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Author Contribution to the Pu Handbook II: Chapter 37 LLNL Integrated Sample Preparation Glovebox (TEM) Section

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Disclaimer

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Instructions to author from Jeremy Mitchell of LANL:

I'd *like to get rough drafts from you by late July; please let me know if you need more time.*

1: Your section: TEM

2; Your section: LLNL integrate sample prep glovebox (talk about other sample preparations for other experiments, DSC, resistivity, having an ante-chamber, laser micro-machining for DAC, X-ray diffraction, beam-line experiments, thermo-mechanical processing for grain growth, lowest common denominator sample preparation example, X-ray encapsulation transmission and

I understand your time constraints and will help in any way possible. I

can address the historical overview for you if you like; please focus on how you perform the work at LLNL.

Chapter 37 Desired Section Contents

Introduction

- Historical overview on application of technique/method to Pu- Key publications that reflect technique development

Methods

- Describe the various methods used to perform measurement as applied to Pu (e..g., for thermal expansion I have reviewed LVDT pushrod, laser pushrod, capacitance, fiber Bragg grating, etc.)
- Identify key publications.
- Do not make this a detailed overview of the technical specifications of the instrument, just identify key aspects for Pu measurements

Sample Considerations

- Describe sample preparation methods, emphasizing any special concerns for Pu samples (e.g., are there any sample prep artifacts?)
- Describe sample size, shapes, etc.

Operational Conditions

- What are the preferred conditions for operation? (e.g., for thermal expansion I have included enclosure type, pushrod load, atmosphere, heating/cooling rates, sample holder materials, etc.)
- What conditions are optimal or undesirable for Pu measurements?

Data Analysis

- Are there special techniques for evaluating data beyond manufacturer's software?
- Are there Pu-specific experimental artifacts that should be discussed? (e.g., for thermal expansion I discuss high-temperature sample deformation, evidence for sample oxidation, etc.)

Examples/Experiment Library

- Are there some representative experiment results worth documenting?

(e.g., I will include phase transformation experiments, alloy homogenization, etc.)
-Are there particular signatures worth including for Pu (e.g., spectroscopic signatures for Pu peaks)

Figures and Tables
- Do not insert figures into text; instead, put in a text line that states “Insert Figure/Table X here”.
- Include a section at the end for Figure/Table Captions.
- Place each Figure/Table on it’s own page at the end of you section.
- The editors will get releases for already-published figures if needed.

References
- I have a Pu Handbook EndNote library that I can send or you can just include reference details that I will place into the collective library.

LLNL Integrated Sample Preparation Glovebox:

Introduction:

The development of our Integrated Actinide Sample Preparation Laboratory (IASPL) commenced in 1998 driven by the need to perform transmission electron microscopy studies on naturally aged plutonium and it's alloys looking for the microstructural effects of the radiological decay process (1). Remodeling and construction of a laboratory within the Chemistry and Materials Science Directorate facilities at LLNL was required to turn a standard radiological laboratory into a Radiological Materials Area (RMA) and Radiological Buffer Area (RBA) containing type I, II and III workplaces. Two inert atmosphere dry-train glove boxes with antechambers and entry/exit fumehoods (Figure 1), having a baseline atmosphere of 1 ppm oxygen and 1 ppm water vapor, a utility fumehood and a portable, and a third double-walled enclosure have been installed and commissioned. These capabilities, along with highly trained technical staff, facilitate the safe operation of sample preparation processes and instrumentation, and sample handling while minimizing oxidation or corrosion of the plutonium. In addition, we are currently developing the capability to safely transfer small metallographically prepared samples to a mini-SEM for microstructural imaging and chemical analysis.

The gloveboxes continue to be the most crucial element of the laboratory allowing nearly oxide-free sample preparation for a wide variety of LLNL-based characterization experiments, which includes transmission electron microscopy, electron energy loss spectroscopy, optical microscopy, electrical resistivity, ion implantation, X-ray diffraction and absorption, magnetometry, metrological surface measurements, high-pressure diamond anvil cell equation-of-state, phonon dispersion measurements, X-ray absorption and emission spectroscopy, and differential scanning calorimetry. The sample preparation and materials processing capabilities in the IASPL have also facilitated experimentation at world-class facilities such as the Advanced Photon Source at Argonne National Laboratory, the European Synchrotron Radiation Facility in Grenoble, France, the Stanford Synchrotron Radiation Facility, the National Synchrotron Light Source at Brookhaven National Laboratory, the Advanced Light Source at Lawrence Berkeley National Laboratory, and the Triumph Accelerator in Canada.

Preparation Methodology

Nearly all of the sample preparation procedures that we utilize have their foundation based upon the well-established and fundamental metallographic preparation procedures of dicing, lapping, polishing, etching, ion milling and electrochemical polishing. The IASPL is located within a radiological facility thus the quantities of plutonium are limited to several grams; and can easily verify by a factor of up to 100 depending on the specific activity (Ci/gr) of a particular isotope. As a result of this mass limitation it has forced us to re-think how we minimize sample material, minimize material loss during preparation

and processing, and maximize productivity. As a result, much of the experimental methodology for sample preparation and design is based upon a lowest common denominator shaped sample. A standard 3mm diameter transmission electron microscopy (TEM) sample shape (weighing in the 10-20mg range for plutonium) is the basic shape for virtually all of the small-scale science experiments. For example: prior to preparing a 3mm diameter cylindrical by 3mm long core weighing ≈ 250mg for TEM it is possible to 1st prepare one end of the sample for optical microscopy, 2nd iteratively cool to induce a phase transformation in a differential scanning calorimeter (DSC), 3rd prepare again for metallographic observation, 4th perform X-ray diffraction on a thin slice from the core, then finally ending with preparing the same sample for TEM studies by slicing off of one end a 200um thick sample. Figure 2 shows some of the results from metallography, X-ray diffraction, DSC and TEM of this one sample. References 2 and 3 discuss the experimental details and results. Adding to this theme, remnants of the used TEM sample disc can then be further thinned and laser cut into many micro-samples for high-pressure EOS diamond anvil experiments.

An important aspect of our methodology takes into consideration that most of the instruments that we use for experimentation are not located inside a contamination control barrier (e.g. glovebox, glovebag or fumehood) and are used by other experimenters for characterization of non-radiological samples. We have utilized and developed a number of encapsulation schemes to safely encapsulate, handle and transfer plutonium samples to these instruments. Examples of these encapsulation techniques will be apparent in the following section that describes various sample preparation successes that we have had. It should be noted that the ability to load/unload plutonium samples into/out-of these encapsulation devices and to keep the exterior of the encapsulation device clean of contamination is greatly aided by the use of a double-door antechamber on our gloveboxes (figure 3), and specifically for TEM the design and construction of an airlock on the side of our gloveboxes that is effectively an airlock that mimics the airlock on the transmission electron microscope (figure 4). Without an antechamber (or TEM airlock) we would need to employ bag-in/out techniques which when utilized, makes it exceedingly difficult to keep the exterior of the encapsulation and transfer devices free of contamination. The exit side of the ante-chambers and TEM specimen holder airlocks are also surrounded by a fumehood so that personnel do not need to wear a respirator while passing the devices out of the glovebox. The engineered contamination control process that we use here is a graded approach much like that of achieving an ultra-clean cleanroom where there are surrounding layers of increasing cleaner chambers. In the case the glovebox is highly contaminated, the airlocks and antechambers are much less contaminated and the fumehoods are minimally contaminated.

Note: Whenever possible we construct specimen encapsulation devices out of aluminum as it is easier to decontaminate and frequently reuse, as compared to stainless steel and plastics.

Sample Preparation with Experimental Result Examples

Within the IASPL we have installed a number of instruments, processing

capabilities, and developed numerous techniques for preparing precision samples for traditional and advanced characterization techniques. The following are examples of the wide range of samples that we have prepared and the science that has been facilitated along with the sample preparation and handling processes that we employed.

