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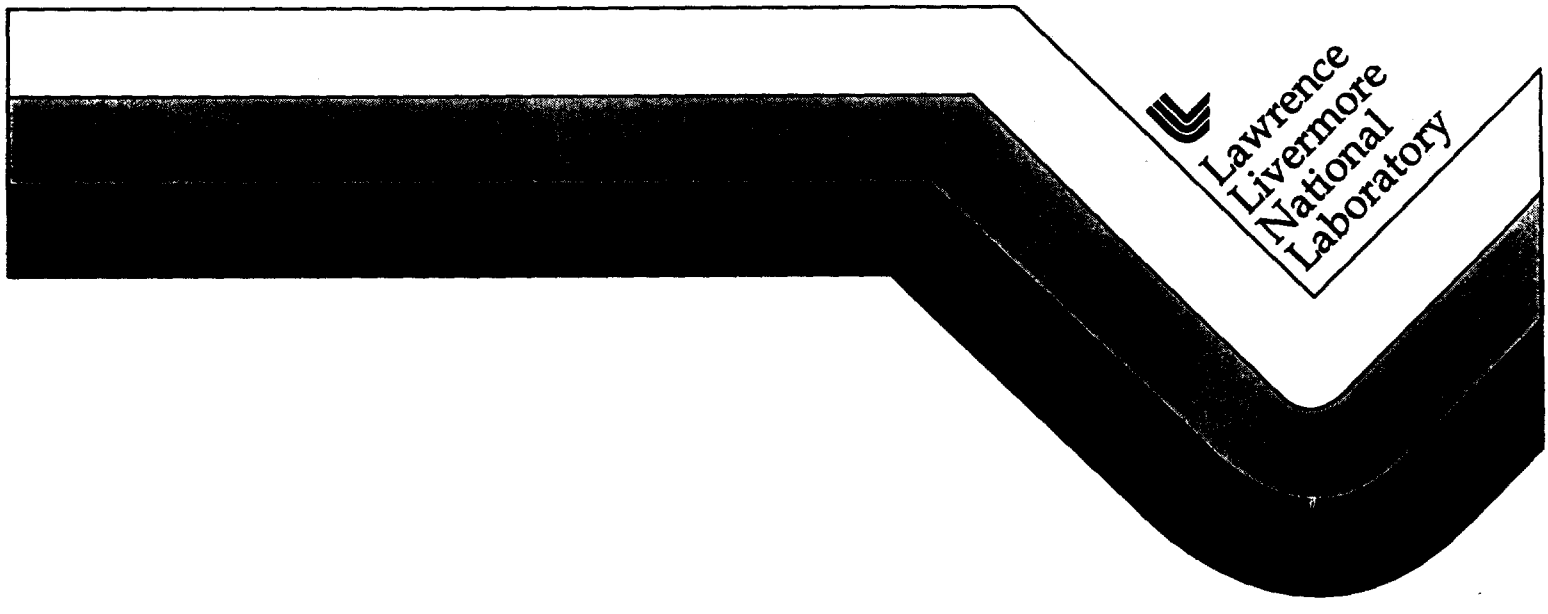
LLNL Workshop on TEM of Pu

Wayne E. King

September 10, 1996



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LLNL Workshop on TEM of Pu

September 10, 1996

I. Executive summary

On September 10, 1996, the Chemistry and Materials Science Directorate of Lawrence Livermore National Laboratory hosted a Workshop aimed at answering the question:

Is it possible to carry out transmission electron microscopy (TEM) on plutonium metal in an electron microscope located outside of the LLNL plutonium facility?

The workshop, attended by scientific staff from C&MS, B-Div., LANL, LLNL Pu facility, LLNL health physics, and Washington State University, focused on the evaluation of a proposed plan for Pu microscopy both from a technical and environment, health, and safety point of view. After review and modification of the plan, the workshop participants unanimously concluded that: (1) the technical plan to carry out TEM on Pu in a TEM facility located outside of the LLNL Pu facility is sound, (2) this technical plan, including a proposal for a new TEM, provides significant improvements and unique capabilities compared with the effort at LANL and is therefore complementary, (3) there is no significant environment, health, and safety obstacle to this plan.

II. Workshop participants

Name	Affiliation	References
Tom Zocco	LANL/MST-5	1-4
Joseph Magana	LLNL/CMAS	
Geoffrey Campbell	LLNL/C&MS	5-13
Adam Schwartz	LLNL/C&MS	14-19
Larry Thomas	WSU	20-27
Mark Wall	LLNL/C&MS	28-34
Dave Lassila	LLNL/B-Div.	35-40
Brooke Buddemeier	LLNL/HCT3	41
Wayne King	LLNL/C&MS	8-13,42-45

Tom Zocco is Materials Characterization Team Leader and Pit Fire Resistance Project Leader at LANL. He has arguably the most experience in the DOE complex at carrying out TEM on Pu. Larry Thomas is a professor at Washington State University. Before joining WSU, Larry was on the staff at PNNL/HEDL where he was the DOE expert on TEM of highly radioactive materials. Brooke Buddemeier is a health physicist for B235 at LLNL. Joe Magana is a chemist in the LLNL Pu facility.

