LA-UR- 00-405

Approved for public release; distribution is unlimited.

Title: Analysis Capabilities for Plutonium-238 Programs

Author(s): A.S. Wong, G. H. Rinehart, M. H. Reimus, M. E. Pansoy-Hielvik, P. F. Moniz, J. C. Brock, S. E.Ferrara, and S. S. Ramsey

NMT-9

Submitted to: Plutonium Futures-The Science
Santa Fe, New Mexico
July 10-13, 2000

Los Alamos
NATIONAL LABORATORY

Analysis Capabilities for Plutonium-238 Programs

A.S. Wong, G. H. Rinehart, M. H. Reimus, M. E. Pansoy-Hielvik,
P. F. Moniz, J. C. Brock, S. E.Ferrara, and S. S. Ramsey

Nuclear Materials Technology Division
Los Alamos National Laboratory
Los Alamos, NM 87545

Introduction

In the past two decades, Los Alamos National Laboratory (LANL) has produced general-purpose heat sources (GPHS) from plutonium-238 oxide for space and other power source applications. One of the most recent GPHS applications was in the Cassini Spacecraft for Saturn exploration[1]. The Power Source Technologies Group (NMT-9) has full capabilities to recover and purify $^{238}PuO_2$ from scrap and aged fuels,[2] and to fabricate oxides into fuel pellets for heat sources.[3]

In this presentation, an overview of analysis capabilities that support ^{238}Pu programs will be discussed. These capabilities include neutron emission rate and calorimetric measurements, metallography/ceramography, ultrasonic examination, particle size determination, and chemical analyses. The data obtained from these measurements provide baseline parameters for fuel clad impact testing, fuel processing, product certifications, and waste disposal. Also several in-line analyses capabilities will be utilized for process control in the full-scale ^{238}Pu Aqueous Scrap Recovery line in FY2001.

Physical Measurement Capabilities

Neutron Emission Rate Measurements. Spontaneous fission of ^{238}Pu produces approximately 2220 neutrons per second per gram (n/s/g) of PuO_2 (at 81% of ^{238}Pu).[4] Energetic alpha particles react with light isotopes such as ^{17}O, ^{18}O, and ^{19}F in PuO_2 producing additions of 5000 to 20,000 n/s/g via (alpha, n) reactions. In order to reduce the neutron emission rate, a minimum amount of hydrofluoric acid is used in the aqueous process, and the fuel oxide is treated in an ^{16}O exchange process to reduce ^{17}O and ^{18}O. The neutron emission rate of oxide is then measured on a thermal neutron counter[5]. The emission rate in the final products (purified oxides and fuel powder) must be less than 7000 n/s/g of ^{238}Pu.

Calorimetric Measurements. Calorimetry is used for determining the power output of heat producing materials. ^{238}Pu has a half-life of 87.74 years and a power output of 0.567 watt per gram. Several types of calorimeters are utilized in the ^{238}Pu lab to measure low- (0 to 5 W with an accuracy of ±0.003 W) and high-wattage (0 to 200 W with an accuracy of ± 0.5 W) fuel.[6] Calorimetry is used to verify the amount of special nuclear materials in incoming fuel materials, outgoing scrap, and finished heat source assemblies.

Metallography/ceramography. Metallographic/ceramographic examinations are performed on test components recovered from impact tests to determine possible failure mechanisms. The microstructure of the GPHS clad material, girth welds, and samples of fuel pellets are also examined. A LECO 300 metallograph with magnification range of 8 to 500 X is interfaced to the glovebox line through a unique hood extension that covers but does not enclose the metallographic stage. Metallographic/ceramographic specimens are prepared with standard metallographic equipment. This includes a wafering saw, automated grinding, rough polishing and fine polishing equipment, and a power supply for electrolytic etching.

Ultrasonic Examination. Ultrasonic testing is performed to examine the weld integrity of the $^{238}PuO_2$-fueled clad and simulant-fueled capsules. The instrument we use to perform ultrasonic testing is a 3.5-MHz transducer inspecting system with a 0.5-inch diameter and a 1.5-inch spherical focus in water. The operator must have a minimum of an ASNT Level I ultrasonic testing certification.

Particle size analysis. NMT-9 has in-line capabilities for determining the particle size distribution of fines that are less than 100 micron recovered from impact tests. Impact tests are conducted to determine the response of GPHS to probable launch accident scenarios. Particle size analysis is also used to verify the particle size of milled oxide prior to dissolution. The instrument we use to perform particle size analysis is the Galai CIS-100. Its measurement range is 0.5 to 600 micron.