Transmission Electron Microscopy (TEM): 3mm diameter cores from bulk material are extracted by either lathe operations, tre-panning on a milling machine or cut with a dicing saw in a facility that can handle larger quantities of plutonium. The extracted samples are then sent to our facility (typically in sealed vials under oil or inert atmospheres), which has a considerably lower inventory limit for plutonium and other radiological materials. Considerable care is used to ensure that the samples are not heated or plastically deformed during these bulk extraction processes as this can cause changes in the microstructure. Cylindrical cores are sliced using a Buehler Isomet saw, figure 5, in glovebox #1 into discs. Discs are individually lapped using precision gravity-feed lapping devices and supplies shown in figure 6, to ≈ 150µm then electro-polished to electron transparency using a Fischione Twin-Jet polishing device, (5) figure 7A.The sample mounting and lapping process is similar to that which is described in reference 4. Only the electrolytic cell of the electro-polishing unit is within the glovebox. The control power supply (figure 7B) is external to the glovebox and is connected via a multi-pin feed-through connection. Electro-polishing of metallic Pu and Pu alloys thus far has worked repeatedly well using a solution of 10% Nitric acid + 45% ethanol +45% butoxyethanol at -15^{o}C with a range of voltage potentials from 40-120 VDC. An alternative to electro-polishing is to pre-thin the 3mm diameter disc using a Gatan Dimpler (figure 8) (6), followed by final thinning using a Gatan PIPS (figure 9) (6) ion milling system in glovebox #2. With either process finished TEM samples are loaded into a vacuum-transfer TEM specimen holder that is loaded into a specially designed airlock (figure 4), attached to both gloveboxes. This specimen holder allows for safe transfer, with minimal oxidation of the specimen, to the TEM and eventual return to the gloveboxes

In order to use the TEM for radiological and non-radiological operations we have adopted several practices for on-going monitoring and decontamination to ensure that non-radiological users of the TEM can have safe access to the TEM and can perform sample holder/specimen exchanges that do not require radiation monitoring or special contamination control training. The following is a list of contamination control practices and procedures to ensure this.

1. The vacuum transfer holder is kept clean, to non-detectable levels, of removable contamination on all surfaces that are exposed to atmosphere. Fixed contamination on exposed surfaces is kept well below 200 DPM alpha.
2. After each Pu sample loading and return to the glovebox the internal parts of the sample holder is decontaminated so that there is no removable contamination. Note: fixed contamination will build up over time to substantial levels, therefore it is necessary to minimize mechanical disturbance to these surfaces as this may result in the loosening of contamination.
3. After removing the radiological sample holder from the TEM airlock it is surveyed for contamination. Cleaned if contamination is detected.

4. Area surveys after each removal of the TEM radiological holder is performed.
5. The airlock of the TEM is indirectly surveyed for radiological contamination after each removal of the radiological sample holder.
6. Weekly swipes by of ES&H technician of the TEM external surfaces are performed.
7. Weekly survey and swipes of the non-radiological sample holders are performed in order to look for cross-contamination. To date (for ≈ 18 years from start of operations) there has been no cross-contamination detected above 0.05nCi.
8. Annually, or prior to servicing of the internal vacuum region of the objective lens/sample area we perform internal surveys and swipes. To date, it is not uncommon to find up to 10nCi of alpha contamination internally. We will remove as much of the contamination as possible. For non-LLNL service personnel to do work on the internal areas of the vacuum system we decontaminate to background levels prior to allowing this service work to be performed.

Note: These contamination control processes are also an example of the practices that we employ of the dual use (radiological and non-radiological) of other instrumentation as well.

***Optical Metallography**:* Instead of the traditional potting of samples in epoxy for metallographic sample preparation and handling, which makes them difficult to extract for further experimentation, we use a single component cyanoacrylate (Sally Hansen)™ to attach specimens to the mounts that come with the lapping devices (figure 6). Alternatively the use of low temperature melting waxes can be used for sample mounting. We often modify our sample mounts by epoxying a thin glass slide to the top of the mount. The glass surface provides electrical isolation for the often-final step of electrolytic etching or polishing that is used for revealing the microstructure. An alternative for electro-polishing/etching/anodization of a sample that has been removed from the lapping mount is to use the TEM electro-polishing system. Mounted samples can either be lapped by hand on a glass plate using a finer succession of diamond lapping films (4), e.g. 30, 9, 3 and 1μm followed by electro-polishing with the same solution and conditions for TEM sample preparation. For room temperature electro-polishing we often use a solution of 3 parts phosphoric acid 4 parts glycerol and 4 parts ethanol in a range of 10-25 volts ref 10. If needed, we can perform slurry polishing with the same lapping device using a Beuhler Mini-Met lapping instrument, figure 10. The Beuhler Mini-Met polisher has been modified to guide the lapping device for final polishing on a lapping cloth of choice. We often skip the traditional final polishing step of a colloidal silica or Al_2O_3 as this often leads to hydride formation and significant rounding of the surface of the small surface area samples. Most often we briefly electro-polish away the damage layer from the 1μm lapping step and then follow with lowering the voltage to get either an etched microstructure, or further lower of the voltage, to get an anodized layer for revealing microstructural details. Figure 2 shows examples of electro-polished vs. an anodized surface preparation in a delta-stabilized Pu-Ga alloy. Imaging is performed on an Olympus PME-3 (figure 11) in glovebox #1. This metallograph was chosen as it is upright in design, thus requires a small footprint inside the glovebox. A digital camera

with a USB electrical feed-through connection to an external PC allows for live viewing and single image acquisition with any number of imaging acquisition software packages. We currently use a Zeiss AxioCam 503 camera with the Zeiss imaging software for image capture. A second macro style camera with portable mini-screen is mounted inside glovebox #1 near workers hands and allows for workers to view small sample handling. With this macro-camera there is an additional RCA style feed-through on the glovebox so that the video camera feed can be recorded as video or still-frame format on a PC. There are several reference sources that we have frequently referred to regarding metallography preparation techniques and microstructure evaluation for Pu and Pu alloys: (7, 8, 9).

Surface Metrology Characterization by Interferometry: As part the development and production process of precision-engineered samples for laser-based experiments we utilize a Zygo white-light interferometer for the quantification of surface roughness, flatness and sample step-height measurements (11). The need for this level of surface quantification arose as a requirement to precision engineer plutonium samples for high-energy-density experiments where surface roughness requirements are below 50nm, surface flatness's are >0.2µm over mm distances and sample thickness needs to be measured to 1µm accuracy. The interferometer is located outside the glovebox (figure 12a) and is also utilized, as the TEM and other instrumentation, by non-radiological workers for inert materials metrology. In order to facilitate safe transfer and to minimize sample oxidation during measurements we transfer the samples in a custom built windowed encapsulation sample holder (figure 12b). Samples are loaded into the encapsulation devise and sealed while under the inert atmosphere of the glovebox. Encapsulated samples are then passed out via the antechamber. The inert atmosphere of the glovebox is trapped inside the encapsulation device thus minimizing oxidation. After assuring the external surface of the encapsulation device are contamination-free (while in the fumehood attached to the exit side of the ante-chamber) it can then be transferred to the interferometer for surface metrology measurements. The encapsulation device window affects the interferometer optics (both phase and path-length); to compensate for this a similar window is placed in the reference leg of the Zygo optics. Without this compensation capability interferometry would not be possible. Reference 4 details the surface measurement and encapsulation process. While encapsulated, typically a few hours, there is little to no observable oxidation of the sample surfaces, even for pure alpha Pu as the encapsulation device is O-ring sealed and (again) has the dry, inert, atmosphere of the glovebox sealed inside. Figure 13 shows a typical example of measuring surface finish and flatness as a function of lapping film particle size. Figure 14 shows a typical step-height measurement of an alpha-Pu part that has been lapped to <10um in thickness using techniques described in ref 4. For example, from this precision lapped foil a 2mm diameter disc will be laser cut from the central region. The surface flatness within the central 2mm diameter region is < 0.2um.