III. Introduction

It is well known that microstructural features such as grain size, grain boundaries, dislocations, impurities, alloying element distributions, and point defect clusters have an important influence the macroscopic mechanical behavior of plutonium, even under extreme deformation conditions. Despite the importance of microstructure, certain aspects of microstructural characterization of Pu have been largely overlooked in the past. Recent events (including the release of the Metals and Alloys

Committee report[46] and developing interest in aging effects) have however heightened interest in microstructure and the related materials science.

TEM is generally accepted in the materials science community as an essential tool for microstructural characterization at length scales extending from atomic dimensions to several microns. TEM is particularly well suited to analyze defects in crystal structures such as grain boundaries, dislocations, precipitates, inclusions, voids, and gas bubbles which often control macroscopic properties. Examples of such defects in Pu are shown in Figures 1-3. Using well established imaging and diffraction techniques, it will be possible to determine dislocation densities, dislocation character, deformation mechanisms, and point defect cluster/bubble geometry, symmetry, and density. In-situ heating experiments can give information about kinetics. Analytical capabilities in the TEM can provide two-dimensional compositional information at resolutions approaching that of the instrument (0.15 nm).

Despite the important role played by TEM in most materials science laboratories, relatively little progress has been made in microscopy of Pu. Roughly a decade ago, LANL set-up a 200 keV TEM for Pu studies in the Pu facility. Over the next five years, significant effort was expended to develop methods for preparation of samples for observation in the TEM. These efforts resulted in the images shown in Figures 1-3 as well as some others. The materials that were investigated were primarily rolled sheet or sputter deposited films (supplied by LLNL) and little or no work was done on materials of direct programmatic interest.

LANL expects to reactivate its effort in this area in FY97 using the electron optical and sample preparation technologies developed previously. This technical plan represents a significant step forward in the microstructural imaging of Pu and the safe preparation of high quality TEM samples and is complementary to the LANL effort.

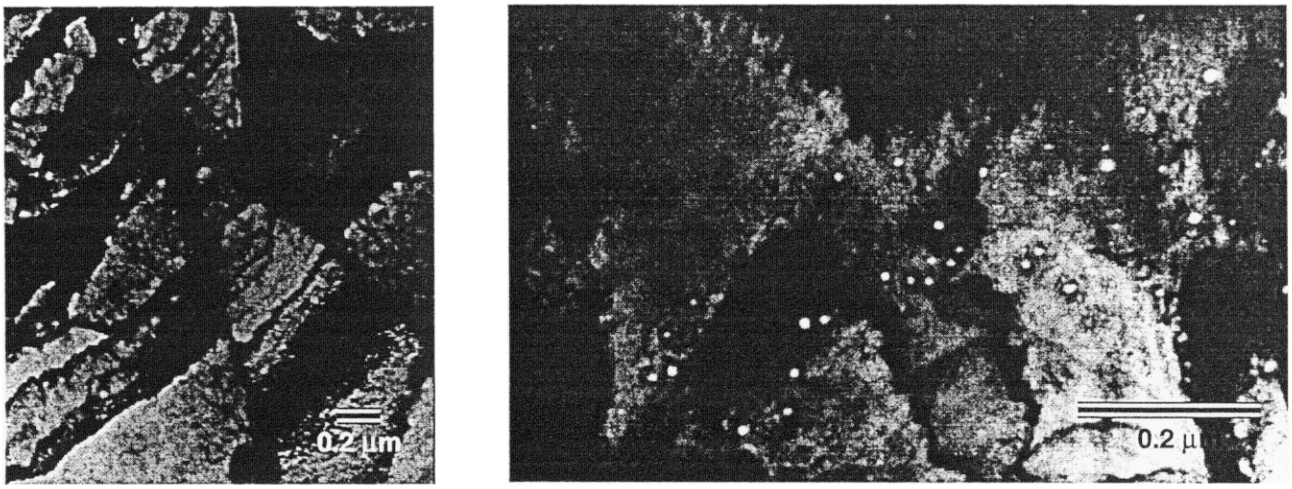

Figure 1. Examples of TEM of Pu acquired at LANL. Material was heat treated at 400°C for 1 hour after 17 years of aging. Observations (these micrographs) were taken approximately 10 years later.

