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Controlling Plutonium Oxidation State for the Development of a Reliable Solution-Phase Assay

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The redox mobility of plutonium is both a blessing and a curse; thus, controlling the oxidation state of plutonium lies at the heart of its solution-state chemistry. One important target oxidation state is Pu(VI) (PuO_2^{2+}) due to its structural similarities to UO_2^{2+} and NpO_2^{2+}, its interaction with common extractants used in separation schemes, and its strong optical absorption peak that can be used for solution-phase quantification. The formation of PuO_2^{2+} typically requires strong chemical oxidants such as ozone, silver(II), cerium(IV), or dichromate. Once formed, PuO_2^{2+} typically survives in isolation for a few hours to a few days depending on environmental and solution conditions. When PuO_2^{2+} is the target for a quantitative analytical assay, it is crucial to understand its stability to ensure reliable and robust measurements. This work investigates the formation and subsequent stability of PuO_2^{2+} in cerium(IV)-containing nitric acid media using near-infrared optical absorption spectroscopy (NIRS). Our work identifies conditions that stabilize PuO_2^{2+} in solution without the exclusion of air for several months. Additionally, we confirmed that sulfate is an excellent ligand for PuO_2^{2+} and readily complexes the ion even in extreme excess of nitrate. These results demonstrate that controlling plutonium oxidation state and speciation in solution enables the development of a robust and reliable quantification technique.

Strain Rate Sensitivity in Actinide Materials Using Nano-Indentation Measurements

Veronica Anghel, Zachary S. Levin, Isaac A. Hernandez, Todd P. Martinez

In recent years, nanoindentation has emerged as a powerful technique for investigating the mechanical response of materials at the micro- and nanoscale. Its advantages include minimal sample preparation, small testing volumes, high spatial resolution, and relatively non-destructive operation. Traditionally, nanoindentation has been used to measure local mechanical properties such as hardness and elastic modulus. However, the deformation behavior of many materials is inherently time dependent.

This work is part of a comprehensive nanoindentation study of α-phase plutonium alloys conducted using a newly customized, glovebox-integrated FemtoTools FT-I04 nanoindenter. The enhanced system stability and precise displacement control enable systematic strain-rate-dependent indentation experiments, including constant strain-rate, strain-rate jump, and load-hold protocols. Such capabilities are particularly important for actinide materials, which exhibit complex thermally activated deformation behavior arising from strong electron correlations, multiple allotropes, and defect-sensitive plasticity.

The strain-rate dependence of deformation in α-phase plutonium alloys was evaluated at constant strain rates of 0.0001 s^{-1}, 0.001 s^{-1}, 0.01 s^{-1} and 0.1 s^{-1}, as well as through strain-rate jump experiments. These measurements provide insight into strain-rate sensitivity, activation volume, and activation energy. We further examine the relationship between indentation loading profiles and the corresponding measured hardness values. Such analysis aids in identifying the governing deformation mechanisms, defect interactions, and potential diffusion-assisted processes.

Overall, this work demonstrates the utility of advanced nanoindentation techniques within a glovebox environment and contributes to the development of a comprehensive micromechanical characterization framework for actinide materials.

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Deployable Modular Electrochemical Decontamination for Nuclear D&D Applications

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Decontamination and decommissioning (D&D) of radiological equipment is frequently constrained by labor-intensive chemical methods that increase operator time-in-containment, drive dose and ergonomic risk, and generate significant secondary waste. These constraints are especially acute for complex geometries such as glovebox interiors, small high-surface-area parts, and long tubing or pipes, where wiping and immersion approaches are slow, inconsistent, and difficult to scale. To address these challenges, Los Alamos National Laboratory developed the Modular Electrochemical Nuclear Decontamination System (MENDS), a scalable, semi-autonomous platform that couples high-strength oxidizing chemistry with closed-loop electrochemical regeneration to deliver rapid decontamination while minimizing liquid waste inventory and reducing hands-on work.

MENDS adapts the cerium redox chemistry used in the Catalyzed Electrochemical Plutonium Oxide Dissolution (CEPOD) process into a closed-loop architecture that continuously regenerates Ce(IV) oxidant from Ce(III) within a flow-through electrolyzer. Continuous oxidant regeneration enables sustained decontamination using a finite electrolyte volume, reducing make-up chemical demand and confining liquid waste to a managed loop. In this approach, oxidizing solution is circulated through an application-specific cleaning interface (e.g., a sealed cleaning head or spray nozzle), returned to the electrochemical cell for re-oxidation, and reused until decontamination endpoints are achieved. This architecture is intended to replace repetitive manual scrubbing with monitored circulation supported by measurable oxidant strength.

