintering process, as described in Section 3.3. A separate test plan will be issued for this task, as well as a summary report detailing the results of the development work.

Milestones:

- Complete AUC UO_2 Baseline Development Plan (November 1997)
- Complete Baseline Process Development Report (August 1998)

3.2 Alternate PuO_2 Feed Test

After the baseline development task has been completed (see Section 3.1), processing parameters will have been established by which MOX fuel can successfully be fabricated using the AUC UO_2 and the LLNL 3-step PuO_2. For the alternative PuO_2 fabrication task, the same parameters will be used to fabricate MOX fuel with the AUC UO_2, but using an alternative PuO_2 source. Sources of PuO_2 feed materials can vary widely, depending on the conversion process utilized. Feed materials from the same process can also exhibit differing powder characteristics, depending on the processing parameters. It is important, therefore, to quantify the effect a different PuO_2 source will have on the established fuel fabrication process.

To utilize a different source of PuO_2 feed material from that used in the previous feed materials baseline development activity (LLNL 3-step), a source will first have to be identified and acquired, if necessary. A few possible sources exist, some of which are already on-site at LANL. LLNL previously made oxide by the 2-step (hydride-oxidation) method, which was shipped to LANL with the 3-step material last year. This material has not yet been used in experiments and could serve as a possible source. Work is ongoing at LANL on a direct oxidation of metal weapons plutonium conversion method, and some of that product might be made available. Other sources resulting from the ARIES/HYDOX development work will need to be examined for their applicability and availability. The effort of identifying and obtaining an alternate source of PuO_2 is covered by another task in the FY98 R&D activities (see Section 2.3).

MOX fuel will be fabricated using the established baseline fabrication processing parameters, the AUC UO_2, and the identified new source of PuO_2 material. The results will then be directly compared against those of the fuel previously made with the LLNL 3-step material for the baseline development efforts (Section 3.1). A separate test plan will be issued for this task, and the results will be incorporated into the Baseline Process Development Report.

Milestones:

- Complete Alternate PuO_2 Feed Test Plan (November 1997)

3.3 Gallium Sintering Study

The main thrust of the sintering studies is to fabricate MOX pellets containing varying amounts of Ga_2O_3, sinter the pellets under a wide range of conditions, then characterize the sintered pellets to determine what effects, if any, the gallium has on the sintering process. Studies will first be performed in the "cold" (without plutonium) laboratory using surrogate materials. Surrogate materials can easily be used to examine a wide range of experimental variables at a lower cost than can be achieved using plutonium-bearing materials. For this particular activity, CeO_2 will be used as a surrogate for PuO_2 in all experiments. In addition, "hot" laboratory studies will be performed in the Plutonium Facility at LANL. Actual surplus-weapons plutonium will be used in all of those experiments.

To accomplish these studies, it is first necessary to obtain the proper feed materials. If AUC-derived UO_2 feed can be obtained early enough in the fiscal year, these studies will be performed using solely that material. Otherwise, the studies will be initiated with UO_2 feed already available, and confirmatory studies will be performed upon receipt of the AUC-derived material. For the surrogate studies, the feed materials will be put through an initial fabrication process to obtain a homogeneous mix. The standard method of fabrication is to press initial pellets, crush and grind the pellets into powder, repress the powder into pellets, then sinter the pellets. This method, albeit time-consuming, will be used in some of the planned experiments. Concurrently, other fabrication processing methods will be investigated, with a focus on improving process efficiency. This investigation will also serve to create feed materials with differing morphologies, one of which should be similar to that of the MOX feed material eventually used in the full-scale disposition MOX fuel fabrication.

For the plutonium studies, a source of PuO_2 feed material must first be identified for use, preferably with varying levels of gallium. Available PuO_2 material from previous activities have initial gallium concentrations of almost 9000 ppm (in PuO_2). (LLNL 3-step material has 8846 ppm gallium, while 2-step material produced some time ago at LANL has 8714 ppm gallium.) This will serve as the upper limit of the range for the sintering studies. PuO_2 feeds with lower concentrations can be obtained from gallium removal R&D experiments, or use could be made of already-processed material from other activities (i.e. the LWR Demonstration Test fabrication efforts). The possibility of obtaining gallium-free PuO_2 will also be investigated. For the surrogate studies, the gallium concentration range will be selected to coordinate with that achievable in surplus-weapons feed materials.