Laser Micro-Machining: In order to precisely shape the lateral dimensions of very small samples for a variety of experiments we have developed a safe laser micro-machining process. This process, like so many that we describe here, allows for the safe handling and encapsulation of plutonium samples and control of associated contamination. Specifically for laser micro-machining, we have developed and utilize an

optically transparent encapsulation sample holder with a transparent window that allows for imaging and laser micro-machining with precision to the micron level. Figure 15 is an image of our encapsulation system where the outer window is made of CR-39, an impact resistant plastic that is non-absorbing for the three wavelengths (red, green and UV) available on the Quick-Laze Tri-light system from New Wave Research (figure 16). We chose the Tri-light system as it is compact, is integrated into a light microscope system for viewing the precise location of cutting and cutting in progress, and has computer aided drawing (CAD) and CAD file import capability. The power of the system is relatively low, and having a repetition rate of only 50HZ, the ability to cut through thick specimens (>100um) leads to very long cutting times, e.g. 4-6 hours for a 2 mm diameter discs. However, with the low power there is little to no heating of the sample.

Figure 17 shows several examples of precision laser cutting from thin foils of plutonium samples for experiments ranging from X-ray diffraction, resistivity and high-pressure diamond anvil cell (DAC) equation-of-state.

As with other sample loadings, sealing and transfers, these samples are loaded into the encapsulation holder while positioned in the antechamber of the glovebox under inert atmosphere of the glovebox. With the use of clean tools, a lay-down of clean Al foil and removable tape covering of exterior surfaces of the two pieces of the encapsulation device, it is routinely possible to load, seal and pass-out the encapsulated sample with little to no external contamination present.

A short procedural description for mounting and sealing for laser cutting: The samples to be laser-cut are mounted with Sally Hansen cyanoacrylate glue to the sample mount that fits into the encapsulation device. **Note:** The sample mounts' top surface has been modified having a glass slide epoxied to it. Using a bare metallic mount will often lead to the micro-welding of the sample to its' surface. The cyanoacrylate is non-absorbing to the laser light and does not appear to interact with the samples; even when coated over the surface of the sample and the laser cuts through it. The mounted sample is placed into the open encapsulation device as it sits on clean aluminum foil in the antechamber. The taped-over lid to the encapsulation device is placed upon the base and twist-locked into place. The sample and surrounding inert atmosphere is now sealed. After removal from the antechamber and into the adjoining fumehood the anti-contamination wrapping is removed and the external surface is decontaminated (if necessary) then transferred to the stage of the laser cutter. After laser cutting the encapsulated sample is returned to the antechamber, exterior surfaces area wrapped with tape as an anti-contamination layer and passed into the glovebox after the antechamber is purged with inert gas. The mounted laser-cut sample(s) are removed in a manor that is opposite of loading. Acetone is used to dissolve the cyanoacrylate glue in a jar that is lined with removable filter paper, or similar material, that can be used to remove the samples from the acetone without directly handling the samples, e.g. with tweezers.

Diamond Anvil Cell Sample Preparation and Loading: For high-pressure and temperature equation-of-state diamond-anvil-cell (DAC) experimentation performed at advanced X-ray synchrotron facilities (12, 13) on Pu and Pu alloys (14), we have developed a process for preparing precision micro-specimens and the ability to safely load the microscopic samples into sub 150um diameter wells within the DACs. Samples are typically lapped to specified thickness, e.g. 10-50µm, then laser micro-machined into

desired shaped, e.g. 20-50μm diameter discs or triangles. The lapping of samples is similar to other sample preparation processes that we have employed for metallography, and as previously described (4), with the additional step of lapping both sides of the sample in a serial process to achieve the final desired thickness. For lapping of delta-stabilized materials that may have undergone a surface phase transition from the lapping process a low-temperature anneal of ≈ 100ºC is incorporated to revert any transformation phase as this would be artifact. Thickness of the sample while lapping is measured with a precision micrometer having an accuracy of +/- 1um. If need be, more precise thickness measurements can be made using the Zygo interferometer. Samples are typically laser cut as described in the prior laser micromachining section. Alternatively, brittle samples can just as easily be ground with a mortar and pestle to a desired size range. The final thickness of the samples is dependent upon the type of experiment. For transmission experiments, thicknesses that allow for considerable transmission of typical energies available at photon sources are in the 8-15um range. For reflection diffraction or experiments that detect outgoing florescence, the samples are more often in the 50um range. Laser cut samples are removed from the laser-cutting mount with acetone, rinsed and transferred (typically in a covered Petrie dish inside a plastic bag) to the DAC loading system (figure 18). This portable system features a long working distance, high resolution, stereoscope and video imaging system that allows the operator to pick up the micro-samples with a sharp needle and precisely place them within the opening of the DAC well. Sealing of the DACs is performed within the enclosure so that inert atmosphere is trapped within the cell for maximum oxidation protection. In order to minimize contamination of the outer surfaces of the DACs they are wrapped with heat-shrink plastic prior to insertion into the DAC enclosure loading system. The DACs are then bagged in clean bags and passed out of the loading system and into a fumehood for removal of the anti-contamination wrapping and their exterior surfaces are cleaned to background levels. Once sealed, specimens appear to have a shelf-life of up to ≈ 60 days, leaving time to ship to experimental facilities. Beyond 60 days there is typically measurable oxidation of metallic samples. The DAC loading system is portable, thus allowing us to remove it from the lab space and to roll in other instrumentation as needed.

Laboratory Based X-ray Diffraction: Samples are typically prepared by metallographic processes previously described here, then sealed in a commercially available encapsulation device (figure 19A) that allows for the penetration of typical energies available with laboratory X-ray diffraction systems. Metallic samples are typically electro-polished as a final step. Electro-polishing removes any surface damage that is present, as well, reduces the rate of surface oxidation. Powder samples are typically mixed in vacuum grease or oil to minimize their dispersal. It is common practice to place a microscopic amount of Si or Cu powders on the surface of the samples as an internal calibration for lattice parameter determination. Again, with the use of an antechamber, a clean aluminum foil surface in the ante-chamber to place the encapsulation parts on, anti-contamination wrapping and clean tools as needed, it is routinely possible to air-lock in the encapsulation device in an open configuration, load the sample, then using a clean tool, close the encapsulation device with inert atmosphere sealed inside. Figure 19B shows the sealed encapsulation holder from Bruker Inc. in place in a Bruker diffractometer. Figure 19C shows typical examples of a powder

diffraction pattern from alpha and delta stabilized plutonium. Routinely, minimal diffraction from surface oxide is detectable.

Synchrotron/Advanced Photon Source Experimentation Using Transmission/Reflection Specimen Holders: We have prepared many bulk and thin specimens by either the before mentioned lapping process, the jet-polishing technique (ref 5) and/or the window-technique electro-polishing techniques (ref. 17) for X-ray transmission, absorption, emission and back-reflection for experimentation at advanced X-ray synchrotron facilities. Figure 20 shows several sample holders that typically have 2 to 3 layers of Kapton (or other low density material/windows, e.g. Be) for the multi-layered encapsulation of the radiological sample and for contamination/oxidation control. For many of the specimens for these experiments the first layer of encapsulation is a dip-coating of liquid Kapton *. The Kapton is cured at a low temperature (<100ºC) inside the glovebox on a low temperature hotplate. The thickness of this first coating layer is typically less then 25um. The thickness of the Kapton coating can be varied by changing the viscosity of the liquid Kapton. We change the viscosity by mixing the liquid Kapton with a thinning agent, butyrolactone. Porosity of the Kapton coating has never been observed. The Kapton has shown excellent stability under constant alpha irradiation from Pu239 based samples. For samples containing higher amounts the Pu238 isotope we have noticed degradation of the first layer of Kapton. Typically alpha particle radiation is still observable (albeit low) through this first layer, yet oxidation is significantly reduced. Samples are then mounted and sealed inside their respective holders having additional layers of Kapton or other low absorption materials relative to the probing X-ray source and out-going emission/transmission signal. The Kapton and/or e.g. Be windows typically have small gold or indium wire seals on their outer perimeters and are held in place with small screws through an annular frame. These samples have been shipped to numerous synchrotron facilities for unique experiments ranging, but not limited to, phonon dispersion curve measurements in single grains (crystals) of delta-stabilized plutonium (ref 18, 19), EXAFS experiments on alpha-prime, delta-phase plutonium and PuO_2 (ref 20, 21), EXAFS measurements of nearest (atom) neighbors distances in several Pu metals, alloys and compounds (22, 23), resonant absorption near-edge X-ray emission (REXS) and X-ray absorption near-edge structure (XANES) (25), Phonon density of states by inelastic X-ray scattering in a polycrystalline Pu(Ga) alloy (26), and transmission X-ray thermal diffuse scattering for phonon density of states measurements (27).