Chemical Analysis Capabilities

Chemical data of ^{238}Pu samples (feed oxides, purified oxides, granular ^{238}Pu, and process solutions) provide necessary baseline parameters and measures for process control, material control and accountability, waste disposal, and product certification. Our chemical analysis are available through collaborative effort with NMT-1 Analytical Chemistry Group.

The purity of plutonium oxide is determined by Pu(III) visible spectrometry. If expected Pu content is low (μg or less), gross alpha counting is used to calculate the ^{238}Pu content. Actinide impurity analyses including ^{234}U, ^{241}Am, ^{237}Np, and ^{236}Pu are determined by radiochemical methods (gross alpha and gamma counting, gamma and alpha spectroscopy, and radionuclide separations.) The plutonium isotopic composition is determined by thermo-ionization mass spectrometry (TIMS). Direct-current arc (DC Arc) and inductively coupled plasma mass spectrometry (ICP-MS) techniques are used to determine non-actinide cationic and anionic impurities.

Detailed description of each chemical measurement capability will be given in the presentation.

In-Line/On-Line Process Monitoring

In FY 2001, the full-scale ^{238}Pu Aqueous Recovery and Purification line will be operational at LANL. We are currently installing an on-line gamma system that will monitor Am, U, and Pu gamma rays during the ion-exchange process[7]. The on-line gamma monitoring system will provide real-time elution profiles of actinide impurities that are important for plutonium loss, waste minimization, and process control.

In addition, a solution in-line alpha counter (SILAC) will be used for in-line monitoring of alpha activity in hydroxide filtrate during the full-scale production. By knowing the approximate alpha concentration, we can adjust the operating parameters of the ultra-filtration process to maximize removal of plutonium and uranium from the waste solutions. Farnham and Fowler at LANL[8] developed this SILAC system for real-time monitoring of plutonium and americium concentration in process solutions in the glovebox lines.

Summary

The Plutonium Facility at LANL is the only place left in the United States with full capabilities of producing ^{238}Pu heat sources. It is very important for us to maintain and improve our valuable resources in order to support future space applications.

Acknowledgment

We thank Liz Foltyn, Tim George, and Kevin Ramsey for leading the efforts discussed here. We also thank NMT-1 Analytical Chemistry Group for providing analytical chemistry results for oxides and process solutions; NMT-4 Material Accountability Group for providing calorimetric measurements; and all the personnel who support ^{238}Pu programs.

References

1. T. G. George, and E. M. Foltyn, "Production of 238PuO2 heat sources for the Cassini mission," AIP Conference Proceedings (CONF-980103), Jan 1998.

2. L. D. Schulte, G. H. Rinehart, J. Espinoza, G. L. Silver, K. B. Ramsey, K, G. D. Jarvinen, G. M. Purdy, "Recycle of scrap plutonium-238 oxide fuel to support future radioisotope applications," AIP Conference Proceedings (CONF-980103), Jan 1998.
3. G. H. Rinehart, "Light weight radioisotope heater unit (LWRHU) production for the Cassini mission," AIP Conference Proceedings (CONF-970115), Jan 1997.
4. G. M. Matlack, and C. F. Metz, "Radiation Characteristics of Plutonium-238," LA-3696, Los Alamos National Laboratory, 1967.
5. N-1 Application Note, "Passive neutron coincidence counters," LALP-92-52, Los Alamos National Laboratory, 1992.
6. NIS-5 Application Note, "Heat-Flow Calorimeters," LALP-99-25, Los Alamos National Laboratory, 1999.
7. A. S. Wong, K. B. Ramsey, and M. E. Pansoy-Hjelvik, "On-line Gamma Monitoring System for the ^{238}Pu Aqueous Recovery Process," ANS Transactions, 15 (81) 1999.
8. J. Farnham and M. M. Fowler, "SILAC Assembly Manual," Los Alamos National Laboratory, August 1998.

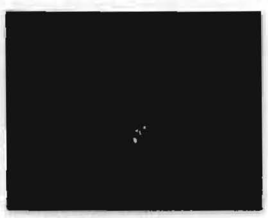
LA-UR-00-405

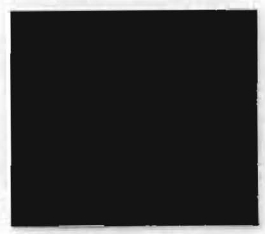
Analysis Capabilities for Plutonium-238 Programs

A. S. Wong, G. H. Rinehart, M. H. Reimus, M. E. Pansoy-Hjelvik, P. F. Moniz, J. C. Brock, S. E. Ferrara, and S. S. Ramsey