Each type of fuel will be sintered according to parameters specified in the Gallium Sintering Study Test Plan. The actual number and type of experiments completed will depend on resource availability, extent of characterization required, and the results of experiments as they are performed. The number of experiments performed during the surrogate studies will not be reproduced as a part of the plutonium fabrication efforts. Instead, a limited number of experiments will be selected to reproduce the more promising of the surrogate results in a more realistic fashion. There are then several types of characterizations that need to be performed to adequately determine the gallium effect on sintering. Focus will be on the green and sintered pellet densities, and further analyses will be specified in the test plan. For example, fuel users are concerned with such characteristics as grain size and pore size and distribution, while fuel fabricators are concerned with material handling issues such as pellet chipping during grinding. It is important to determine if such characteristics are affected by gallium during sintering.

A separate activity in this task is the continuation of the phase relations assessment begun as part of last year's R&D activities. During that effort, an assessment was initiated of the phase relations between UO_2, PuO_2, and Ga_2O_3, also using CeO_2 as a surrogate for PuO_2. This information is needed in the gallium sintering studies (as well as several other of this year's R&D activities), as the basic thermodynamic information learned here will help to better predict and understand behavior seen in the sintering studies. Efforts will continue on assessment of the PuO_2-Ga_2O_3 phase diagram as part of the TIGR development activity. This task will also include the evaluation of the plutonium-gallium perovskite stability and will continue the assessment of the solubility of gallium in plutonium oxide. Studies similar to these will be performed for CeO_2 behavior with Ga_2O_3. Finally, the ternary system (UO_2- PuO_2-Ga_2O_3) will be evaluated based on the information obtained from study of the binary systems.

The results of the gallium sintering studies and the phase relations assessment achieved during the course of the fiscal year will be summarized in a single report.

Milestones:

- Issue Gallium Sintering Study Test Plan (December 1997)
- Issue Gallium Sintering Study Test Report (September 1998)

4.0 Analytical Methods Development

The continued development of analytical techniques used in conjunction with the fabrication of MOX fuel is considered necessary for several areas this fiscal year. The technical tasks included in this activity are:

- Continue development of a MXRF system to measure the spatial distribution and bulk concentration of gallium in PuO_2 feedstock, unsintered MOX fuel, and sintered fuel pellets.
- Continue development and implementation of O/M measurement techniques (as resources permit).
- Assess the ability to measure gallium concentrations using the existing LIBS capability in TA-55.
- Complete implementation of autoradiography measurement techniques.

These techniques will directly support other ongoing MD program activities this year, including R&D and test fuel fabrication efforts. The MXRF and LIBS techniques will be needed by the TIGR R&D efforts, while the O/M and autoradiography measurements will be needed for both the Fuel Fabrication R&D and ATR test fuel fabrication. In addition, these techniques and the results obtained from their various analyses this year will ultimately support the successful fabrication of MOX fuel for the plutonium disposition mission.

4.1 MXRF Development

As work has progressed in ongoing R&D efforts, a need has been demonstrated for a sensitive, spatially resolved method of gallium detection. The goal of this task then is to further develop the MXRF system to measure the spatial distribution and bulk concentration of gallium in PuO_2 feedstock, unsintered MOX fuel, and sintered fuel pellets. This task is a continuation of past efforts, where MXRF has demonstrated its unique ability to provide rapid elemental information that cannot be obtained with any other analytical method in elucidating the movement of gallium in surrogate MOX fuel pellets under reducing conditions. In addition to being nondestructive and requiring minimal sample preparation, MXRF offers a number of other advantages over conventional electron microprobe techniques for acquiring elemental distributions from a sample including higher sensitivity, greater penetration depth, the ability to operate in air, and large area sample analysis. Studies performed with MXRF include: (1) elemental detection capabilities of the technique including spatial resolution, (2) demonstration of phase identification abilities, and (3) developmental work for MXRF as a more widely-known measurement technique, especially for gallium in MOX fuel surrogates.