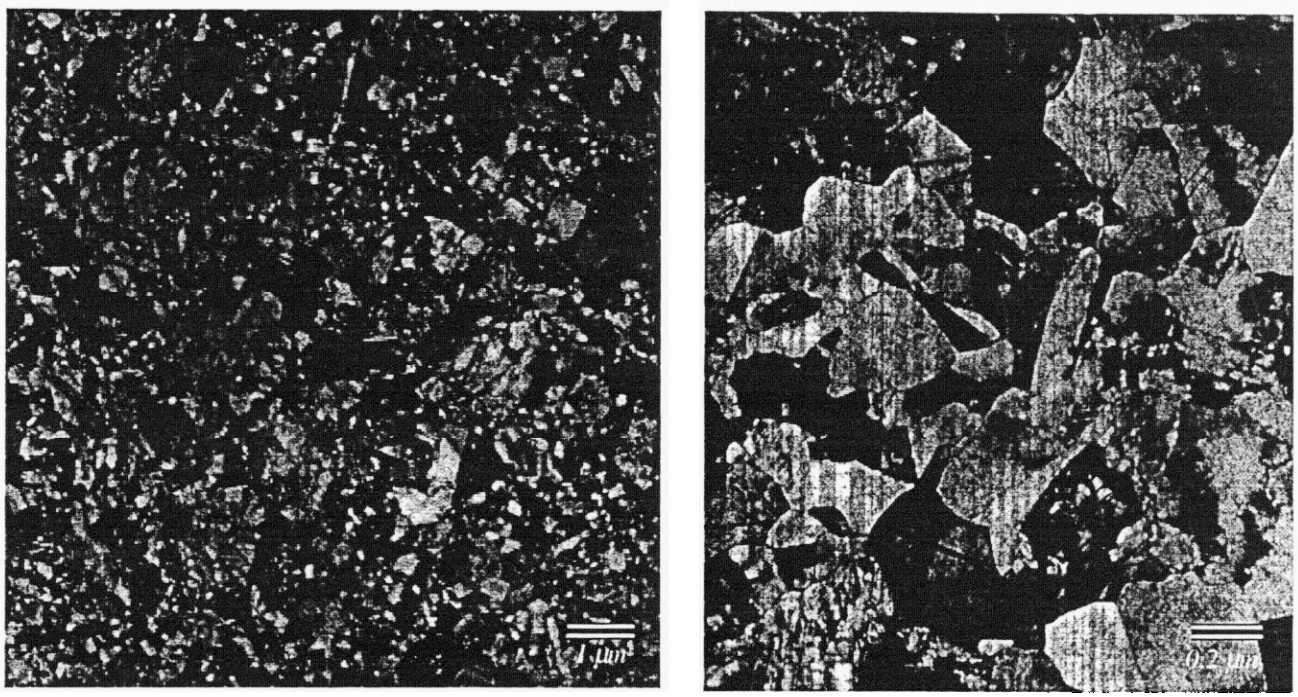

Figure 2. TEM observation of sputtered plutonium. Sputtered samples were prepared by A. Echeverria and Harry Rizzo (LLNL).