Commercial off-the-shelf (COTS) electrolyzers were evaluated but did not meet facility-driven constraints on size, oxidation efficiency, electrical demand, and material compatability. Consequently, a LANL-designed electrochemical flow cell fabricated via additive manufacturing was developed to maximize active electrode area while controlling hydrodynamics and reducing power draw. Performance testing demonstrated substantial reductions in system mass and footprint relative to comparable commercial units (approximately 6% of the weight) while providing markedly higher electrochemical turnover (approximately a 7× improvement), achieved at substantially lower operating voltage and current. A larger-capacity cell variant has also been fabricated to increase throughput via expanded electrode area, supporting future scaling to higher-flow applications.



Size comparison of COTS electrolyzer versus LANL-developed electrochemical flow cell. Two versions of bespoke flow cell are shown with the short cell in front, and the tall cell in back.

A common circulation/oxidation backbone supports multiple modular implementations for different D&D tasks. In the glovebox configuration (GB-Decon), the system uses a closed-loop reservoir and compatible pumps with temperature control to maintain kinetics and stability. An inline UV–Vis flow cell tracks Ce(III)/Ce(IV) conversion in real time, enabling verification of oxidant strength during operation. For glovebox surface decontamination, GB-Decon employs interchangeable cleaning heads manufactured to conform to flats, corners, and obstructed

geometries. Each head incorporates a compressible gasket that forms a sealed, narrow void against the target surface; negative pressure assists adhesion and draws oxidizing solution uniformly across a defined treatment zone, while quick-connect fittings enable rapid head changes. Preliminary results demonstrate substrate-dependent decontamination to low-level waste (LLW) thresholds on operationally relevant timescales, with 304 stainless steel reaching LLW criteria in approximately 20 minutes and 316 stainless steel in approximately 30 minutes under the tested conditions. By replacing iterative spray-and-wipe cycles with recirculating treatment, GB-Decon is positioned to reduce operator time-in-glovebox, lower dose and ergonomic strain, and improve campaign efficiency.

MENDS also targets long tubing and small-parts decontamination—persistent drivers of D&D cost and complexity. The tube module seals end caps to the tube outer diameter and uses an internal spray to wet interior walls, returning solution for re-oxidation and reuse, thereby enabling decontamination prior to size reduction or disposal. For small, textured, or high-surface-area parts, the enclosed MENDS Recirculator provides an alternative to traditional nitric/hydrofluoric acid soak-and-wipe protocols. In validation tests on Haynes 25 coupons contaminated to high activity levels (up to approximately 10 million dpm/cm², with a release criterion of <100 dpm/cm²), a legacy mixed-acid bath required greater than 12 hours to reach endpoint conditions, whereas the MENDS Recirculator achieved the same endpoint in 19 minutes followed by a brief deionized-water rinse, demonstrating substantial cycle-time reduction without reliance on extended mixed-acid exposure. In addition, the MENDS Recirculator allows for batch decontamination of multiple components in a single pass, all without direct handling or manipulation by operators.

Because MENDS regenerates oxidant electrochemically rather than through continual chemical replenishment, it operates with a fixed solution volume and correspondingly limited secondary liquid waste. Semi-autonomous circulation reduces manual handling of contaminated components and surfaces, while onboard UV–Vis flow monitoring provides a direct measure of oxidant availability and process condition. Across glovebox, tube/pipe, and small-parts demonstrations, MENDS integrates compact, modular hardware with closed-loop chemistry regeneration to enable faster processing, reduced secondary waste, and improved operational safety. Ongoing work focuses on throughput scaling, cleaning-interface optimization, and development of qualification metrics and deployment workflows to transition MENDS into routine production D&D operations.

Machine-Learning Assisted Plutonium Quantification Using High-Purity Germanium Gamma Detectors

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Accurate quantification of plutonium concentration is essential to nuclear safeguards, materials accountability, and environmental stewardship and is governed by federal regulations and institutional directives requiring defensible analytical methods. Current analytical methods for quantifying plutonium require a combination of isotopic determination using High-Purity Germanium (HPGe) gamma spectroscopy coupled with an additional method to determine sample activity. Complementary approaches such as calorimetry and neutron counting are effective but time consuming and logistically intensive within the operating bounds of the facility. We propose a rapid, single-measurement method that integrates HPGe spectroscopy in a fixed, well-characterized geometry coupled with machine learning based regression to provide absolute quantification in a quick and singular measurement.

