Preprint
UCRL-JC-137917

Nondestructive Evaluation and Assay for the Plutonium Ceramification Test Facility

M. Mitchell, D. Pugh, T.-F. Wang

This article was submitted to
Spent Fuel and Fissile Material Management, San Diego, CA., June 4-8, 2000

U.S. Department of Energy

Lawrence
Livermore
National
Laboratory

March 7, 2000

This work was performed under the auspices of the United States Department of Energy by the University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

NONDESTRUCTIVE EVALUATION AND ASSAY FOR THE PLUTONIUM CERAMIFICATION TEST FACILITY

Mark Mitchell[1], LLNL
Darrell Pugh, LLNL
Tzu-Fang Wang, LLNL
Livermore, California*

INTRODUCTION

Lawrence Livermore National Laboratory (LLNL) has conducted design and testing activities of the Nondestructive Assay/Evaluation (NDA/NDE) system that will be installed to support the Plutonium Ceramification Test Facility (PuCTF). PuCTF immobilizes plutonium using the ceramic can-in-canister technology. The overall function of the NDA/NDE System is to ensure that sintered pucks contain the appropriate materials for ceramification process control, special nuclear materials (SNM) accountability, and repository acceptance. The system accepts sample pucks from the ceramification system, performs measurements, and determines if the product pucks are acceptable. This report details the conceptual system that is being developed.

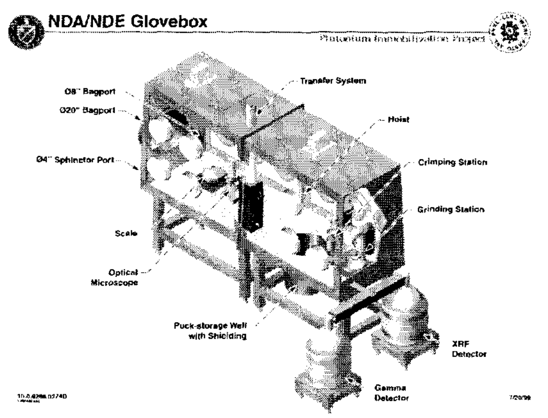

Figure 1. NDA/NDE system conceptual glove box

Objectives

The NDA/NDE system will provide a range of analytical capabilities that will provide quantitative data in support of the PuCTF operations. Data to be provided includes: 1) puck dimensions and density, 2) puck integrity assurance, 3) puck elemental composition including presence of major matrix components and neutron poisons, 4) mineralogy, and 5) puck actinide / fissile material loading. This information assures that the repository data requirements are satisfied and validates the Product Control Model (PCM) that will be used for process control. This information is also required for Materials Control & Accountability (MC&A). The NDA/NDE system will develop analytical techniques and equipment for use in the plant design.

Techniques

Four techniques are used in the NDA/NDE system. First, X-ray fluorescence (XRF) is used for identifying and quantifying major matrix components and neutron poisons. In the case of XRF analysis, two excitation options are available: 1) self-induced fluorescence by the alpha activity from the actinide decay or 2) external excitation. Self-induced fluorescence has the advantage of providing efficient excitation of the K shell x-rays of the higher Z elements, such as Hf and Gd, which can be measured simultaneously with the gamma ray spectra. Externally excited x-rays are more effective for measuring the lower Z materials, such as Ca and Ti, which have softer x-ray signatures. Second, gamma ray spectroscopy is used for determining isotopics and actinide loading/fissile material content. This technique is based on analyzing the active gamma ray emissions of the various isotopes of Pu and U. Third, laser dilatometry is used for determining puck dimensions and density. Fourth, X-ray diffraction (XRD) is used to validating mineralogy of the ceramic. All NDE activities pertaining to XRD are conducted by SRTC, and thus are not covered in this paper. As a rough check on mineralogy and phase

[1] Author to whom questions should be addressed.

formation, an optical microscope x100-x3000 was used by LLNL.

Overview of Research

This project has several goals. These studies were pursued by first testing surrogate ceramics, then uranium, and finally plutonium. The techniques must be nondestructive and well suited for a glovebox environment. Since detectors have a high maintenance requirement and use liquid nitrogen, this requires removing as many components as possible from the interior of the glovebox. This goal was met by using self-induced fluorescence and sealed source excitation for XRF, in conjunction with a 50 mm^2 SiLi detector, instead of a traditional all-in-one research XRF containing an x-ray tube and secondary targets. Preliminary tests investigated several excitation sources. As a result of these tests, ^{109}Cd was found to be the best exciter for Hf, Gd, Ce, and Ti, with a lesser ability to excite Ca in quantitative amounts. ^{55}Fe is a good exciter for Ca and Ti. Used in conjunction, ^{55}Fe and ^{109}Cd selectively excite the key matrix elements and neutron poisons, namely, Ca, Ti, Hf, and Gd. A tight ring configuration is optimal for location external to the glovebox.

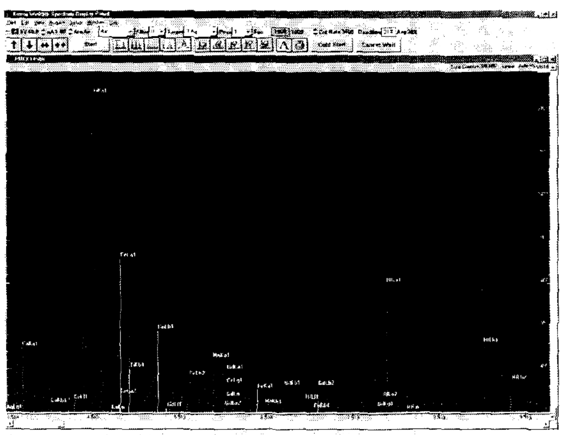

Figure 2. XRF spectrum of Hf/Ce/Ce puck using both ^{55}Fe and ^{109}Cd radioisotope sources.