Nuclear Materials and Technology Division

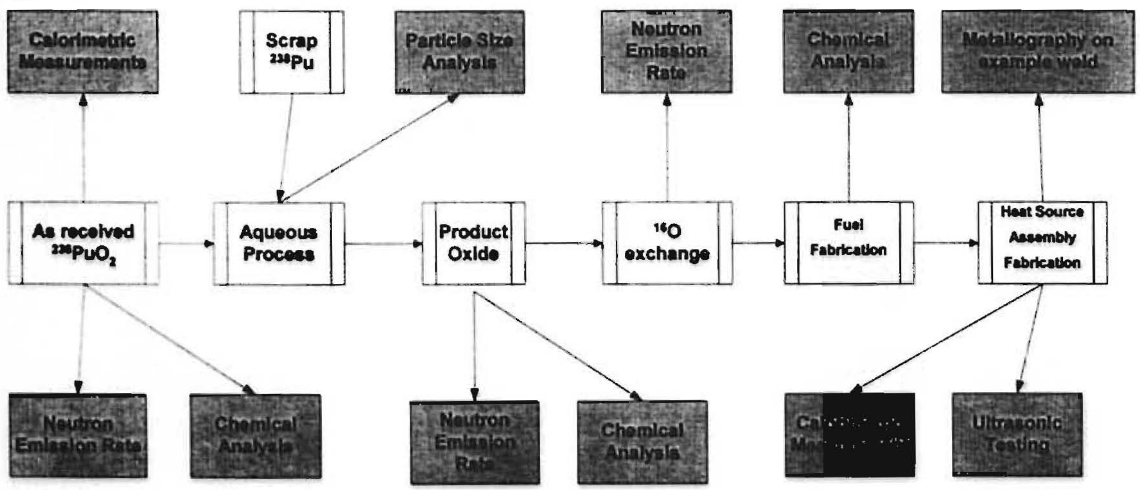
Los Alamos National Laboratory





Introduction

Plutonium-238 Group (NMT-9) at Los Alamos Plutonium Facility has full capabilities to recover and purify $^{238}PuO_2$ from scrap and aged fuels (*aqueous scrap recovery*), to fabricate oxides into fuel pellets, and to assemble into general purpose heat source (GPHS). An overview of analysis capabilities that support ^{238}Pu programs are presented here.



^{238}Pu Heat Source Fabrication

Impact Testing

Chemical Analysis of ^{238}Pu Materials

Chemical data of ^{238}Pu materials (feed oxide, purified oxides, granular ^{238}Pu, and process solutions) provide necessary baseline parameters and measures for process control, material control and accountability, waste disposal, and product certifications.

Analytical Chemistry Group provides information on plutonium assay, actinide analysis (^{234}U, ^{241}Am, ^{237}Np, and ^{236}Pu), Pu isotopic composition, and non-actinide cationic and anionic impurities.

^{238}Pu group has several in-line analyses capabilities to provide real-time chemical information. These include SILAC (Solution In-Line Alpha Counter) and PPM (Plutonium Process Monitoring) System.

SME: Amy Wong of NMT-9 and Analytical Chemistry Group

Chemical Analysis-Methodology

Plutonium assay: utilize Pu(III) visible spectrometry

Actinide analysis: Radiochemical methods

- ^{241}Am by high resolution gamma-ray spectroscopy
- ^{237}Np and ^{234}U by ion-exchange, gross alpha & alpha spectroscopy
- ^{236}Pu by alpha spectroscopy
- thorium by inductively coupled plasma mass spectrometry (ICP-MS)

Isotopic composition: thermal ionization mass spectrometry

Non-actinide cationic impurities: direct-current arc (DC Arc)

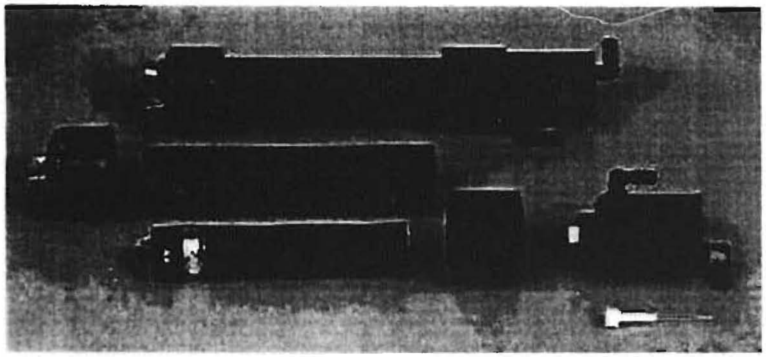
(Al, B, Be, Ca, Cd, Cr, Cu, Fe, Mg, Mn, Mo, Na, Ni, Pb, Si, Sn, Zn)