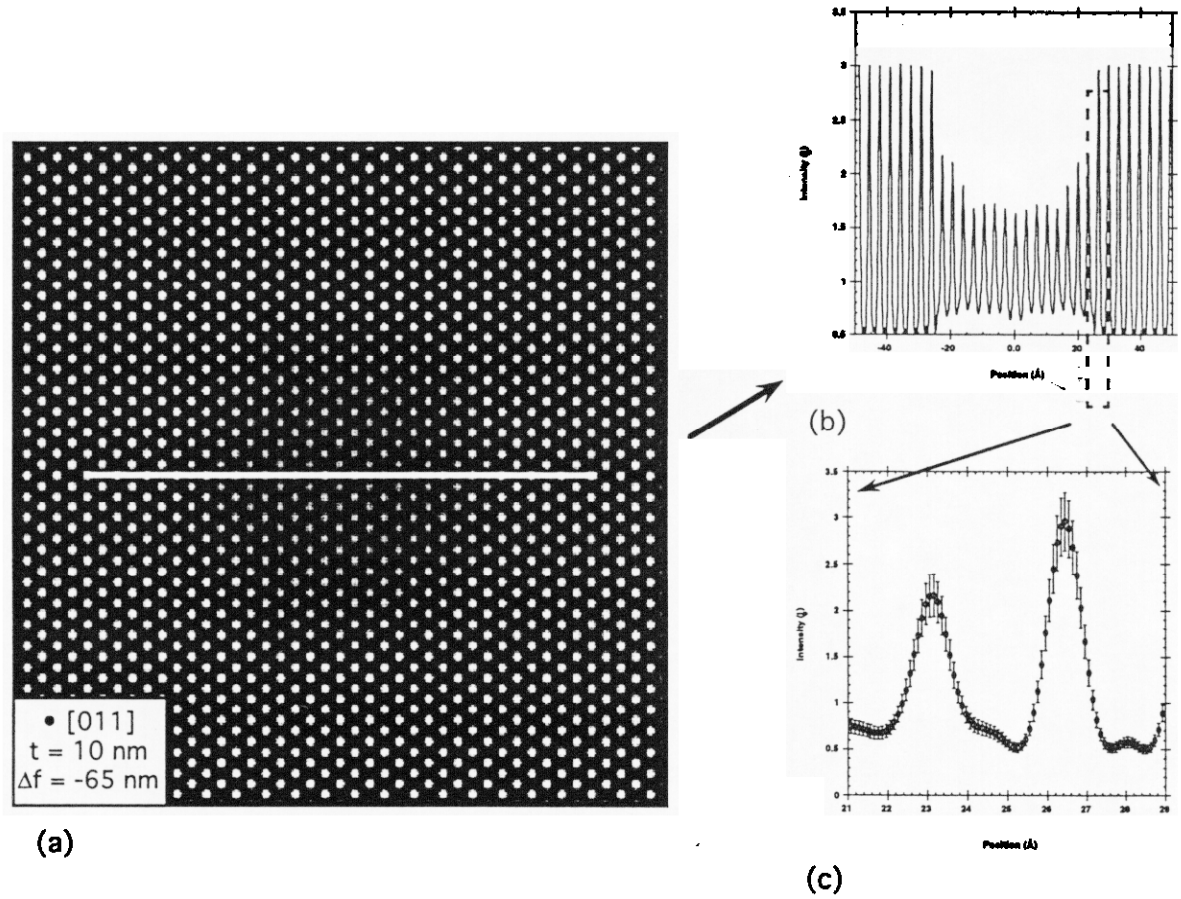

Figure 3. (a) Simulated high resolution electon microscope image of a 50 Å void in the center of a 100 Å thick foil of Pu. Conditions used were for a Philips CM300FEG-Ultratwin. To estimate the smallest void discernable by HREM, a line scan was taken across the center of the void (b). The contrast at the edge of the void was extracted and plotted along with error bars corresponding to 95% confidence level (c). The void at this point is 2.5 nm thick. It is concluded that it is possible to discern a 2.5 nm void in Pu at 95% confidence level for thes imaging conditions.

IV. Evaluation of the technical plan

The details of the technical plan are available on request. We have elected to include only the workshop participant's evaluation of the plan in this briefing. Most details are addressed by the external participant's statements.

A. Harvesting of representative samples from the bulk

Sufficient capabilities already exist in the Pu facility to harvest representative samples from the bulk plutonium.

B. Fabrication of "as low as reasonably achievable" (ALARA) samples

A unique aspect of this plan, which is based on the work of L. E. Thomas on highly radioactive materials,[27] is the intent to minimize the mass of Pu used (<5 mg) for the TEM sample thus facilitating TEM outside of the Pu facility and to adopt a policy of zero acceptable contamination for the TEM (required for the proper function of analytical capabilities and beneficial to avoid cross contamination of samples). This part of the plan involves work in the Pu facility and preparation of samples in glove boxes under inert atmosphere (LANL samples are produced in air). No significant obstacles to success have been identified. Electropolishing techniques have been developed and success rates ~50% (acceptable for TEM sample preparation) have been obtained at LANL. Typical extents of electron optically thin areas for Pu are ~50 μm.

C. Transfer of sample to TEM

Because of the small mass of Pu involved and the proposed method of sample transfer between the Pu facility and the TEM, the workshop participants find that the TEM can be located in any Radioactive Management Area (RMA) at the Lab permitted by the FSP. Indeed, more severe constraints on the TEM location are imposed by the manufacturer's specifications on the site which include vibration, electric fields, temperature control, sound levels, and electrical services. At least one lab in B235 (after some modifications) is expected to meet the manufacturer's requirements.

A significant technical and ES&H issue is the formation of an oxide layer on the surface of the sample during transfer to the TEM. This oxide is known to be only weakly adherent and spalls readily resulting in loose contamination. This problem will be minimized using a commercially available vacuum transfer sample holder such as that shown in Figure 4. If oxide layers form even under these conditions, a thin passivating coating will be sputtered onto the sample before transfer.

The Pu "Sample" will be delivered by Materials Management Transportation to the Building Health and Safety technician who will "receive" it. The technician will then give it to the TEM operator for their use. The TEM operator will requre Rad worker I (HS6010 1-4 hr course), X-ray Safety (HS6070 - 2-3 hr. course) and Contamination Control (HS6300, 1hr). Retraining is required every 2 years.

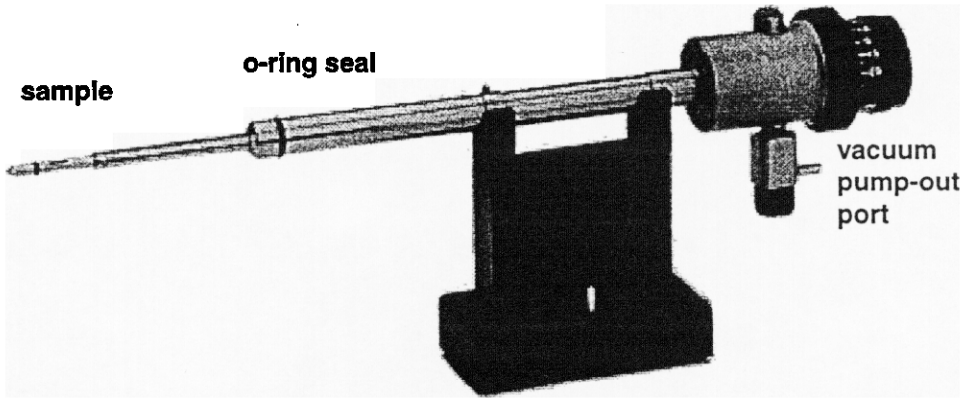

Figure 4. Example of vacuum transfer sample holder appropriate for transfer of Pu samples to the TEM without exposure of sample to air or exposure of staff to Pu.