More specific activities have been identified for this fiscal year, based on results obtained through previous experiments. A new high-power x-ray tube for the system will be purchased and installed, boosting both flux and sensitivity. The alignment of the previously installed capillary optic will be fine-tuned to gain sensitivity for gallium detection. Demonstration of the phase identification work will also continue, through

studies of Raman spectra and micro-x-ray diffraction work at Oak Ridge National Laboratory to determine the composition of unknown phases observed in previous experiments.

Milestones:

- Complete Installation of High Power X-Ray Tube (May 1998)
- Complete Installation of Monolithic Capillary (May 1998)
- Demonstrate Direct MXRF Method for Gallium Detection (September 1998)

4.2 O/M Measurement Technique Evaluation

This fiscal year, efforts will be undertaken to identify potential O/M measurement techniques for eventual use in TA-55. While the existing O/M measurement technique is sufficient for current activities, it is time-consuming, and it is likely that future efforts could require more accurate measurements. Last fiscal year, information was gathered from various sources on the techniques currently used for measurement of O/M ratio. This activity will continue this fiscal year, as resources permit. A literature survey will be performed, from which potential methods will be identified and evaluated. The ultimate result will be a recommendation of which method(s) should be pursued and implemented for use with fuel fabrication activities at TA-55.

4.3 LIBS Capability Implementation

The objective of this task is to develop an on-line method for determining trace gallium concentration in PuO_2 in real time, as the product is treated for gallium removal. Preliminary investigations suggested that LIBS (Laser Induced Breakdown Spectroscopy) was the best candidate for on-line analysis because (1) critical LIBS components can remain external to the glovebox, and (2) the technique was already in the early stages of setup in the plutonium facility. Under this effort, the system will be installed in TA-55 and demonstrated for gallium and plutonium. Gallium and plutonium standards will also be developed for use with the system. The ultimate use for this technique will be as a real-time measurement system for the TIGR R&D (see Section 5.0). In fact, this effort will continue into next fiscal year at which point it will be funded directly under the TIGR R&D task. As such, all efforts associated with this task will be integrated with the TIGR R&D activity, and plans will concurrently be developed for the system's implementation into the Phase II ARIES demonstration line.

Milestones:

- Complete On-Line LIBS Assessment (December 1997)
- Demonstrate LIBS in TA-55 (July 1998)
- Calibrate LIBS for Gallium in PuO_2 (September 1998)

4.4 Autoradiography Development

The goal of this task this fiscal year is to evaluate improvements to the current autoradiography capability at LANL and provide information that will be useful for a future MOX fabricator. A need has been identified to develop this in-house homogeneity measurement capability further, for use with R&D and test fuel fabrication activities. As part of this effort, the data obtained from the autoradiography method will be correlated with microprobe measurements to determine the method's accuracy. First, there is a very limited amount of the cellulose nitrate film typically used with this technique available. A new source and/or a new film type needs to be located for this effort to continue. A few standards of known PuO_2 particle size and distribution will be fabricated to evaluate the use

of these techniques as funding and schedule permit. Characteristics such as size distribution will then be correlated between the autoradiography and microprobe measurements as appropriate. Other experiments, such as studying the sensitivity of the autoradiography technique to subsurface plutonium relative to the alpha particle escape depth, are desired but will be postponed pending funding. The ultimate goal is to develop an autoradiograph technique for MOX fuel fabricated with surplus-weapons plutonium which is semiquantifiable and quality assurance certified as an effective homogeneity measurement technique.