The second goal is to optimize plant design by using a single detector for gamma ray spectroscopy, needed for isotopics and actinide loading, and measurement of self-induced K shell x-rays, needed for quantifying neutron poisons. A planar 16 mm HPGE detector, with a thin Be window, was selected for its ability to detect the x-rays and gamma rays of interest. The only operation necessary to convert from one measurement to the other is the insertion of an absorber.

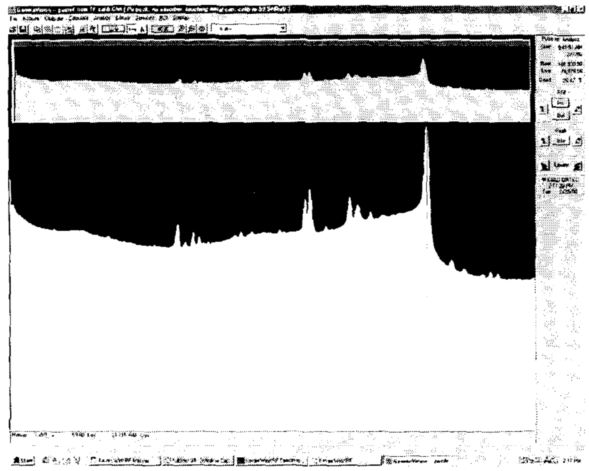

Figure 3. Spectra of Hf/Pu/U puck - preliminary self-excitation XRF experiment with Pu. HPGe planar detector.

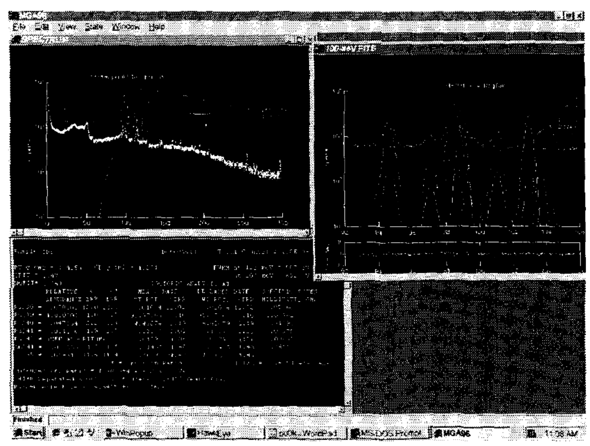

Figure 4. Spectra and MGA data of Hf/Pu/U puck, using Cd absorber. HPGe planar detector.

In order to achieve this result, a system was built to test surrogate and uranium pucks, and prototype the arrangement for the Pu glovebox.

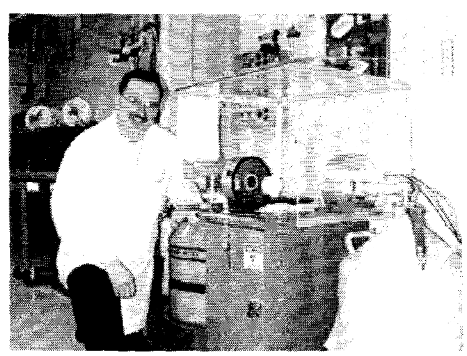

Figure 5. Detector setup for surrogate and uranium tests.

Finally, a glovebox was designed and fabrication commenced to support PuCTF.

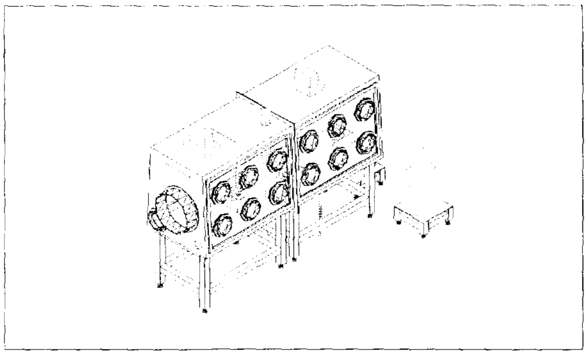

Figure 6. Glovebox isometric view

CONCLUSION

Over the course of two years, investigations moved forward from initial surrogate and uranium measurements to actual tests on plutonium ceramic pucks. Nondestructive techniques were developed to provide data on major matrix elements, neutron poisons, SNM loading, and isotopics. Forthcoming tests on Pu pucks in optimal conditions will provide XRF spectra of Ca, Ti, Hf, and Gd in a region swamped by the Compton effects of decaying ^{241}Am in aged plutonium. The Plutonium Immobilization Plant will be able to nondestructively evaluate and assay the ceramic pucks in a manufacturing, in-line setting.

ACKNOWLEDGEMENTS:

*Work performed under the auspices of the U.S. Department of Energy by the University of California Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

REFERENCES:

Guy Armantrout. *Assessment of NDE Measurements For the In-Line Analysis of Sintered Pu Pucks: Evaluation of X-Ray Fluorescence Analysis For NDE Product Characterization And Process Control.* (Milestone 6.5a), Plutonium Immobilization Project September 30, 1998.

Bonner et al. *Trace Element Analysis Using X-ray Fluorescence.* Chemical Instrumentation, 6(1), pp. 1-36 (1975).

Mark Mitchell, *Conceptual Design for PuCTF NDE Support System (Project Milestone 2.6.4.a)*, Lawrence Livermore National Laboratory, Livermore, CA, PIP-99-081. July 22, 1999

Mark Mitchell, Art Jurgensen. *Draft System Design Description for NDA/NDE (Rev. A): Module 2.6.4, Puck NDE for Process Control, and Module 2.6.5 SNM Accountability.* Lawrence Livermore National Laboratory, Livermore, CA. December 14, 1999.