Anionic impurities: phosphorus by ICP-MS

SME: Amy Wong of NMT-9 and Analytical Chemistry Group

In-Line Chemical Monitoring Systems

SILAC



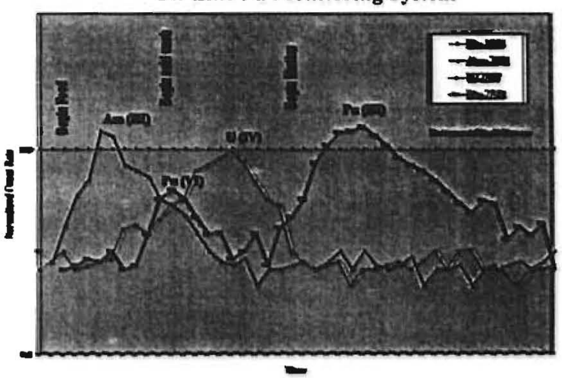
Provide alpha concentration in line without sample-out for analysis

- **Semi-quantitative analysis**
- **Large dynamic range for Pu detection: ~0.1 to 6000 mCi/L**

Monitor ^{241}Am, ^{237}U, ^{239}Pu, ^{238}Pu, and ^{241}Pu gamma rays during aqueous scrap recovery process. Provide real-time trend plot of loading, washing and elution profiles.

Provide information for process monitoring and improvement. Help to minimize solution volume and waste generation.

On-Line Pu Monitoring System



SME: Amy Wong

Plutonium Process Monitoring (PPM) System

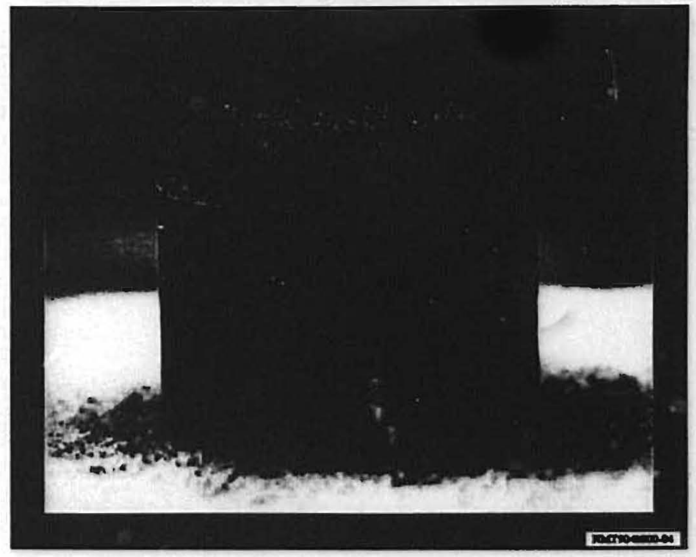
Calorimetric Measurements

^{238}Pu, $t_{1/2}$ = 87.74 years

Power output = 0.567 W/g

Two types of calorimeters used in ^{238}Pu laboratory:

- **Low wattage, 0 to 5 W ± 0.003 W**
- **High wattage, 0 to 200 W ± 0.5 W**



Calorimetry is used to verify the amount of special nuclear materials in incoming fuel materials, outgoing scrap, and finished heat source assemblies.

SME: NMT-4/NIS-5 Calorimetric Measurement Team

Neutron Emission Rate (NER) Measurements

Spontaneous fission of ^{238}Pu produces ~ 2220 n/s/g of PuO_2 (at 81% of ^{238}Pu). Energetic alpha particles react with light isotopes such as ^{17}O, ^{18}O, ^{19}F in $^{238}PuO_2$ producing additions of 5000 to 20,000 n/s/g via (alpha, n) reactions.

The neutron emission rate is measured in a thermal neutron counter.