Milestones:

- Complete Autoradiography Implementation (September 1998)

5.0 Gallium Removal System

The purpose of this task is to initiate the design of a Thermally-Induced Gallium Removal (TIGR) System to be built and tested during FY98 and FY99. One of the important steps in preparing surplus-weapons plutonium for use as MOX fuel in the plutonium disposition project is the removal of the gallium. In fabricating MOX fuel it is important to have consistent feed material so the sintering parameters that produce acceptable fuel pellets can be established. At high concentrations, gallium may affect the sintering behavior of the ceramic (to be studied as part of the Fuel Fabrication Development activity; see Section 3.3), as well as the possibility of adversely interacting with the fuel rod cladding during irradiation. Therefore, to ensure consistent feed and processing parameters for sintering of pellets, as well as protecting the sintering furnace and fuel rod cladding from adverse material interactions, it is important to reduce the levels of gallium in the powder. Thus, it has been decided programmatically that the removal of gallium to levels as low as reasonably achievable will be undertaken using the TIGR system. This system is a simple, low-cost method for removing the gallium from the PuO_2 powder derived from pit plutonium processed through the HYDOX system. It has a low environmental impact and should have few waste management activities associated with it. The system design will be based on the requirements for the production-scale TIGR system that will be designed, fabricated and integrated into the PDCF. The gallium removal system will be directly integrated with the HYDOX system and must produce oxide that meets MOX fuel fabrication requirements.

This R&D activity is divided into two major tasks: (1) process development which leads to the prototypic design, and (2) prototypic design and testing. Both of these tasks will be accomplished through testing with the use of cold surrogate (CeO_2) and hot (PuO_2) feed materials. Following feed fabrication development and validation, the surrogate feed material will be used to verify furnace material compatibility and process material handling for the prototypic equipment. The PuO_2 feed material will be used to demonstrate adequate gallium separation and to optimize process operating conditions. This entire activity is a two-year effort culminating in the final prototypic demonstration of the TIGR process. The specifics of the FY99 effort, however, are not addressed in this document.

Milestones:

- Issue TIGR R&D Test Plan (January 1998)

5.1 Process Development

This task will focus on several areas to demonstrate the TIGR process adequacy and to aid in the selection of prototypic processing equipment and processing conditions. To accomplish these goals, an adequate supply of well characterized prototypic surrogate (CeO_2) and PuO_2 feed material must be produced. Surrogate testing will then be performed

to determine the phase structure of cerium-gallium oxide and the rate limiting mechanisms of gallium oxide evolution from cerium oxide. Testing with PuO_2 will be performed to validate the surrogate applicability. In addition, PuO_2 testing will be performed to begin the determination of prototypic processing conditions.

To conduct both the process development and prototypic testing tasks, adequate supplies of both surrogate-based and plutonium-based feed material must be fabricated. It is essential that this material, for both surrogate-based and plutonium-based feedstock, be as representative of prototypic material as possible. For this reason, multiple surrogate fabrication processes will be evaluated. For the surrogate-based efforts, several different methods for producing the CeO_2 will be evaluated to obtain a feed material with the proper phase structure and particle morphology. For the plutonium-based efforts, there are three sources of weapons-based PuO_2 containing gallium currently being considered (LLNL 3-step, LLNL 2-step, and directly oxided material). This feed material in general must be characterized with regard to gallium concentration and particle morphology.

The main objective of the process development effort is the selection of prototypic processing conditions. These process conditions will dictate the type of processing equipment and materials required. The plutonium-gallium oxide phase structure will dictate the maximum amount of gallium which can be separated by the TIGR process. While a single Ga_2O_3 phase should be easily reduced with hydrogen to the volatile Ga_2O species, a more complex phase with both plutonium and gallium in solution might not be amenable to Ga_2O formation. Currently, a method for characterizing the phase structure of plutonium-gallium compounds at gallium concentrations less than 10 wt% has not been found. Consequently, determining the phase structure for prototypic weapons-based material is not yet possible. However, it is possible to determine the CeO_2 surrogate phase structure with prototypic gallium concentrations of less than 1 wt%. In light of this information, surrogate phase structure studies will be performed to ascertain the thermodynamic similarities between cerium-gallium oxide and plutonium-gallium oxide, at gallium concentrations amenable to phase structure analysis for both the surrogate and plutonium systems. Following validation of the cerium and plutonium phase structure similarities, conclusions drawn from surrogate phase structure analysis at prototypic gallium concentrations will be extrapolated to the plutonium phase structure to provide data regarding the theoretical maximum gallium separation from plutonium by the TIGR process. Prototypic PuO_2 will then be used to establish the processing conditions which are required for the system design. A separate test plan will be issued detailing the processing parameters to be studied (time, temperature, gas velocity, gas partial pressure, and batch size). Characterization will include gallium concentration and the particle morphology (i.e. size distribution and surface area).