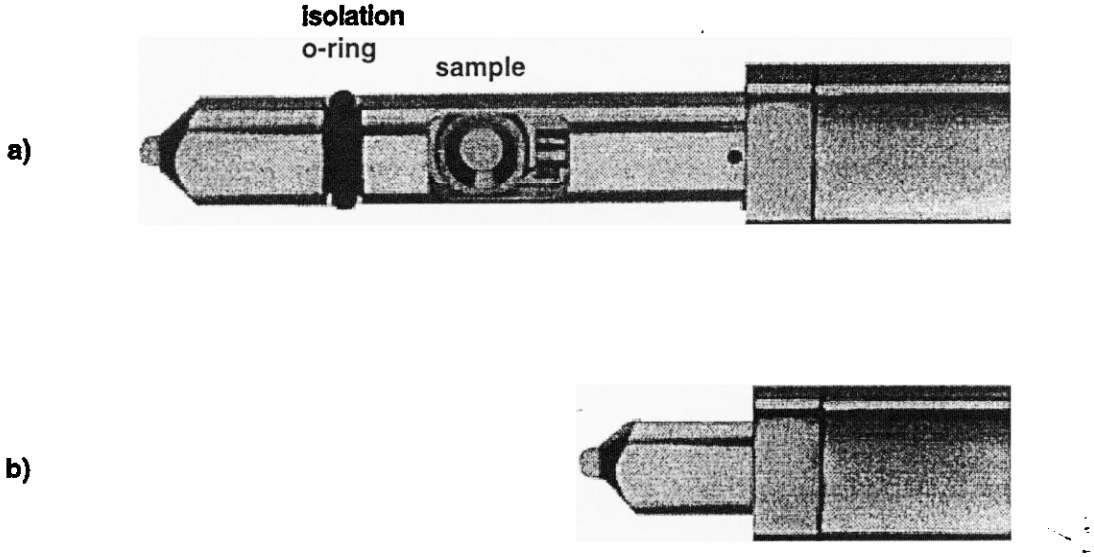

Figure 4. Tip of vacuum transfer sample holder in (a) extended position for sample loading and TEM observation and (b) in retracted position for transfer.

The building Health and Safety techician and Materials Management will then be used again to send the source back to the Pu facility for disposal or archiving. The Health and Safety technician will also be the one to perform the release survey on the TEM (if possible) or on subsequent samples (by swiping the transfer mechanism near the sample).

D. Sample disposal

Following transfer of the sample back to the Pu facility, it can be handled or disposed of using standard practice.

E. Location of the electron microscope

There are strong arguments for performing TEM outside of the Pu facility:

1) The users of the microscope will need to be trained, at most, at the radiation worker level and not at the Pu handler level.

2) The instrument will be available for use even in the event of a temporary shut-down of the Pu facility.

3) Location facilitates use of the machine by outside users and consultants.

The workshop participants find that TEM on Pu can be carried out in any LLNL building that is an RMA where the FSP/OSP have been appropriately prepared.

F. Statements by external participants

Statement from T. Zocco:

Wayne, Thank you for giving me the opportunity to attend and contribute to your workshop yesterday. I have greatly enjoyed my past interactions with personnel at LLNL and hope this will continue in the future.

I would like to summarize the discussions and primary conclusions made at the workshop.

On September 10, 1996, I met with Wayne King, Larry Thomas and other LLNL personnel to discuss a proposed method for performing Transmission Electron Microscopy (TEM) on plutonium metal and alloys. This proposed method suggested that microscopy could be performed on plutonium using a TEM located outside the LLNL Pu facility. Issues discussed included 1) sample preparation methods, 2) ES&H issues pertaining to the handling and transportation of plutonium, 3) equipment requirements and 4) analyses to be performed.

The method proposed would require that all preparation of the plutonium samples be conducted within the confines of the LLNL Pu facility. This would require use of existing facilities as well as an upgrade/new facilities for some of the specific TEM preparations methods discussed at the workshop. Once prepared, the samples would be removed from the Pu facility and transferred to the microscope laboratory.

The technical details of the proposed method would utilize a number of commercially available instrumentation. Bulk Pu metal would initially be processed using conventional metallographic techniques (i.e. cutting, grinding, mechanical/electropolishing). Once a thin disk/wafer of material was prepared, it would be mounted into a fixture which would act as a supporting structure for the Pu sample. At this point final thinning of the sample could be performed in a variety of ways such as jet electropolishing or ion thinning. With the sample thinned to electron transparency, the final step would involve the deposition of

a thin layer of material which would act as an encapsulation shell to protect the Pu metal sample from oxidation. At the same time, this coating would provide protection to the local environment near the specimen, including the microscope chamber and vacuum system.

Several suggested coatings included B_4C, Ni, SiO_2 and others. Further investigation is required to finalize which materials would offer the best encapsulation. The material must provide oxidation resistance while still remaining electron transparent, and the material must not hinder the collection of high resolution imaging or interfere with collection of electron diffraction data. In some cases the coating will also need to withstand some mechanical damage, as might be caused by frictional forces upon loading and unloading.

New LLNL equipment would allow the following advantages (as compared to existing LANL facilities which currently handle Pu samples and perform TEM analysis): 1) state-of-the-art high resolution imaging of the Pu materials, 2) preparation of samples in an inert environment, potentially eliminating the presence of oxide surface films from the current preparation methods used at LANL, 3) use of increased microscope accelerating voltages for observation of thicker specimens and 4) use of energy filtering to collect both microstructural and chemical information from thick samples.

If an encapsulation material can be clearly identified and deposited using a method which will not alter the microstructure of the Pu materials to be examined, then I see no reason why this method would not produce observable Pu microstructures, while maintaining a non-radioactive microscope environment.