A major factor that contributes to the analytical uncertainty and computational load is Compton continuum, the removal of which requires either simple linear removal or higher-order approaches such as Whitaker filtering. These approaches introduce additional uncertainty and processing overhead. By establishing a geometry that allows the sample to be considered a point source, the effects of Compton background should be minimized, simplifying these corrections and improving peak isolation.

The use of fixed geometry also enables the development of robust calibration curves based on known, non-interfering gamma lines with well-characterized emission probabilities. Limiting the machine-learning input space to explicitly defined peak features improves stability and preserves physical interpretability of the model. Because the method operates on selectively specified peaks, it can be extended to additional isotopes without modification to the measurement configuration, allowing for greater flexibility.

This framework is intended to reduce assay time and operational burden while maintaining analytical rigor for plutonium quantification, within the established safety systems and envelope of the facility.

Invar Effect in δ-Pu Alloys

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Several long-debated dynamical and elastic anomalies of δ-Pu and its alloys (Pu-Al/Ga/In) [1,2] have been captured within an unprecedented unified theoretical framework. This achievement [3] is made possible by machine learning-accelerated *ab initio* simulations [4,5], which enable an accurate description of explicit temperature effects [6].

For pure δ-Pu, the present simulations—within this unified framework—successfully reproduce the experimental observations of negative thermal expansion, equilibrium volume, and anomalous softening of elastic properties. Our analysis demonstrates that the distinctive behavior of δ-Pu is almost entirely attributable to large anharmonic effects, which are linked to the extreme softening of the phonon spectrum [3].

This simulation approach has recently been extended to Pu-Al/Ga/In alloys. The results reveal significant anharmonic effects upon the addition of δ-stabilizing elements and reproduce the well-known invar effect in δ-Pu alloys, where thermal expansion transitions from negative to positive as a function of Al/Ga/In content. These results are a step forward toward a better understanding of alloying effects in plutonium.

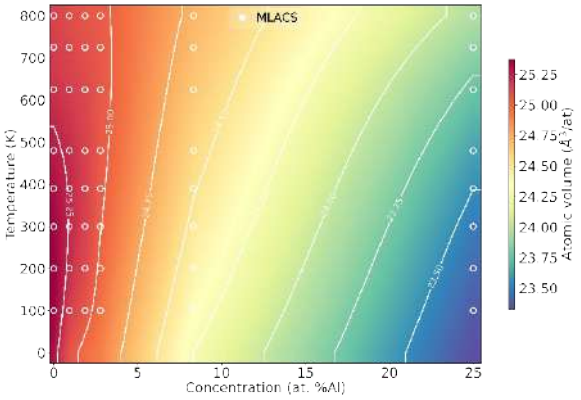


Figure 1: Atomic volume as a function of Al concentration and temperature.

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Investigations Into the Spectroscopic Changes of Actinyl Based on Hydroxide and Nitrate Ligation

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As nuclear energy mitigates the increase in global energy demands, processing spent nuclear fuel (SNF) efficiently is increasingly more crucial.[1,2] Especially relevant when considering waste from nuclear activities such as that stored at U.S. DOE sites, actinide speciation is important and guides the selectivity of the extraction process.[3] While on-line and real-time monitoring techniques such as Raman spectroscopy are being prepared for implementation in tank waste processing, complementary techniques such as luminescence spectroscopy for actinyl species, which are important in the nuclear fuel cycle, could add supporting evidence with additional spectroscopic signatures that harness their optical properties.[4,5] Herein, the goal of this work is to investigate the changes in luminescence of actinyls based on changes in ligand coordination, nitrate to hydroxide, in the solid-state. By measuring the emission of both ligand to metal and f-f transitions of these species, advancement to fundamental photophysical knowledge as well as application in monitoring and nuclear forensics through completed benchmark studies would be added.[5] The far-reaching, scientific motivation of these luminescence studies is to understand actinyl speciation in alkaline conditions as reprocessing SNF in alkaline media could improve the possibility of its reuse in nuclear fuel production.[3,6-8] With a rise in pH and increase in hydroxide concentration in tanks, such as those at the U.S. DOE Hanford Site, understanding both actinyl nitrate to hydroxide species is important.[7] Using single-crystal X-ray diffraction (SCXRD) and measured emission via flourimeter, metal complex structures can be directly compared to their emission spectra.