The CeO_2 will be validated for acceptance as a surrogate by duplicating selected tests with PuO_2. This approach will allow leveraging of the optimization data for surrogate validation. The PuO_2 process optimization test conditions will be defined in a separate test plan. A balanced selection of the most significant test conditions will be chosen for duplication with the PuO_2 feed.

Milestones:

- Issue Surrogate Validity Report (July 1998)

5.2 Prototype Design and Testing

The prototype design effort will be focused on two primary activities. First, a system design will be selected which can provide the process conditions selected from the process development test results. As much as possible, this system design will consist of off-the-

shelf equipment. Second, materials will be selected which allow operating the selected system at the design conditions. In particular, this relates to the concern over gallium reactivity with system components and any moving parts at elevated temperatures. The prototypic testing will consist of (1) cold checkout phase, (2) hot checkout phase, and a (3) prototypic demonstration. These tests will be conducted during FY99 and are not discussed here, although the test planning will be initiated later in FY98.

Particular attention must be paid to both mechanical and material considerations when designing a new system. For example, if it is determined that a fixed-bed device cannot provide adequate gas/particle mixing, then the design will become more complicated, possibly employing an inclined-rotary continuous-type device. This will add complexity to the design with regard to including more moving parts at elevated operating temperatures. In addition, locating an off-the-shelf design for TIGR operating temperatures while matching the processing rate and satisfying the criticality requirements will be challenging. Two additional questions arise with regard to materials: (1) what furnace fabrication materials must be changed if the off-the-shelf equipment is operated at significantly higher than original design conditions, and (2) what will be the effect of highly reactive gallium on the furnace system materials? Once a system is selected, a formal design review will be held to ensure that issues such as these are adequately addressed. Because of schedule constraints the final design will be done in parallel with process development. This means the design will be conservative and include all features potentially needed to optimize gallium removal. The tradeoff will be higher costs but faster delivery of a finished working system.

Milestones:

- Issue Final Systems Requirement Document (April 1998)
- Initiate System Design Review (July 1998)

6.0 R&D Coordination/Integration

This activity has been included to allow for overall planning, coordination, integration, monitoring, and reporting of the Nuclear Fuels Technologies Project R&D activities at LANL. In general, staff assigned to this task will provide programmatic approval of R&D project deliverables and oversight of activities. They will serve as the primary interface between R&D project activities and staff of other program participants, the PDCF project, and with LANL senior management staff. Under this task, a report will be issued that summarizes the results of the FY97 R&D activities. In addition, an FY98 R&D Test Plan will be published, and monthly progress reports will be issued.

Milestones:

- Issue FY97 R&D Summary Report (October 1997)
- Issue FY98 R&D Test Plan (November 1997)

7.0 Schedule and Milestones

A summary of the FY98 research and development milestones for the Nuclear Fuels Technologies Project is given in Table III. An overall schedule of activities for FY98 is provided in Figures 1 and 2.