Finally, it is anticipated that a delay period will exist during establishment of the LLNL facility. During this period, LANL facilities will be available to assist LLNL Pu TEM needs. Again, thank you Wayne for allowing me to participate in the workshop and I look forward to further collaboration with you and your team. If any of my comments above appear incorrect, please let me know.

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Statement from L. E. Thomas

Wayne, Thank you for the opportunity to participate in the workshop. My summary of the discussions and primary conclusions follow:

Summary of workshop on TEM of Pu, held at LLNL, Sept 10, 1996

Participants: Wayne King Mark Wall Tom Zocco (LANL) Joe Magana Geoff Campbell Adam Schwartz David Lassila Brooke Buddemeier Larry Thomas (WSU)

On Sept. 10, 1996, I met with the above participants to discuss a technical plan for carrying out TEM examination of Pu metal/alloy samples outside a Pu containment facility. Key elements of the plan that came from this discussion were: (a) a method for preparing contamination-free TEM samples suitable for high-resolution microstructural examination and microchemical analysis in a state-of-the-art TEM; (b) Minimization of active material in the samples in order to reduce laboratory safety requirements and avoid the need for material accountability; (c) a method for transferring samples from the preparation facility to the TEM without exposing the samples to air or risking contamination of the TEM laboratory or personnel; and (d) (optionally) a means to ensure the contamination-free status of the TEM to allow its unrestricted use for examining nonradioactive samples. The plan as conceived incorporates technically and radiologically sound practices that, in my experience, should allow high-resolution TEM analyses of Pu samples in an unmodified TEM with normal radiological controls.

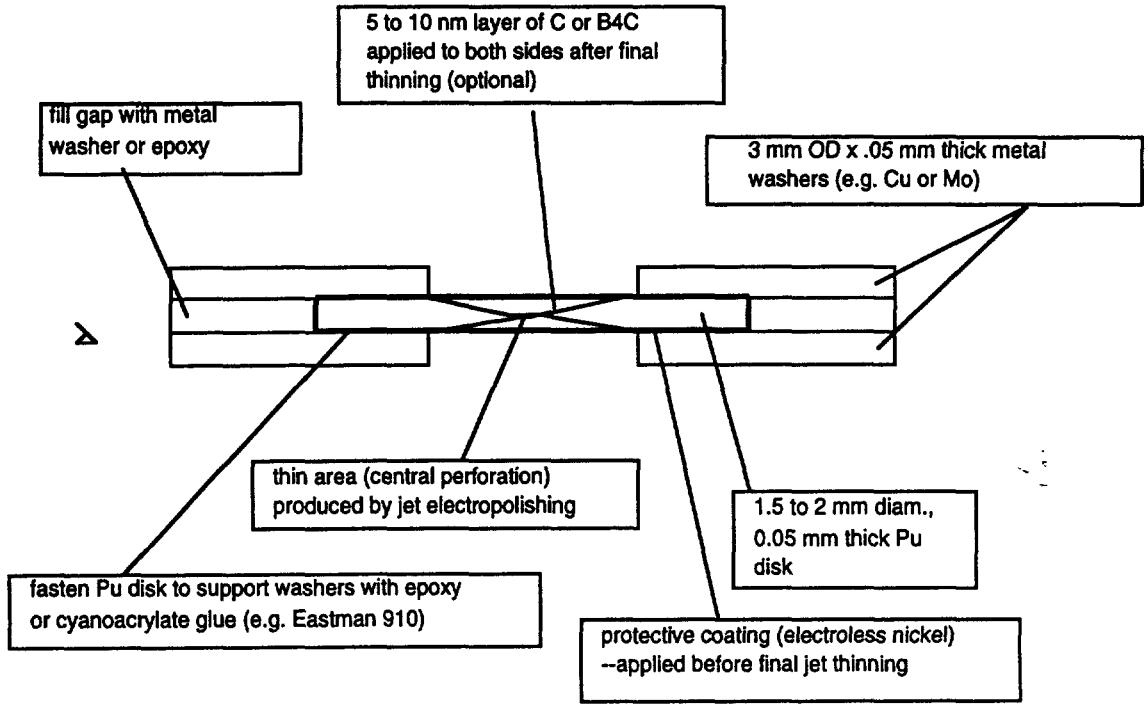
In this plan, all sample preparation operations would be conducted in an inert atmosphere glovebox within a Pu containment facility. A commercially available vacuum or inert-gas transfer sample holder would be used to move prepared samples from the preparation facility directly into the TEM. Directly transferring the samples into the TEM in the sealed and inerted transfer holder would virtually eliminate the risk of contaminating either the laboratory or personnel.. After examination, samples would be returned to the preparation facility in the same transfer holder, and unloaded there for disposal.

Although perhaps not essential from a radiological control viewpoint, a goal of this plan is to conduct to sample preparation and transfers in a manner that would also involve neglegible risk of contaminating the TEM interior. This goal requires producing dust free samples in which exposed Pu surfaces cannot contact the sample holder or microscope interior, and surface oxide on exposed Pu surfaces is either avoided or contained by an appropriate protective coating. Intermittent monitoring of the unloaded transfer holder, microscope airlock, and microscope sample chamber could be used to ensure that the contamination-free status of the TEM is maintained.