For the starting material and initial target, plutonyl (VI) dinitrate was first synthesized and crystalized (Figure 1). From Using this plutonium starting material, a bridging hydroxide and tetrahydroxide plutonyl species can follow (Figure 2).[9] Emission of all these species can then be compared.

The following long-term goal will be to set-up a similar design with a liquid sample holder where we can compare solid-state emission to solution emission and support a hypothesis that the species exists in solution. To support a hexavalent oxidation state in solution at a high pH, absorbance of plutonyl (VI) nitrate in 2 M nitric acid, 1 M tetrapropylammonium hydroxide and 1 M tetrapropylammonium hydroxide (TPAOH) is shown after 7 days (Figure 3).[10] For complimentary speciation determination, XANES and EXAFs experiments were performed.

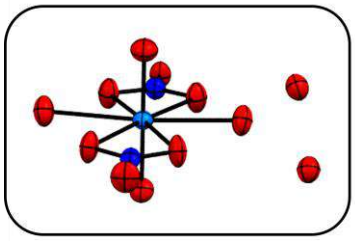


Figure 1: SCXRD of plutonyl (VI) nitrate

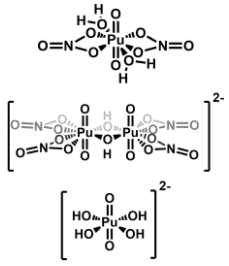


Figure 2: Target Pu (VI) nitrate and hydroxide species

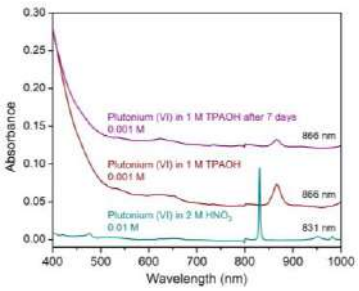


Figure 3: Absorbance spectra of Pu (VI)

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Growth kinetics of plutonium hydride corrosion sites

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Plutonium hydride formed below 100 °C is a finely divided, face-centred cubic material that can spontaneously ignite when exposed to air at room temperature [1]. During long-term storage of plutonium metal in sealed containers, hydrogen can accumulate due to radiolysis of packaging materials and reactions between the metal and moisture released from those materials. As a result, the formation of plutonium hydride becomes a credible possibility during extended storage [2]. This raises concerns about the long-term integrity of vessels containing plutonium, as corrosion and associated products may compromise containment [2]. To ensure safe and reliable storage over long periods, it is important to be able to predict storage behaviour [3], which in turn requires an understanding of the variability in hydride growth kinetics.

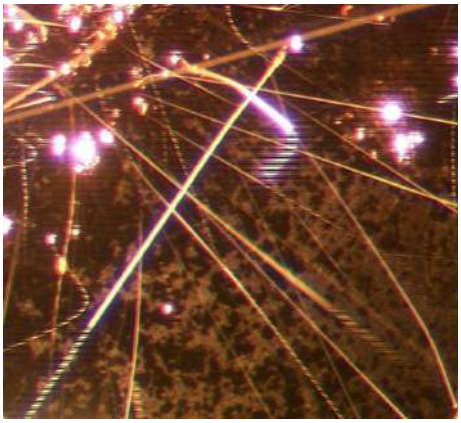


Figure 1 – Photograph capturing the ignition of some plutonium hydride on the surface of a partially corroded sample of a plutonium alloy.

The kinetics of plutonium hydride formation have been reported numerous times, yet the results across studies can be inconsistent [4]. A key challenge in obtaining reliable measurements for this highly exothermic reaction is controlling temperature. The reaction between plutonium and hydrogen proceeds rapidly, and even a small sample of less than 10 g can experience a temperature rise exceeding 50 °C if the heat is not effectively dissipated. Although methods such as using a heat sink [5] can reduce this temperature rise, uncertainty in the actual reacting surface area can still lead to misleading results when using conventional gravimetric or barometric techniques. In contrast, hot-stage microscopy provides a more robust approach [6], offering not only improved temperature control but also the ability to directly observe the nucleation and growth behaviour of individual corrosion sites.