Resistivity Experimentation Combined With Heavy Ion Irradiation: We have prepared thin foil samples for 4-point resistivity measurement experiments. For the resistivity experiments foils of delta-stabilized Pu(Ga) material were formed by using a small rolling mill to a near final thickness of ≈10µm. Foils were cross-rolled between thin Ta sheets with 450ºC/2hr vacuum annealing iterations in order to achieve a uniformly thin foil. Foils were mounted into 4-point resistivity holder (figure 21) within the glovebox and checked for electrical continuity while positioned on a contamination-free tray. The sample and mounting fixture was then dip-coated in Kapton for a first primary seal for contamination and oxidation control. The encapsulated samples were air-locked

out via the antechamber and checked for external contamination and cleaned to minimal levels. A portable vacuum system which could also be operated as a glovebox enclosure (figure 21) was rolled into our sample preparation lab where the mounted resistivity samples were transferred (via a bag-in process) into the vacuum system (at atmospheric pressure and actively purged with dry N_2) and mounted on the end of a cryogenic cooling rod; electrical connections were made and checked for continuity followed by vacuum evacuation of the chamber. The chamber was then transported to the accelerator lab and connected to a 4 Mev heavy ion irradiation accelerator. The Kapton coating is essentially transparent to the high-energy ions, hence there was no noticeable degradation of the Kapton upon removal of the sample after the experiments were finished. As well there was little to no released contamination within the vacuum chamber. Resistivity measurements were made as a function of dose, temperature and time. See reference (28) for technical results and discussion.

Magnetometry Measurements: Samples prepared for magnetometry measurements have been prepared by standard slicing, lapping and electro-polishing processes as described herein. For these samples it was imperative to not use tools and handling tweezers that contained magnetically susceptible materials, e.g. Fe, Ni. After electro-polishing ≈ 1mm x 1mm x 2 mm samples, samples were dip-coated in liquid Kapton and cured. Several layers were applied. As can be seen in figure 22 the sample is then glued in place using GE varnish (which can be thinned or dissolved in ethanol) onto a half-circle high-purity brass tube. At this point, the assembly can be transferred out of the glovebox, cleaned of external removable contamination and placed in a glovebag located inside a fumehood. The half-tube assembly is sealed inside a brass tube having a compressed Au seal on one end. The glovebag is purged with dry nitrogen so as to displace any moisture that could be trapped inside the assembly, as the sample and encapsulation device will see cryogenic temperatures, thus minimizing condensation of ice on the sample. Reference (29) contains the results of these experiments.

He Ion Beam Implantation Experiments: In glovebox #2 we made use of a small hand operated rolling mill to produce a uniformly thin, ≈ 100um thick foil of delta-stabilized Pu(Ga) alloy. An iterative process of annealing (≈440oC for 4 hrs) and multi-directional 5-10% rolling reduction (between 250um thick Ta foil) was utilized to get to final thickness. The foil was then high-vacuum annealed @ 440^{o}C for several hours to remove mechanical damage. Surface oxide was removed by polishing with 1um diamond paste on a cotton-tipped applicator as the sample was mounted with dissolvable adhesive (Sally Hansen**) on a secured mount. Once the sample was clean of visible oxide, the sample was dip-coated in liquid Kapton and heat-cured at ≈100^{o}C for several hours. The foil specimen, while wet with Kapton, is placed on a sheet of Teflon (as a nonstick surface) so that once the heat-cure is finished and the Kapton is hardened, it is removable from this surface that it is sitting upon. The specimen was then loaded into a vacuum transfer chamber, sealed and evacuated, then transferred to the Center for Accelerator for Mass Spectroscopy at LLNL (30) for He implantation. After He implantation, the sample was returned to our facility and prepared for TEM studies characterizing the He bubble size distribution at an age equivalent of 200 years of He ingrowth from the Pu239 decay process. (This work has not been published) The Kapton coating processes in this and

many other sample sealing processes has worked quite well. The ability to take advantage of the properties of liquid Kapton for sealing and protecting samples has been very advantageous and cannot be understated. Additionally, we have working evidence (by appearance of color) that even over a period of 2 years that the amount of oxide growth on well polished and Kapton dip-coated samples can be less than 50nm for delta-stabilized Pu-(Ga) alloys.

***Differential Calorimetry Experiments*:** Samples prepared for differential scanning calorimetry (DSC) are straightforward. The basic requirements are that they are small enough to fit into the pan, have the oxide removed (typically by electropolishing) and have at least one flat surface. Having a flat, oxide free surface, ensures that there is good thermal contact between the specimen and the DSC pan. Without good thermal contact getting good thermal signal, with minimal time delay, would be difficult to achieve. We use a hermetically sealable pan and lid supplied by the manufacture (31). The pan has a threaded portion that allows for a screw-top lid to compress the gold foil lid in order to make a hermetic seal. The hermetic seal allows for contamination control of the radiological sample and to contain inert atmosphere around the specimen thus minimizing oxidation. The oxidation process is not only the primary source of radiological contamination, but reduces the thermal conductivity of the specimen, which again is vital for good DSC measurements. Figure 23 shows the DSC, the pan sealing system, and typical results from an exothermic reaction from a martensitic transformation in a Pu(Ga) alloy upon cooling below room temperature. References (2, 33, 34) are examples of our DSC efforts regarding phase stability in the Pu(Ga) system.

Micro-metallurgy: In glovebox #2, we have a small UHV resistive heating furnace having a base pressure of < 5.0e-9 Torr and a temperature range from room temperature to >1400^{o}C, figure 24. At high temperatures the vacuum conditions temporally degrade from outgassing to the high 10e-5torr range but quickly recover into the low 1-e-7 torr range. The primary uses of this furnace system have been to 1) melt plutonium samples in order to evolve any He gas that has in-grown as a result of the radiological decay of Pu239, 2) to near-net-shape small sample in the liquid state so that final mechanical processing, lapping and polishing is minimized, 3) to anneal plutonium alloys for solute homogenization, and 4) to make alloys. With UHV conditions there is minimal oxidation at high temperatures, hence loss of material from small samples is minimized. This is important for working with small sample volumes else the majority, if not all, of the sample materials would be consumed by oxidation. As well, with near-net shaping we can make maximum use of the small masses (e.g. <50mg) of material that we are allowed to have in our radiological facility inventory. Figures 25 shows an alloy of Pu-4.3at%Ga that was produced by melting the two pure elements together in a Ta-10W at 900^{o}C in UHV conditions. This small (≈75mg) ingot shows little oxidation. The near-net shaping of pure Pu metal into small thin plate shaped samples has also been produced under UHV conditions which have resulted in minimal oxidation. Near-net-shaping processes that we have utilized is facilitated with having machined flat-bottom Ta-10W crucible with a floating piston (of same material) that presses the liquid metal into thin plates. The thickness of the plate is controlled by an annular washer (Ta-10W) of specified thickness (as thin as 100um) that sits at the bottom of the crucible and acts as a

stop for the floating piston. We have found that for the Ta-10W if we anneal the parts in air @ 500°C for 1-2 hrs to form a black color oxide that there is little to interaction with the liquid Pu at high temperatures. Higher temperatures and longer times will produce a white oxide and this has not worked so well as a barrier to liquid Pu.