Sample preparation: A wide variety of methods can be used to prepare TEM samples, depending on material properties such as electrical conductivity, hardness, and friability, and on the physical state (shape, density, particle size, etc). In the present method, bulk Pu metal would be reduced to a thin sheet by standard metallographic methods such as cutting, grinding, and chemical or electropolishing. A thin disk of the sheet material would be mounted with most of the material protected by surface coating or masking by TEM support washers, and the central region would be thinned to electron transparency by electropolishing or ion micromilling. Based on the experience of T. Zocco (LANL) in preparing Pu metal for TEM examination, jet electropolishing with commonly used electrolytic solutions such as dilute perchloric acid in methanol, ethanol, or acetic acid would be the method of choice for final thinning. Jet electropolishing works for metallic materials and is generally preferred over other methods for its simplicity, rapid sample production and low equipment cost. The small amount of mixed waste (used electrolyte) generated by this method, typically 500 to 1000 ml per year, can be disposed by normal laboratory waste management practices within the Pu facility. Ion milling could also be applied if the need arose for finishing the electropolished samples to remove surface irregularities or anodic films or to improve the amount of electron-transparent area. However, radiation damage produced by ion milling might interfere with microstructural observations in Pu samples.

Basic requirements for a noncontaminating TEM sample of Pu metal are that the sample should be nonfriable, free of dust or other loose pieces, and not contain a dispersible oxide film on the Pu metal surfaces. In addition, the sample should contain the minimum amount of Pu material necessary to prepare suitable electron-transparent areas and maintain the sample integrity. Unthinned sample areas needed for mechanical support should have a protective coating or be embedded in a protective layer of material such as epoxy resin. Sample design should allow the finished samples to be loaded in the TEM holder so that no area of exposed Pu metal can directly contact the holder or internal parts of the TEM. If the formation of surface oxide on exposed areas of the thin sample proves unavoidable, a thin protective coating of noninterfering material (e.g., 10 nm of sputter-deposited carbon or boron carbide) should be applied to the sample surfaces after final thinning. However, with sample preparation and transfer under inert gas or vacuum, the use of a final protective coating might be avoided to improve the quality of TEM observations.

A sketch of the proposed sample as I envision (is shown below) .



Due to their inherently delicate nature, TEM samples and sample holders (especially the precision tilting mechanisms in the sample holder cup) do not allow smear-testing for contamination. The presence of contamination on the unloaded sample holder or microscope may be assessed by noncontact proximity monitoring or by smear testing of external areas up to the vacuum seals on the holder or sample exchange airlock.

Transfer holder:

Vacuum transfer sample holders that interface with the side-entry goniometers and sample airlock ecxchange mechanism of most electron microscopes (Philips, JEOL, LEO, Hitachi) are commercially available from Gatan Inc, Pleasanton, CA (tel . 510 463-0200) and Oxford Instruments (tel. 508-369-9933). These include x-y tilting holders and variations for cryo (LN temperature) as well as vacuum and inert gas transfers. The Gatan Model 648 double-tilting holder from Gatan (about $25K) appeared especially attractive for present purposes because it retracts the sample holder tip inside the sealed holder shaft (see photo in page 17 of Gatan catalog) while the holder is transferred through the microscope airlock. This protects the airlock from contamination as well as the microscope exterior.

Besides providing noncontaminating samples, a considerable advantage of this sample preparation method over the in-air preparation and transfer previously used by T. Zocco and others at LANL is that avoiding surface oxidation of the Pu samples can eliminate interfering strains and image artifacts caused by the oxide films. Thus, the samples in a capable TEM should allow high- resolution microstructural observations and microchemical analyses.

Issues:

Issues to be addressed to make this technical plan feasible are:

1. Locate and prepare suitable gloveboxes for sample preparation under inert cover gas. 2. Provide jet electropolisher, sample coater, and appropriate airlock to allow airlocked sample exchanges with vacuum sample holder. 3. Arrange disposal of ~1000 ml of mixed waste from the electropolisher. 4. Train technician to perform sample preparation in Pu containment facility. 5. Develop and test complete sample preparation procedure with nonradioactive samples on bench outside Pu containment facility. 6. (Optionally) Obtain equipment for sample preparation by ion milling (dimple grinder and ion mill), and install in glovebox.

Statement from B. Buddemeier

Wayne,

It was a pleasure to be involved in the discussion of the TEM analysis of Pu. This a modified version of my original hazard assessment which was undergoing peer review when you released the First draft of Workshop report and all of the subsequent discussion ensued. I hope that I have captured many of the issues that were brought up since you released your draft report. It is the intention of this analysis to identify any major pitfalls for the possibility of locating the TEM outside of the LLNL Pu facility. A more detailed analysis will be performed on subsequent Operational Safety Procedures (OSP) as they are written. It is presumed that the sample preparation will be performed in the Pu facility and is not covered under this analysis.

One of the discussed sample sizes, 2 mm diameter and 0.05 mm thick disk, would equate to roughly 3 mg or 0.25 mCi. Normally this quantity of material would be of significant concern but it's solid metal configuration and encapsulation in

the TEM sample preparation process significantly minimizes the likelihood of the Pu being dispersed (see distributed sketch). The Sample Transfer holder adds another level of containment by keeping the Pu under vacuum or inert atmosphere to avoid oxidation of the small portion of the Pu that is exposed.