Previous research reported sub-ambient reaction kinetics as part of an ongoing work to understand storage corrosion [7]. In this study, the growth rate of corrosion sites has been measured between 0.01 and 7 bar. The rate of reaction varies with a square root pressure dependence as seen with sub ambient kinetics. However, considering the individual rates measured from different corrosion features allows variability to be considered. Where previous reports have indicated a singular kinetic value for any given condition [4] this study highlights the variability in the growth kinetics of each corrosion site.

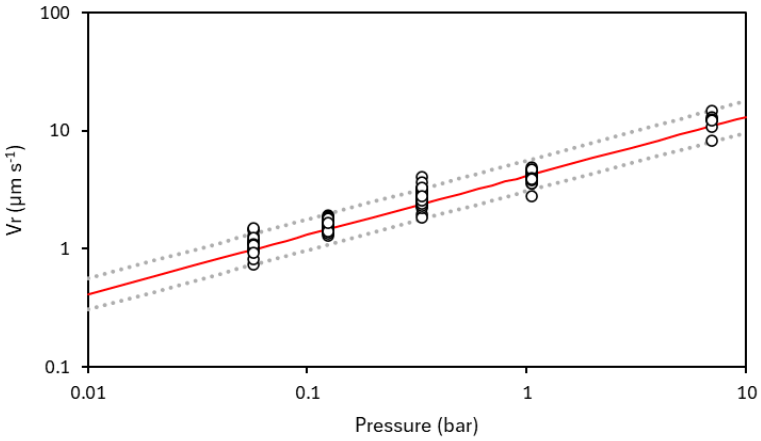


Figure 2 – The pressure dependence of the radial growth velocity (V_r) of plutonium hydride corrosion sites, a best fit is shown (red) alongside the upper and lower 95 % prediction limits (dotted).

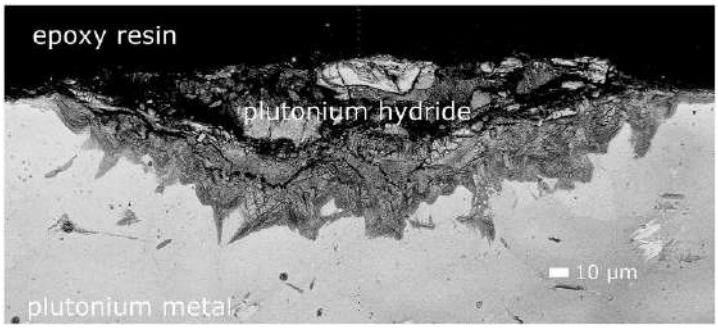


Figure 3 - Cross section of a typical plutonium hydride reaction site imaged using backscattered electrons, noting the discontinuous interface, reproduced from [8].

This study demonstrates that plutonium hydride formation during long-term storage is influenced not only by environmental conditions such as temperature and hydrogen pressure, but also by the intrinsic variability of individual corrosion sites. While the overall reaction rate follows the expected square-root pressure dependence, direct observation using hot-stage microscopy reveals substantial differences in growth behaviour from site to site. These variations arise from microstructural features and discontinuities at the metal-hydride interface, underscoring the limitations of reporting a single kinetic value for complex, heterogeneous systems. Recognising and accounting for this variability is essential for improving predictive models of plutonium storage behaviour and ensuring the continued safety and integrity of sealed containment over long periods.

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The UK's plutonium package surveillance programme: Long-term storage and aging of PuO_2 powder up to 50 years old

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The UK has accumulated a large inventory of separated plutonium from spent fuel reprocessing operations that is largely stored as plutonium dioxide (PuO_2). The PuO_2 has been in storage for up to 50 years already and continued storage will be required until an immobilisation technology is implemented. Consequently, some materials may require storage for over 100 years.

During storage, the PuO_2 powder and the storage containers are subject to conditions of elevated temperature and intense radiation. To understand the effects of long-term storage, Sellafield Ltd undertakes a package surveillance programme that, in addition to inspections within the storage facilities, includes a programme of destructive examination of PuO_2 packages. Characterisation is performed in the High Alpha Inventory Laboratories within the United Kingdom National Nuclear Laboratory's (UKNNL) Central Laboratory.