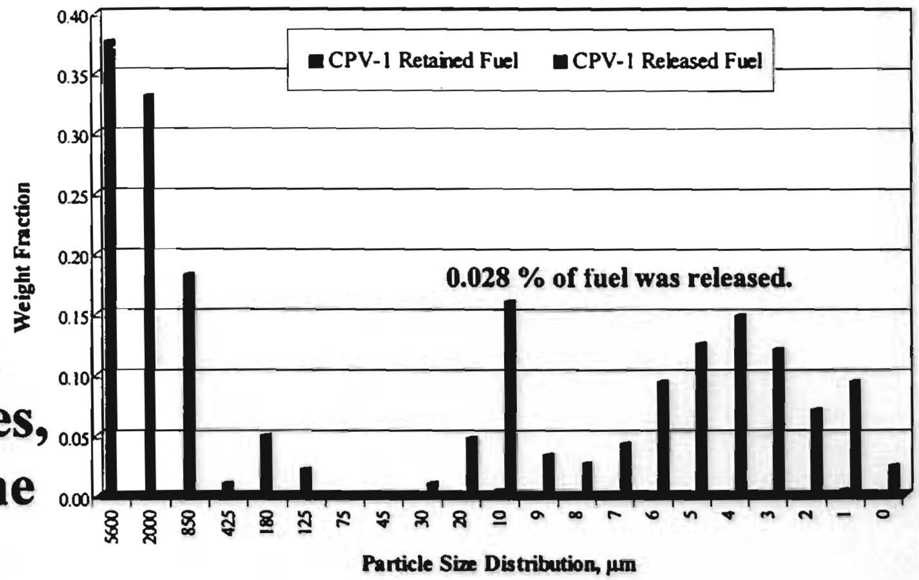
Neutron Emission Rate (n/s/g oxide)		
Fuel Fabrication	Fuel lots as received	^{16}O Exchanged
Lot # L620	18803	4459
Lot # L641	19884	9881
Aqueous Recovery	Oxalate Precipitated	Ion-Exchange Oxalate Precipitated
ASPPDTQ1	18671	12756
ASPPDTQ5	22558	13319

NER data for ^{238}Pu Materials in aqueous (IX and oxalate) and fuel fabrication lines (^{16}O)

SME: Jason Brock, John Brown, Gary Rinehart

Particle Size Analysis

Utilize Galai CIS-100 instrumentation to determine particle size distribution (0.5 to 600 micron) of ^{238}Pu and simulant fuels from impact testing (mainly fine particles, < 100 micron) and verify the particle size of milled ^{238}Pu oxide prior to dissolution for scrap recovery.



Particle size analysis of impact testing results (velocity at 55 m/s) of a hot- pressed simulant fuel (urania pellets)

SME: MaryAnn Reimus, Paul Moniz

Ultrasonic Test (UT)

UT is performed to examine the weld integrity of the $^{238}PuO_2$-fueled clad and simulant-fueled capsules. The cracks are not visible on the external surface of the weld, and if left undetected, could compromise the containment of $^{238}PuO_2$ in the unlikely event of an aborted launch or re-entry into the earth's atmosphere.

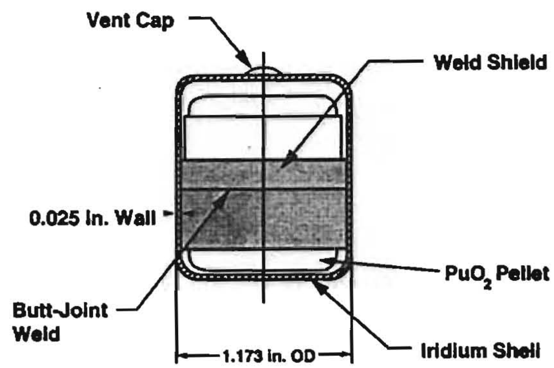
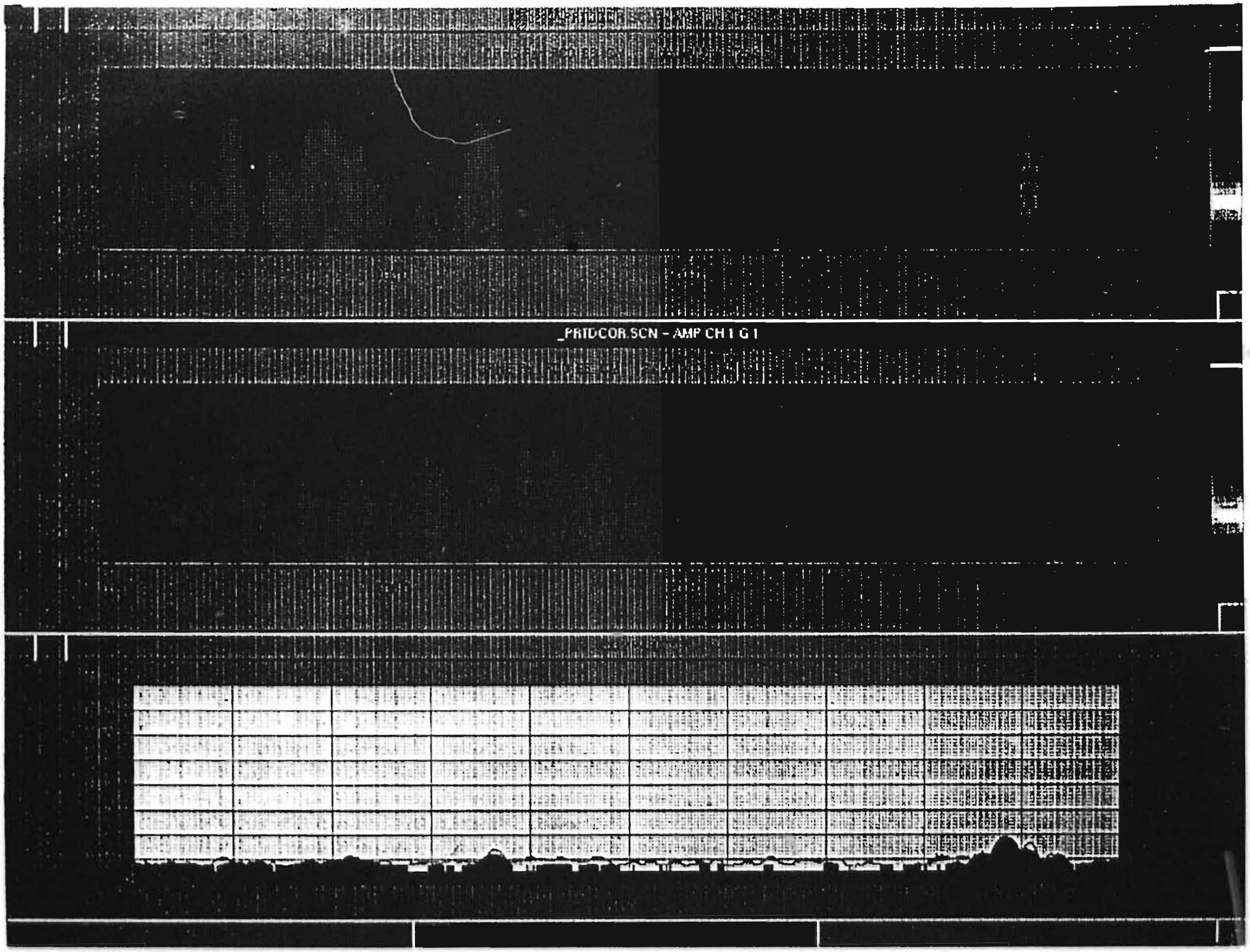