Table II. Fiscal Year 1998 R&D Milestone Summary

Applicable Section	Milestone	Expected Completion Date
2.2	Complete MD MOX Feed Database Architecture	November 1997
2.3	Obtain PuO_2 for R&D and Test Fuel Fabrication	January 1998
2.3	Obtain AUC UO_2	January 1998
2.4	Issue Draft MD PuO_2 Feed Specification	June 1998
3.1	Complete AUC UO_2 Baseline Development Plan	November 1997
3.1	Complete Baseline Process Development Report	August 1998
3.2	Complete Alternate PuO_2 Feed Test Plan	November 1997
3.3	Issue Gallium Sintering Study Test Plan	December 1997
3.3	Issue Gallium Sintering Study Test Report	September 1998
4.1	Complete Installation of High Power X-Ray Tube	May 1998
4.1	Complete Installation of Monolithic Capillary	May 1998
4.1	Demonstrate MXRF Method for Ga Detection	September 1998
4.3	Complete On-Line LIBS Assessment	December 1997
4.3	Demonstrate LIBS in TA-55	July 1998
4.3	Calibrate LIBS for Gallium in PuO_2	September 1998
4.4	Complete Autoradiography Implementation	September 1998
5.0	Issue TIGR R&D Test Plan	January 1998
5.1	Issue Surrogate Validity Report	July 1998
5.2	Issue Final Systems Requirement Document	April 1998
5.2	Initiate System Design Review	July 1998
6.0	Issue FY97 R&D Summary Report	October 1997
6.0	Issue FY98 R&D Test Plan	November 1997

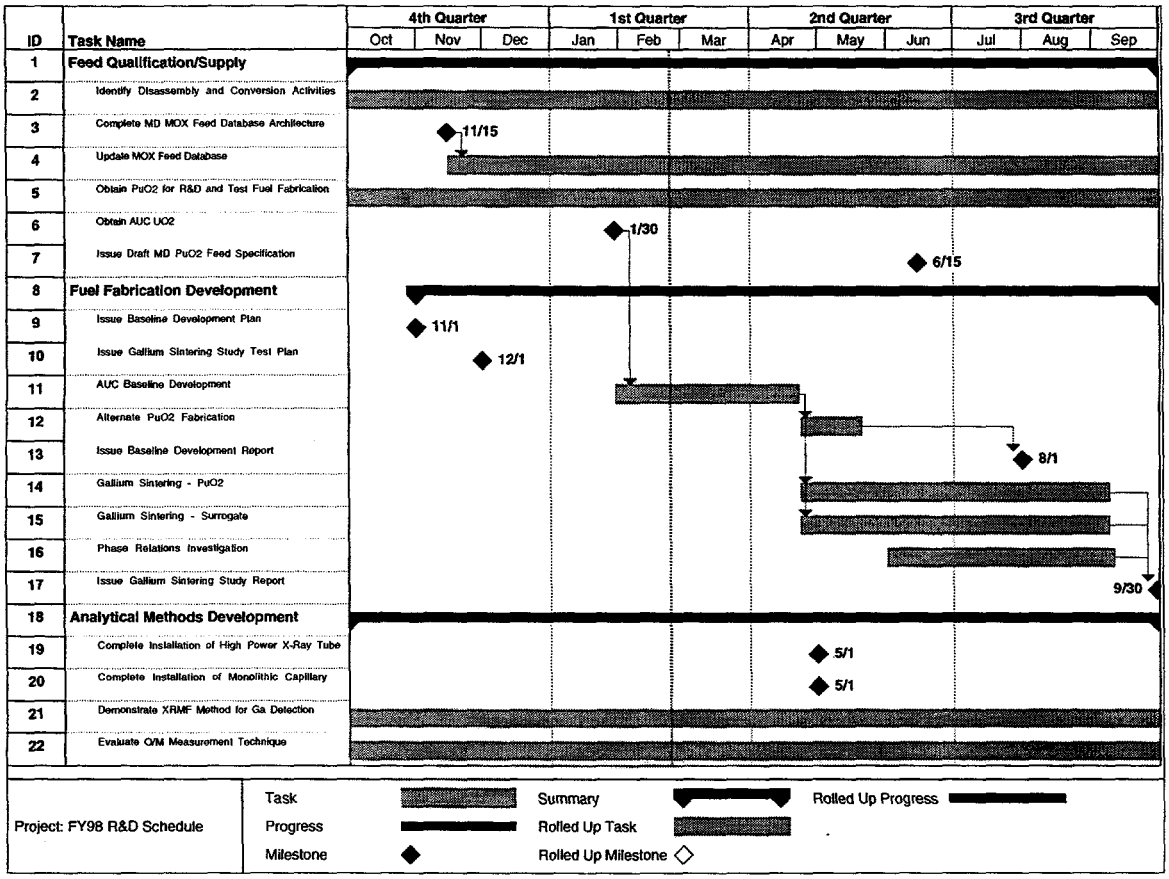

Figure 1. Schedule of Fiscal Year 1998 R&D Activities (page 1)