Conclusion

The combination of small sample handling and preparation methodologies, adaptation of conventional sample preparation techniques to the environment of a glovebox, and a custom built room and gloveboxes has lead to versatile, efficient, reproducible sample preparation of actinide samples for a wide variety of small-scale experimentation. When we combine our sample preparation capabilities with encapsulation into sample holders that can facilitate safe handling and experimentation outside of the glovebox, a wide variety of scientific measurements can be made safely and routinely.

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find Kerri's phase transformation reference

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- Liquid Kapton, "Probimide 9753" available from Microelectronic Materials, Inc. West Patterson, New Jersey 07424 USA.

** SallyHansen ™ cyanoacrylate single component, brush-on, epoxy (dissolvable in acetone).

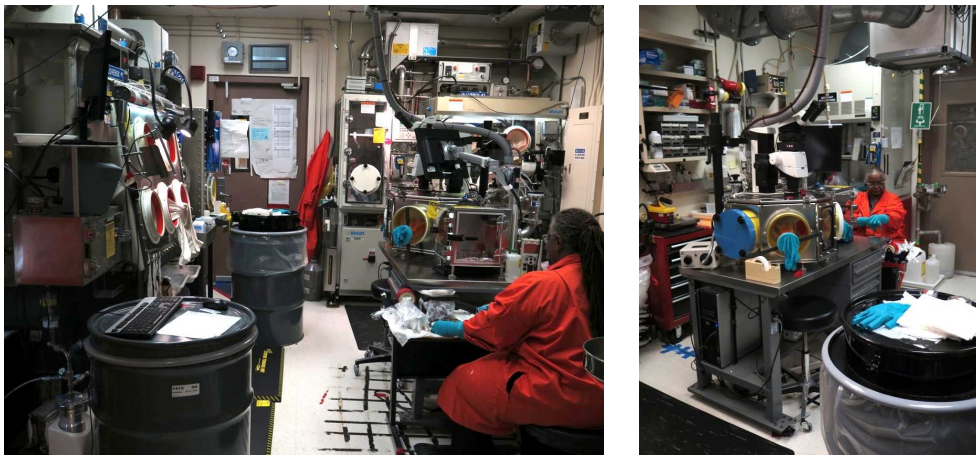
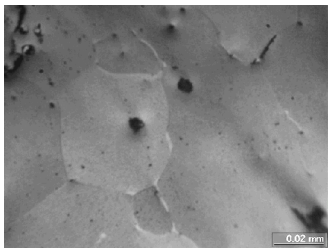
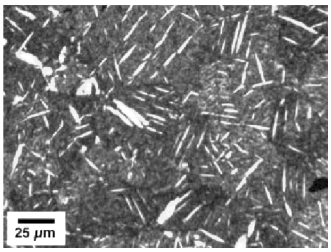


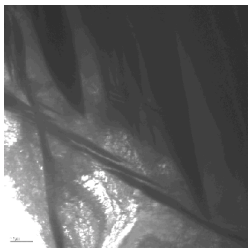
Figure 1 Radiological sample preparation facility at LLNL designed and fabricated for small scale, integrated, precision sample preparation in support of characterization and experimentation.



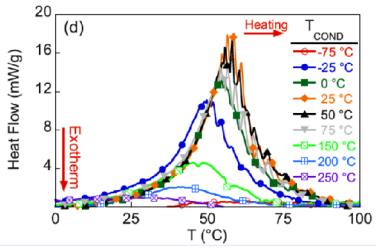
BF metallography of an as-annealed Pu-1.9at%Ga alloy showing equiaxed delta-phase grains with typical grain boundary and inter-granular second phase precipitates from impurities. Final sample preparation is electro-polishing.



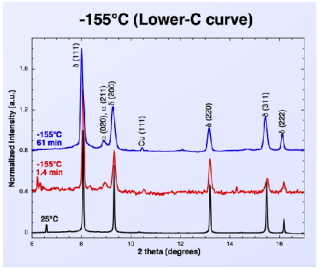
BF image with polarization of the same Pu-1.9at%Ga alloy partially transformed to alpha' at -155C for 4 hrs. Final sample preparation was electro-polishing followed by electro-anodization.



BF TEM image of the same Pu-1.9at%Ga alloy partially transformed to alpha' at -155C for 4 hrs.



DSC results from a series of partial delta to alpha' cooling experiments where the sample was conditioned at 9 different temperatures to measure the effect of conditioning on the amount of phase transformation when cooled to -155ºC. heat flow is the measure of the amount of transformation.



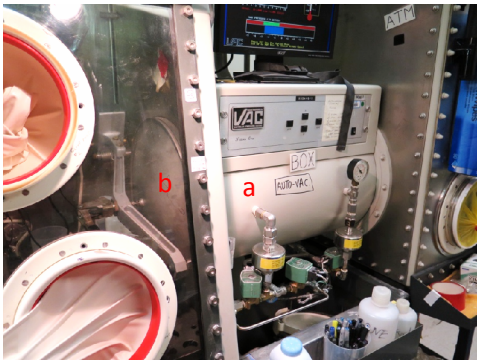
In-situ X-ray diffraction of the same Pu-1.9at%Ga alloy partially transformed to alpha' at -155C.

Figure 2

Figure 2 Excerpts of series of metallographic imaging, X-ray diffraction, DSC and TEM results from a series cooling experiments on one small piece (≈250mg) of a δ-stabilized Pu-1.9at%Ga alloy demonstrating the ability to perform multi-characterization experiments on a small quantity of material. From: "Evidence For Nascent Equilibrium Nuclei as Progenitors of Anomalous Transformation Kinetics in a Pu-Ga Alloy" J. R. Jeffries, K. M. Blobaum, M. A. Wall and A. J. Schwartz, Physical Review B, vol. 80, no. 9, Sept. 17, 2009. and "In-situ X-ray Diffraction of the Delta to Alpha-Prime Transformation in Pu-Ga Alloys" K. J. Blobaum, J. R. Jeffries, M. A. Wall, H. Cynn and W. J. Evans, Available at https://e-reports-int.llnl.gov/pdf/400747.pdf

Figure 3 A and B

A



B

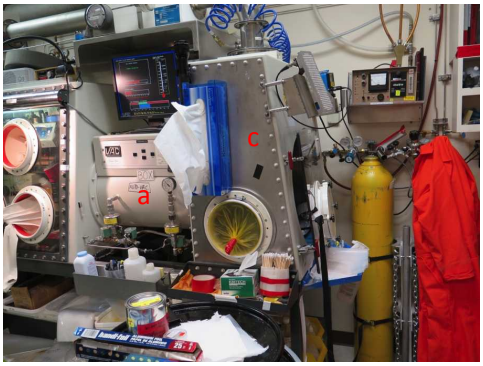
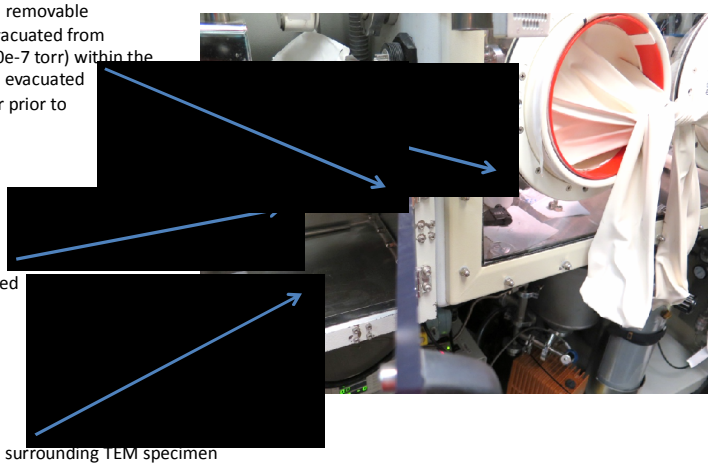


Figure 3. Pass-through ante-chamber (a) with inner and outer guillotine doors. The left-hand door (b) seen here on the inside of the glovebox seals one side and the other end of the ante-chamber is sealed with an Identical door that is located inside a fumehood (c).