Figure 1. General-Purpose Heat Source (GPHS)

We use a 3.5-MHz transducer inspecting system with a 0.5-inch diameter and 1.5-inch spherical focus in water. The operator must have a minimum of an ASNT Level-I ultrasonic testing certification.

SME: MaryAnn Reimus, Paul Moniz

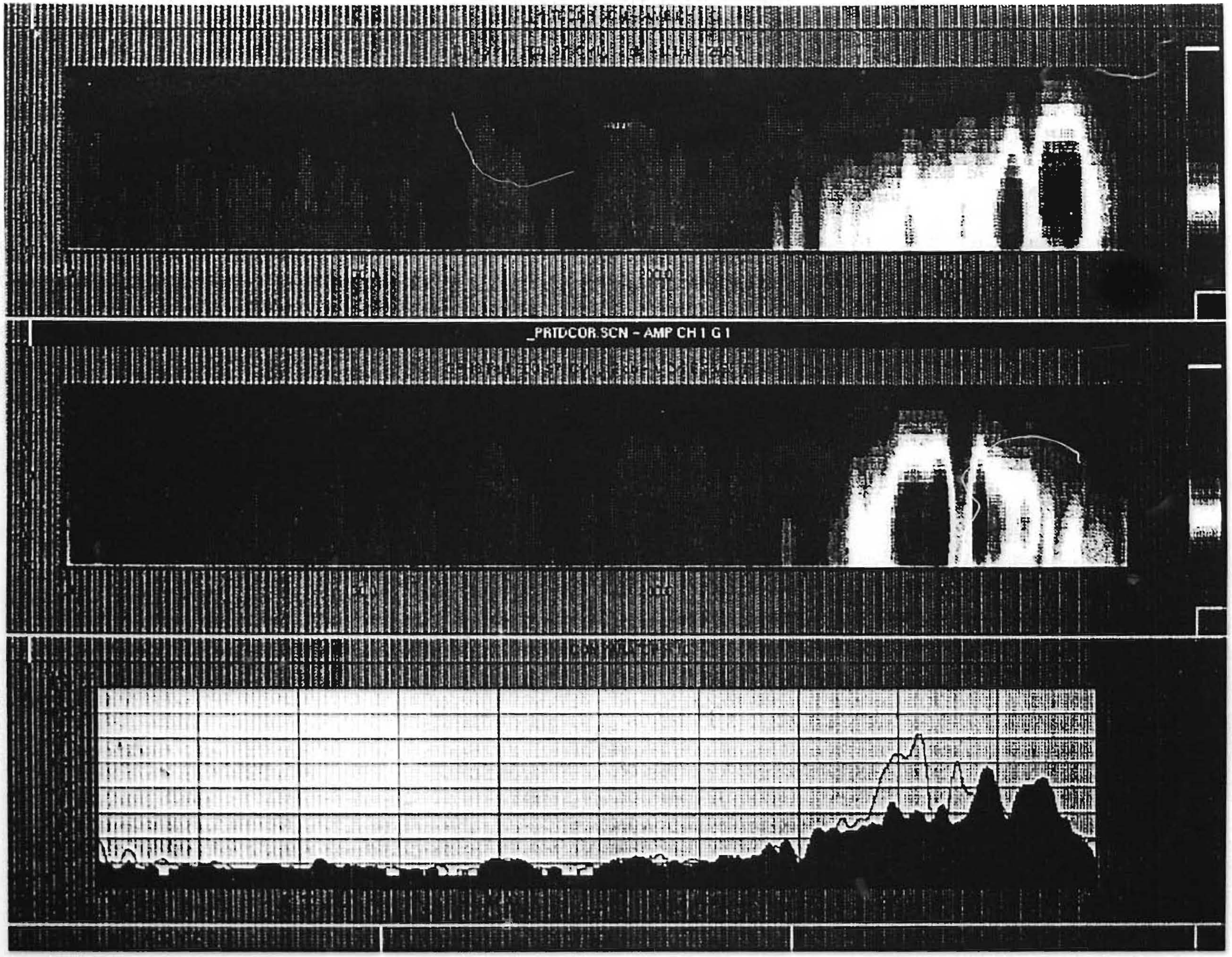

_PRTDCOR.SCN – AMP CH1 G1

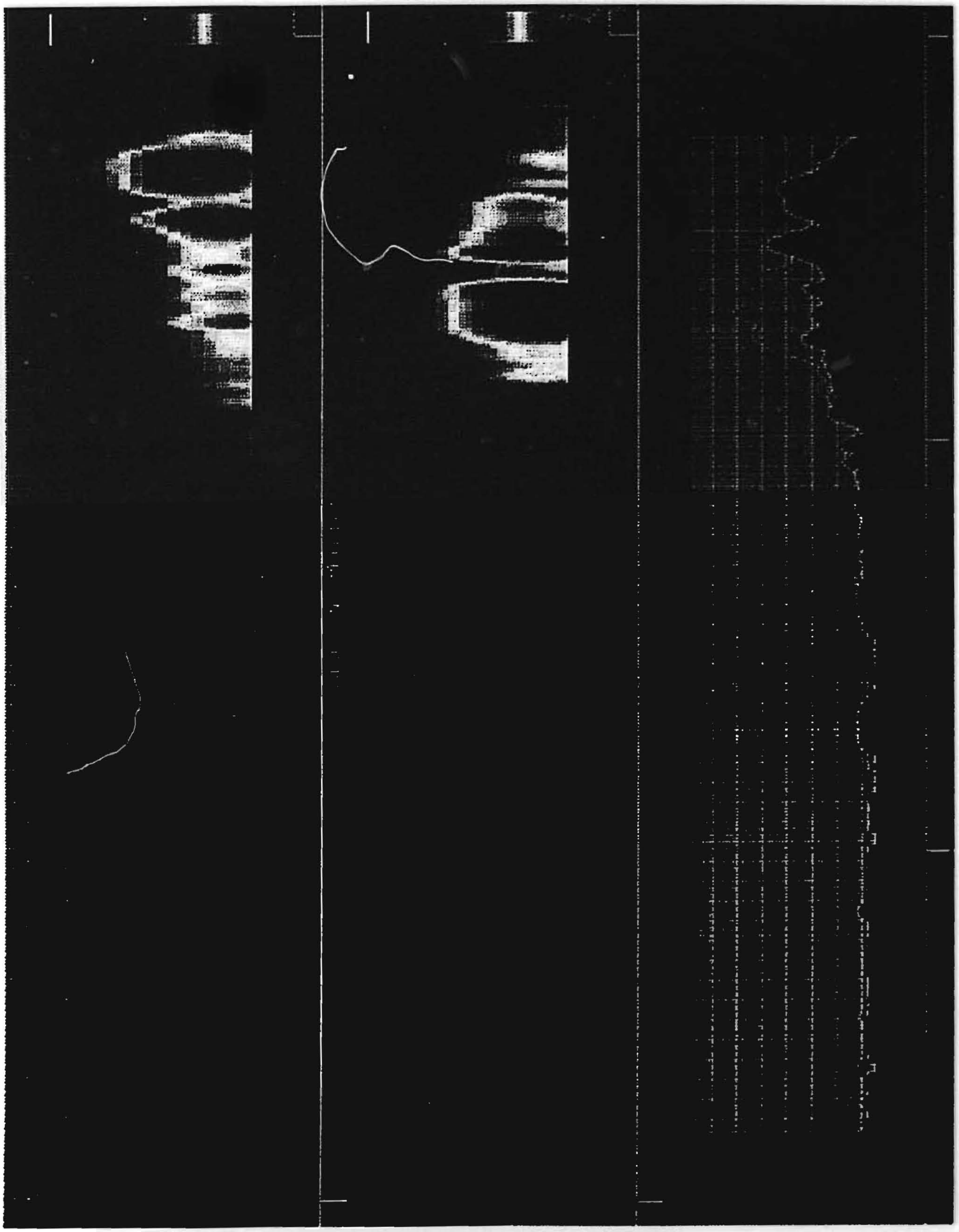
[redacted]