Outside of the sample preparation area, the area that has the highest potential for contamination is the inside of the transfer holder and the TEM chamber. If there is surface contamination of the sample (or shaft of the sample holder) then this contamination might transfer to the TEM chamber during sample position adjustments. Therefore, the TEM will be considered to have "Potential Internal Contamination" once a Pu sample has been analyzed. Since releasing the TEM chamber after each plutonium analysis might not be feasible, I recommend simply releasing subsequent non radioactive samples by surveying and swiping the area near the sample in the transfer holder.

The likelihood of spreading contamination in the TEM or the sample holder can be minimized by coating the sample after it has been prepared for TEM. In any case, the levels of contamination would be expected to be relatively low even if the sample did oxidize; however program management may wish to take additional steps to avoid contaminating the TEM which could result in cross contaminating other samples. Administrative controls to avoid the possibility of sample sputtering due to operator error should be addressed in the OSP.

The vacuum system that pumps down the transfer holder and TEM would also be an area of potential internal contamination. Placing a filter in the vacuum line between the TEM and the roughing pump could be used to evaluate the possibility of chamber contamination and certify cleanliness of the vacuum pump.

Until the durability of the sample can be established, the possibility of a catastrophic sample failure within the TEM will need to be addressed. This health and safety concern can be mitigated by keeping the TEM chamber air pressure slightly negative during sample removal or by a simple "bag out" procedure for the transfer holder until it can be surveyed.

There were several options for sample preparation and coating methods discussed. Regardless of the decided method, I recommend testing the proposed sample configurations, including coatings, with actual plutonium samples for durability. This testing can be done at the Pu facility. If the samples are not coated then we would need to know the effects of exposing an "uncoated" prepared sample suddenly to air (in a glovebox). It would be very helpful to know how fast the sample will oxidize, how easily the oxidation comes off (when shaken or rubbed), and whether the sample will spontaneously ignite. It is important to know the consequences if the vacuum fails on the transfer holder while it is in transit.

As for site location, most of the buildings on site in which radioactive materials are used are radiological and not nuclear facilities and therefore have an upper limit of 8.4 g of Pu-239. An OSP will have to be written and a NEPA review would have to be conducted. The room where the TEM will be located will be a RMA

(Radioactive Material Handling/Storage Area). Release limits for potentially Pu contaminated equipment are 20 dpm/100 cm^2 removable and 500 dpm/100 cm^2 fixed contamination. Materials Management will need to perform the transfer and should be contacted for their comments. When not in use the sample would need to be located in a fireproof safe.

Brooke Buddemeier LLNL Team 3 health physicist with assistance from health physicists Gary Mansfield, Jim Mecozzi, and Kathy Shingleton.

V. Issues

A. Training of Pu facility technicians

Certified Pu handlers will need to be trained to prepare TEM samples. This can be done initially outside of the Pu facility on materials which thin similarly to Pu.

B. Glove box line.

Fabrication of the samples for TEM will require set up or renovation a glove box line in the LLNL Pu facility to facilitate final thinning of the sample by mechanical dimpling, jet polishing, or ion beam thinning.

C. Potential for contamination

Despite our intent to obtain zero contamination of the TEM, it is possible that a contamination could occur. Therefore, we must treat several parts of the microscope as disposable including: specimen holder, airlock, objective lens polepiece, and objective aperture blade

D. Delay for procurement of TEM and Pu facility installations

The lead time for procurement of the 300 keV, in-column energy filtered TEM is approximately one year. Similarly, it is expected that set-up of the glove-box line in the Pu facility and training of the Pu facility technicians will take a similar amount of time. During that time, preliminary work will be carried out at LANL (see Zocco statement above).

E. Suppression of sputtering

The possibility of electron beam-induced losses of material in a TEM could conceivably be significant. Local thinning of metal samples under the spread illumination used for imaging in a TEM is normally undetectable, even for light elements (Be, B, Al) that can receive relatively large energy transfers from 300 KeV electrons. The maximum energy transfer is significantly less for heavy elements like Pu than it is for light elements, and displacement energies tend to be higher, so sputtering should be insignificant.

On the other hand, hole drilling in metals under a focused nanoprobe in the field emission gun TEM is fairly common. In experience with the 2010F FEG microscope at PNNL, for example, it is not

unusual to get nm-sized holes in stainless steel and Ni-based alloys during nanoprobe analyses. This drilling effect appeared in samples with anodic oxide films or other surface layers, but did not appear in clean samples. It was attributed to beam-induced evaporation, sputtering or chemisorption of oxide layers, and seemed to be eliminated by cleaning samples in an ion mill. Sample cleaning with the new plasma cleaners was also being evaluated to see if it would eliminate the hole-drilling.

To put this in perspective, the "worst case" of drilling a 5 nm diameter hole in a 50 nm thick foil would release less than 10^{-18} g of material, not a very large amount.

VI. Findings

- *The technical plan to carry out TEM on Pu in a TEM facility located outside of the LLNL Pu facility is sound.*

- *This technical plan, including a plan for a new 300 keV, energy-filtered TEM, provides significant improvements and unique capabilities compared with the effort at LANL and is therefore complementary not competitive.*

- *There is no significant health and safety obstacle to this plan.*

VII. Acknowledgments

This work is performed under the auspices of U.S. Department of Energy and Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

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