Figure 4



Airlock with internal removable cover that can be evacuated from a vacuum source (10e-7 torr) within the glovebox in order to evacuated the specimen holder prior to transfer to the TEM

Double-tilt vacuum-transfer TEM specimen holder inserted In the airlock on the side of Glovebox #2.

Fumehood surrounding TEM specimen holder allows workers to remove the evacuated specimen holder and survey without wearing respirators. Any contamination on the external surfaces of the specimen holder that was exposed to the internal glovebox atmosphere can be cleaned off prior to removal from the fumehood.

Buehler Isomet saw

https://shop.buehler.com/

Figure 5 – Isomet dicing saw

Figure 6 – Lapping device and lapping supplies

Lapping supplies from South Bay Technology. Model 150 lapping device, diamond lapping Films 30, 9, 3 an 1 um grit, flat lapping plate Digital micrometer and Sally Hansen Cyanoacrylate epoxy and ethanol for Lapping solution.

http://www.southbaytech.com

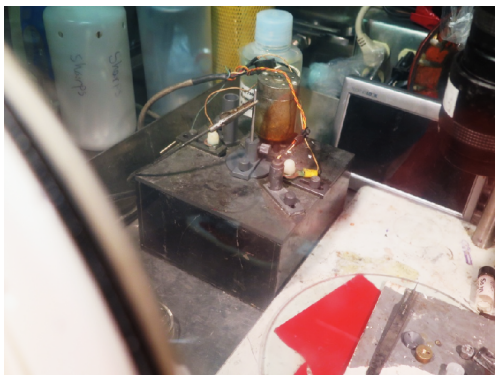


Figure 7A

Figure 7A Photo of a Fischione TEM electro-polishing unit as see through the window of glovebox #1.

The power supply for the electro-polishing unit is located and an electrical feed-through on the back wall of the glovebox allow for remote control.

Figure 7B

External power supply for Fischione TEM electropolishing system. A multi-pin feed-through connects the power supply to the polishing unit inside the glovebox as seen in figure 7A.

Figure 8 Gatan Dimpler for pre-thinning a TEM disc Prior to ion milling.

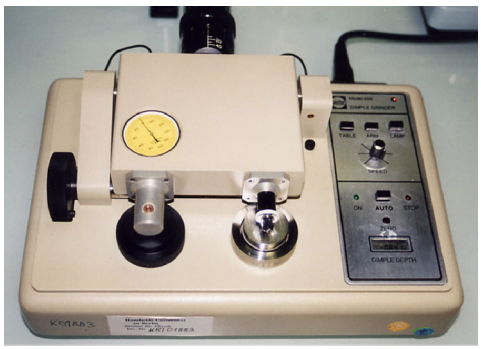
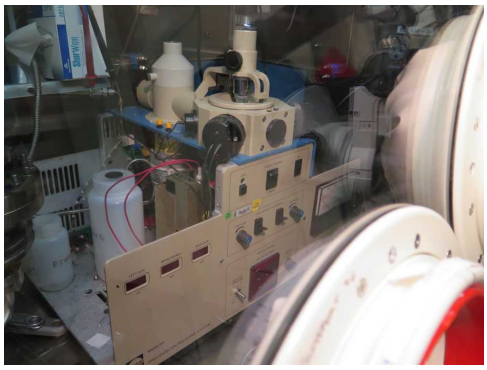


Figure 9 PIPS ion milling system for final thinning Of TEM sample.



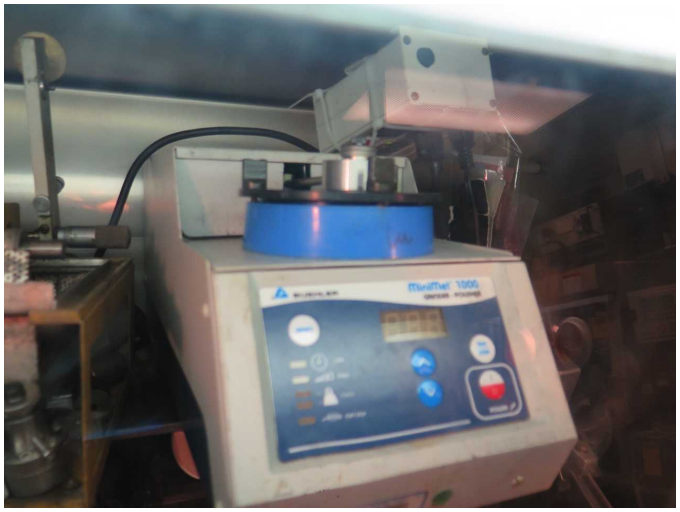


Figure 10 Beuhler Mini-Met lapping instrument. Modified to hold the SouthBay Technology Model 150 lapping device.

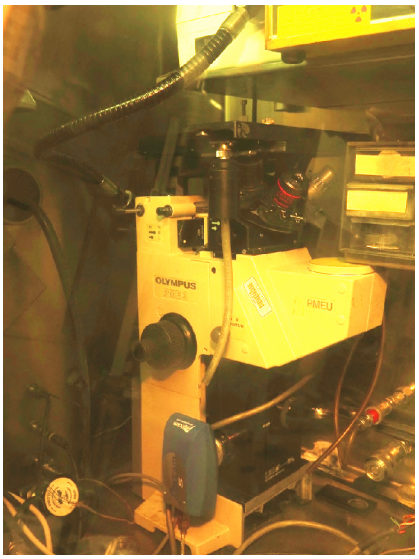
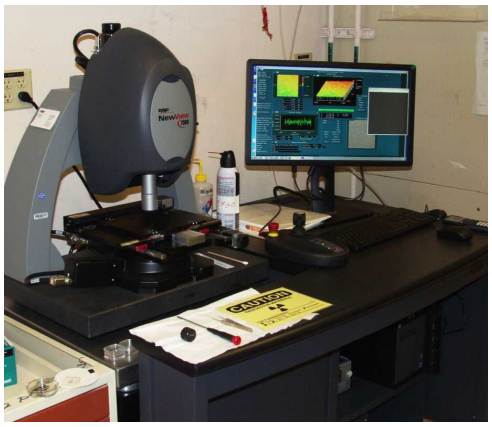


Figure 11 Optical light microscope. Olympus PME-3, inverted and up-right with digital camera, BF, DF, DIC and polarized capability.

Figure 12a,b. Zygo white-light interferometer and encapsulation holder



12a Zygo NewView 7000 white-light interferometer.

12b Image of a specimen mount (with specimen) being loaded into an opened Zygo encapsulation specimen holder in the ante-chamber of the glovebox. Inset image is of the sealed encapsulation specimen holder with top-center window.

Figure 13 Zygo white-light interferometer measurements of surface roughness and flatness measurement of precision lapped Pu foil. The pure Pu metal sample was lapping on 1um diamond lapping film using the SBT gravity-feed lapping fixtures.

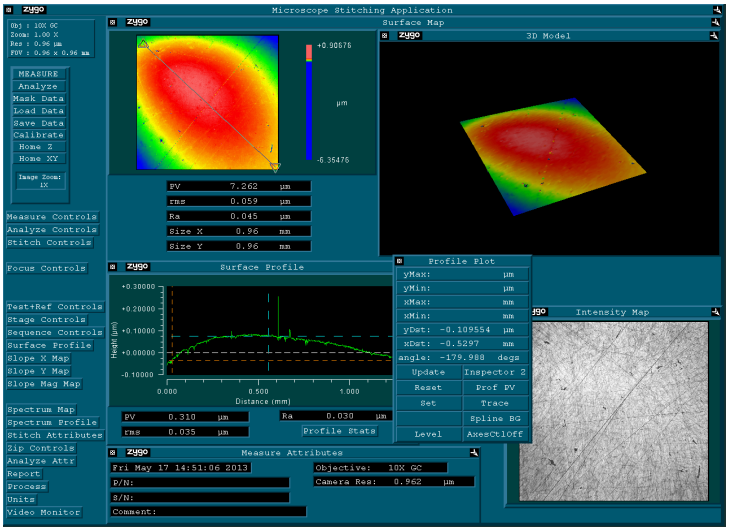


Figure 14 An example of a step height measurement of a precision lapped foil of pure Pu metal foil that will be used for transmission X-ray diffraction experiments. The step height measurement
(sample surface to sample mount surface) indicates a sample thickness of ≈ 8.9 um which includes the cyanoacrylate adhesive which we typically estimate at less than 1um in thickness. Surface flatness over a central 2mm diameter is within 0.2um.