[redacted]

[redacted]

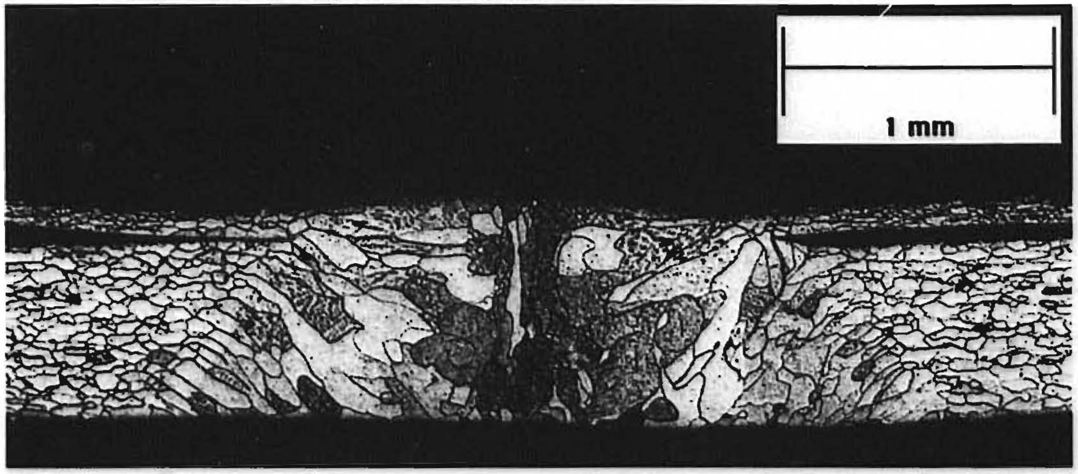
[redacted]





Metallography/Ceramography

Metallography/ceramography examinations are performed on test components recovered from impact tests of $^{238}PuO_2$-fueled and simulant-fueled clads. The microstructure of the GPHS clad material, girth welds, and samples of fuel pellets are also examined.



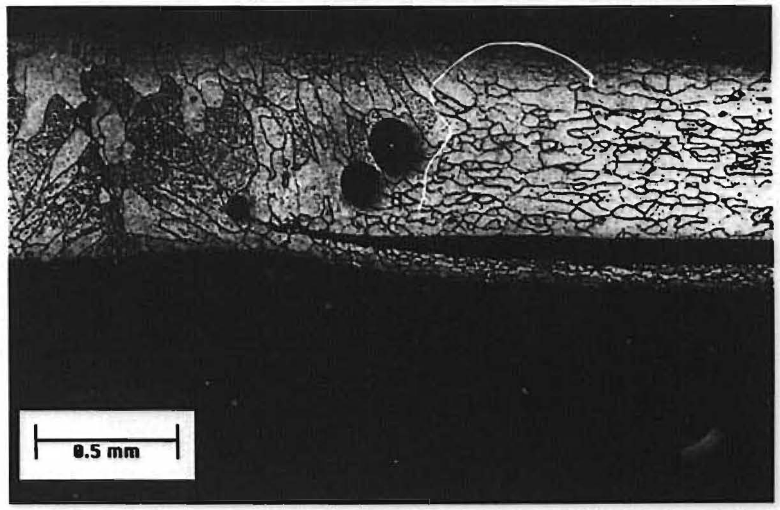
Metallography-conforming graph

SME: MaryAnn Reimus, Paul Moniz

Metallography/Ceramography

A LECO 300 metallograph with magnification range of 8 to 500 X is interfaced to the glovebox line through a unique hood extension that covers but does not enclose the metallographic stage.

The specimens are prepared using wafering saw, automated grinding, rough and fine polishing equipment.



Metallography-nonconforming graph

SME: MaryAnn Reimus, Paul Moniz

Acknowledgement

We thank Liz Foltyn, Tim George, and Kevin Ramsey for leading the past, current, and future ^{238}Pu programs at LANL.