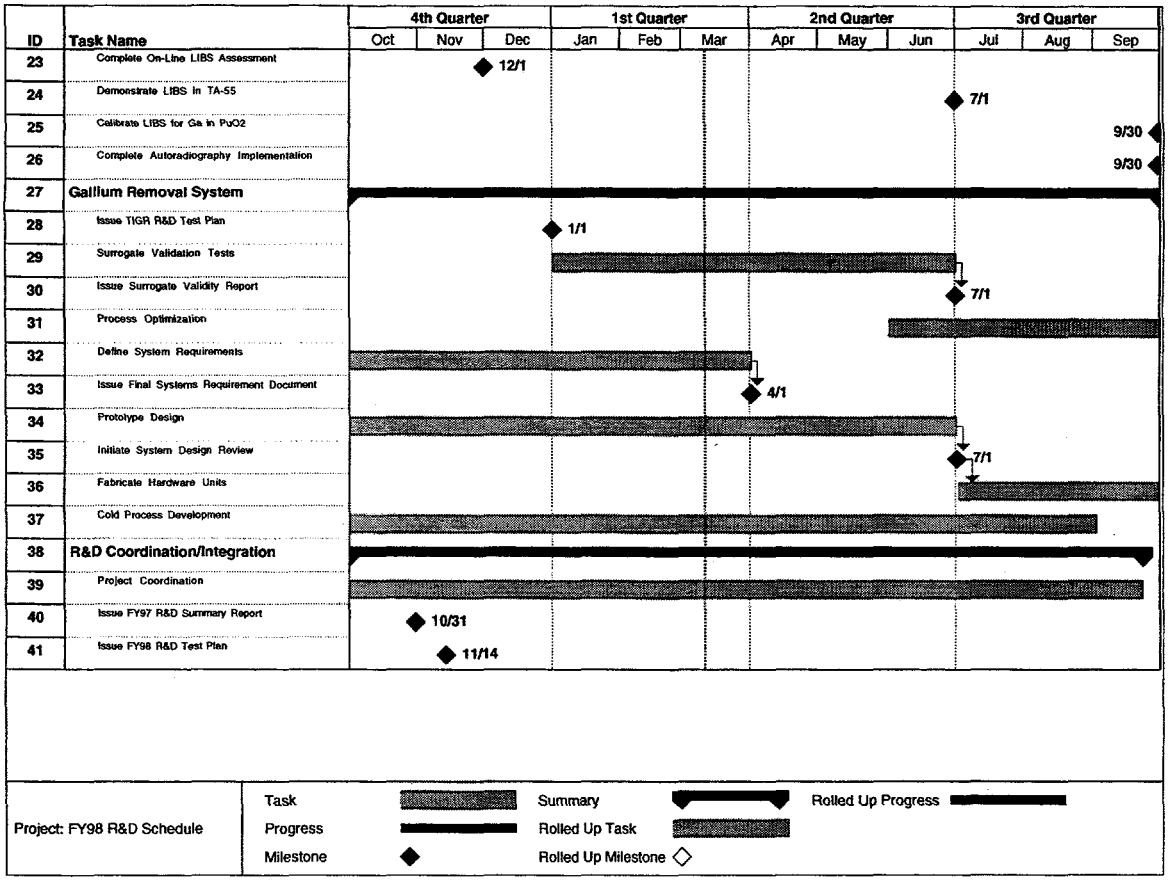

Figure 2. Schedule of Fiscal Year 1998 R&D Activities (page 2)