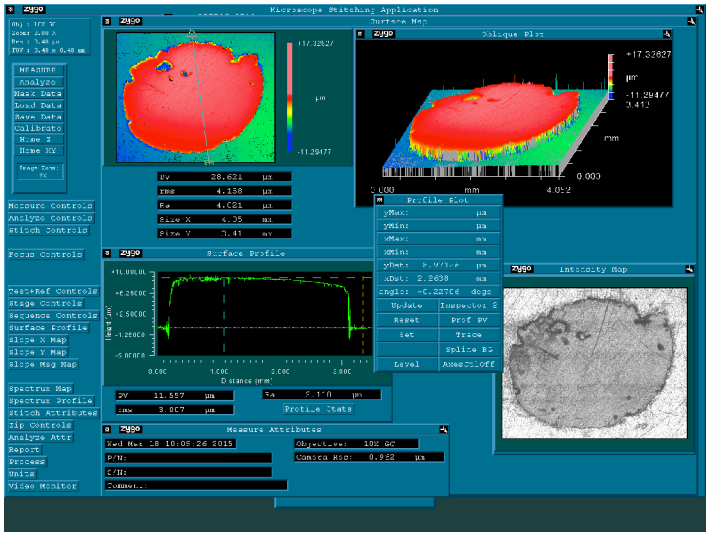


Figure 16 New Wave Research model Quick-Laze 50, tri-light laser micro-machining system with CAD drawing import for precision laser X-Y control. Stage precision is ≈ 1um with a range of spot-sizes from 5 – 100um.

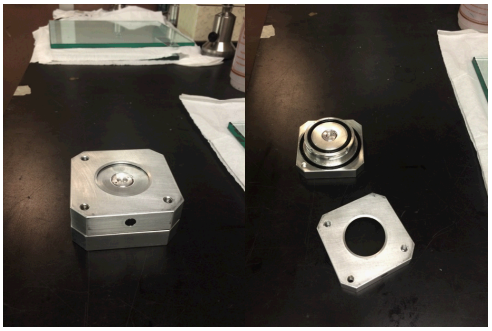
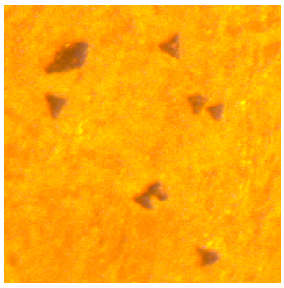


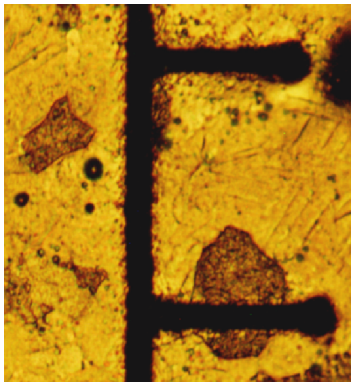
Figure 15 Double-layer encapsulation system for encapsulation of radiological and/or reactive samples. Contains ablated radiological material and minimizes oxidation of samples as inert atmosphere is sealed inside.

Figure 17 Examples of laser cut Pu samples

1. Laser cut triangles, 50um on edge, for diamond anvil cell high pressure experiments.
2. Laser cutting surface marking of a partially transformed (2-phase, delta grains and alpha' plates) specimen for high spatial resolution orientation and strain mapping experiments at the APS facility. The laser cut regions will not yield Laue patterns. combined optical map with point-by-point Laue map will facilitate alignment of microstructure with the back-reflection Laue X-ray diffraction data.

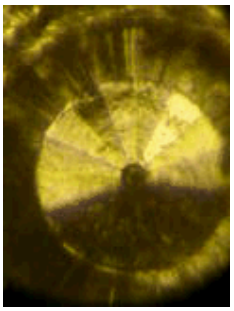


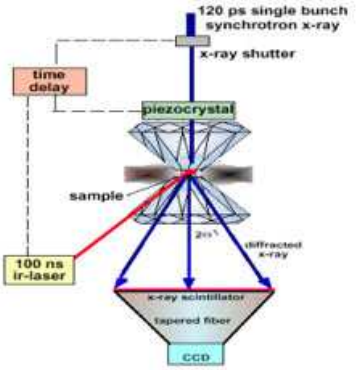
1. Laser cut triangles, 50um on edge, from 10um thick alpha Pu foil



2. Laser cut trenches 5um wide relative to 25um delta grains containing alpha' plates. trenches will yield no back-reflection Laue diffraction' patterns thus allowing correlation of optical microstructure image with strain-mapping.

Figure 18. Photograph of workers loading micro-samples into diamond anvil cells within a double-layered safety enclosure. The inner layer is a engineered plastic glove-bag that is replaced when the internal contamination gets too high thus decontaminating them to background levels becomes to difficult.





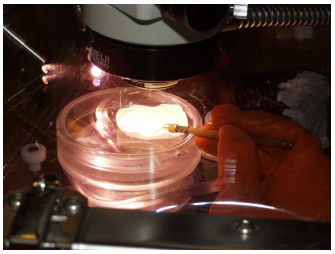
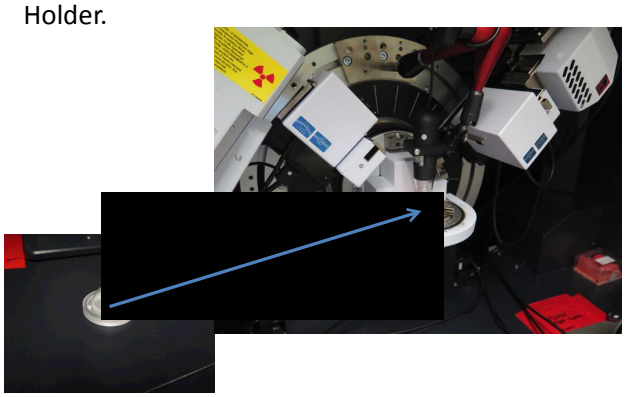


Figure 19A) Photograph of a Bruker environmental specimen holder that contains a sealed Pu sample. Sealing contains radiological contamination and holds the inert Atmosphere around the sample thus minimizing oxidation over time.

B) Image of the sealed environmental holder in place within the Bruker X-ray Diffractometer.

C) Example of a X-ray diffraction spectra acquired from delta-stabilized Pu-Ga alloy and Alpha Pu. The large
Smooth peak at < 20 deg theta is from the amorphous plastic material of the environmental Holder.



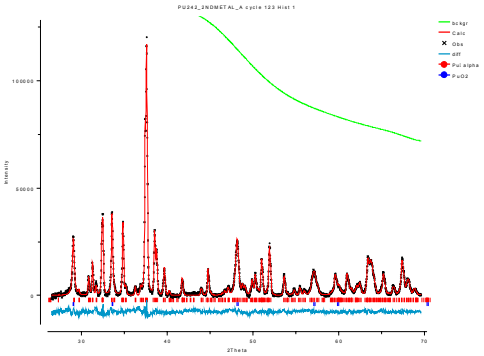
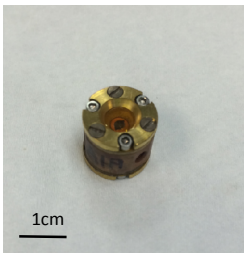
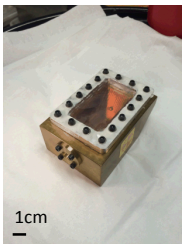


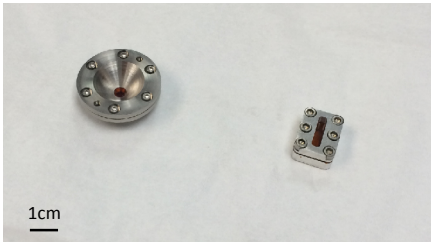
Figure 20



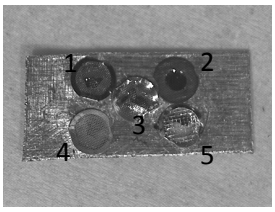
Triple sealed metallic Pu sample for
Transmission X-ray diffraction. Sample
Is sealed in a 1st layer of dip-coated and
Cured of Kapton. Two more layers of
Kapton windows ≈ 100um thick add
Assurance to keeping potential contamination
Inside, as well to reduce oxidation of the small
(10um thin X 3mm diameter) sample.



3mm diameter delta-phase Pu
Alloy specimen dip-coated sealed in
A Kapton windowed back-reflection
Laue sample holder for focused probe
Strain-mapping experiments at the APS
Facility.



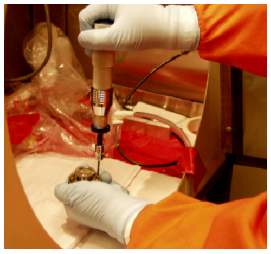
Triple sealed metallic Pu sample for
Transmission X-ray diffraction, small angle scattering and
XAFS. Sample
Is sealed in a 1st layer of dip-coated and
Cured of Kapton. Two more layers of
Kapton windows ≈ 50-100um thick add
Assurance to keeping potential contamination
Inside, as well to reduce oxidation of the small
(10um thin X 3mm diameter) sample.



Example of 5 different sample all thinned
to less than 10um for EXAFS experiments.
Shown here prior to final encapsulation inside
a Kapton windowed system.

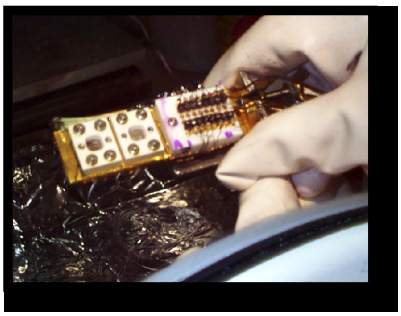
1. Alpha Pu on 3mm TEM grid
2. Alpha U e-polished
3. Delta Pu-Ga on 3mm TEM grid
4. Am on3mm TEM gird
5. Cm on 3mm TEM grid

6. EXAFS experiments

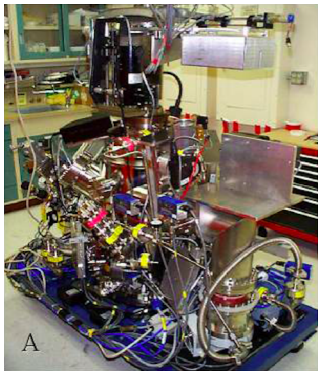


Secondary sealing of muon experimental holder. The
5mm diameter disc. Alpha-Pu sample is first dip-coated with Kapton
And sealed inside this stainless steel housing with
A front Be window.

Figure 21) 4-point electrical resistivity combined with heavy ion irradiation.

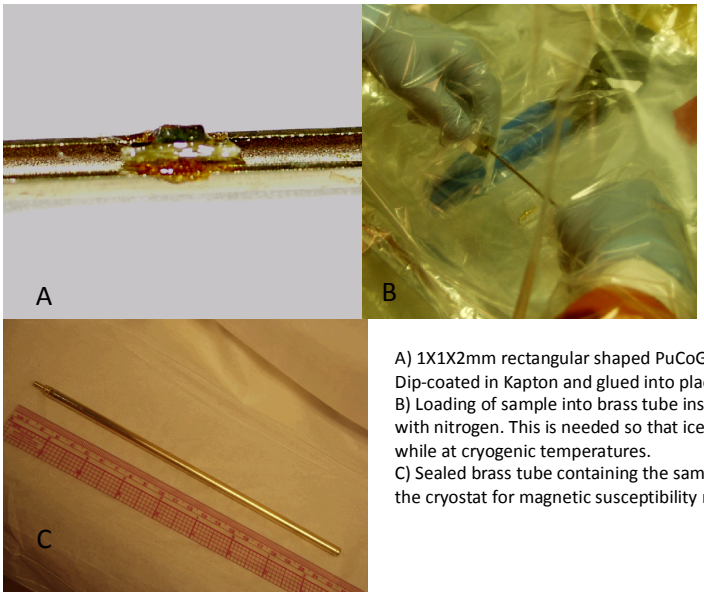


2 each, Delta-stabilized Pu-Ga foils in a 4-point resistivity holder with electrical connections. After mounting and checking electrical continuity The sample regions of this specimen holder are dip-coated in liquid Kapton and then warmed for curing. The solid Kapton minimizes oxidation of the specimens as well as controls contamination. The Kapton is non-conductive thus will not affect the resistivity measurements, and its thinness is transparent to the ion irradiation.



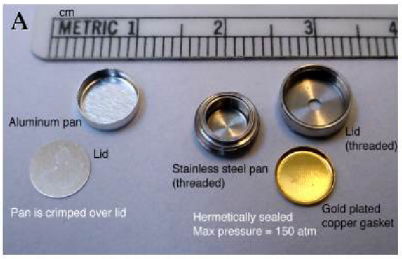
Combination vacuum system/portable glovebox. The electrical resistivity sample holder is bagged Into the vacuum system and attached to the vertical cryostat. Electrical connections are tested and then the chamber is evacuated to high vacuum. Next the chamber is transferred and attached to a 4Mev ion accelerator for combined resistivity heavy ion Irradiation experiments.

Fig. 22 sample preparation and sealing for magnetic measurements

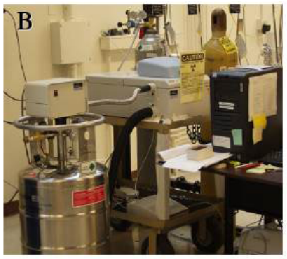


A) 1X1X2mm rectangular shaped PuCoG5 sample that has been Dip-coated in Kapton and glued into place using GE varnish.
B) Loading of sample into brass tube inside a glovebag that is purged with nitrogen. This is needed so that ice will no condense on the sample while at cryogenic temperatures.
C) Sealed brass tube containing the sample and ready for loading into the cryostat for magnetic susceptibility measurements.

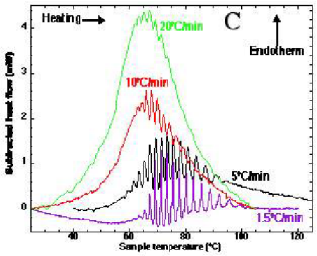
Figure 23) Differential scanning calorimetry.



A) Showing the size and differences between the typical crimp-seal DSC pan and lid (right side) and the Au sealed pan-seal-lid combination used for hermetically sealing reactive/radiological samples in for DSC measurements.



B) Differential Scanning Calorimeter (DSC). The instrument is rolled into our Pu sample preparation lab, and sealed samples are loaded into the instrument. The DSC is rolled back to another lab where measurements are taken.



C) Example of endothermic measurements burst-like martensitic transformation phenomena during the cooling of a Pu-Ga alloy at different cooling rates.

Figure 24) Ultra-high vacuum furnace in glovebox #2.

UHV vacuum furnace

Examples of small Ta-10W crucibles used for melting Pu and Pu alloys. Machined or formed crucibles are annealed in air prior to use @ 500C for 2-4 hrs. The dark oxide-nitride layer that forms protects against chemical reaction with the molten Pu materials.

Figure 25 Example Of making an alloy In UHV furnace

A 75mg alloy (pu4.3at%Ga) made By melting the 2 elements in a Ta10W crucible @ 900oC for 15 minutes under UHV conditions. The dark golden color indicates that The thickness of the oxide is in the 20-40 nm range. The diameter of this Ingot is ≈ 1.5mm.