In the UK, PuO_2 powder was mainly produced by either the reprocessing of Magnox metallic fuel in the Magnox reprocessing plant or from reprocessing oxide fuel in the THORP reprocessing plant. Magnox PuO_2 is packaged in a screw-top aluminium inner can that holds the PuO_2, a layer of polythene that is used to contain the inner can when it is removed from the glovebox and a welded stainless-steel outer can. The THORP package is an all-stainless-steel package design with a screw-top stainless-steel inner can, a breathable intermediate stainless steel can fitted with a sintered metal filter and a welded stainless steel outer can. Both package types can be opened for destructive examination.

Packages are punctured, opened, and sampled before conditioning PuO_2 by heating in a furnace and repacking it for continued storage. Characterisation of the PuO_2 includes Specific Surface Area (SSA) analysis, ThermoGravimetric Analysis (TGA), pore-size analysis, powder X-ray Diffraction (XRD), water adsorption, chemical and isotopic analysis. In addition, the gas composition within each package is determined, the polythene is examined and metallographic examination of the cans is performed.

The UK population of PuO_2 powders provides a unique opportunity to study the effects of aging of many decades because there are PuO_2 powders with a range of initial physical properties that were characterised at the time of production. The evolution of the gas composition in storage packages and changes in the physical properties of PuO_2 occur over several years and decades. These changes may influence disposal as well as behaviour during continued long-term storage. The results of these studies will be summarised and the implications discussed.

The modelling of the dissolution of actinide compounds – the use of ab initio methods to calculate thermodynamic data from first principles

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Understanding the dissolution and aqueous speciation of actinide compounds with a range of different acid media is important for decommissioning activities. A chemical thermodynamic model (Pitzer) was developed to describe the dissolution of actinide compounds in concentrated nitric/hydrofluoric acid (HNO_3/HF) solutions at high temperatures (<120°C).

A critical review of the literature identified several gaps relating to the description of species e.g. actinide fluoride and nitride species were poorly described missing Log K and/or enthalpy values Where data existed, the literature review identified inconsistencies in the reported thermodynamic data e.g. for the fluorides.

To fill the data gap on speciation, Density Functional Theory (DFT) calculations were performed. DFT outcomes were 2-fold - 1) to validate literature data, 2) to generate missing data. Implicit solvation models were generated to calculate Gibbs energy of formation for aqueous actinide fluoride species. Gibbs energy was used to calculate Log K and enthalpy values using the van't Hoff equation. The DFT results showed a strong correlation with experimental Log K's but overpredicted the stability of the formed complex, agreeing with similar published work. Using the developed DFT solvation models and predicted correlations, literature Log K values were validated for the fluorides and enthalpy calculated from first principles.

Developments in Plutonium Recovery at AWE

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Aqueous plutonium recovery is a critical component of the plutonium lifecycle, enabling the UK to maintain responsible stewardship of its plutonium inventory by returning recoverable material to the product stream. Over time, a diverse range of plutonium-bearing residues accumulates from machining, pyro-processing, and analytical operations. Identifying efficient, robust, and optimised methods for recovering plutonium from these residues is therefore essential for closing the material management loop.

AWE and NNL are currently undertaking a coordinated programme of experimental work to generate the data required to inform the design of a future plutonium recovery facility. Ongoing studies include determining optimum dissolution conditions for chloride and non-chloride residues in nitric and hydrochloric acid, evaluating salt-washing strategies, and assessing ion-exchange resin performance to establish achievable decontamination factors. Additional research focuses on precipitation processes—specifically plutonium oxalate to understand product yield and decontamination behaviour. Work is also progressing on final waste encapsulation and long-term storage approaches.

This presentation will outline the latest findings from these activities and highlight future research and development priorities in aqueous plutonium recovery at AWE.

Investigation of Plutonium Metal for Materials Compatibility Testing

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Ensuring material compatibility with plutonium is essential to minimize chemical and mechanical instability, as these interactions can affect system performance, plutonium purity, and safety across a range of processing environments. The authors present an investigation into material compatibility testing of molten plutonium metal at elevated temperatures (500°C). Molten plutonium compatibility with other materials, such as Inconel 718, graphite, tantalum, and tantalum carbide, was investigated with exposure tests and characterization to evaluate any metallurgical interactions. Other metals that have similar metallurgical properties to molten plutonium, such as cerium and gallium, are also tested on the same materials using the same procedure. These additional results are presented and discussed to enable more accessible and comprehensive scientific research where plutonium experiments cannot be performed. The samples are characterized with SEM/EDS to examine the plutonium and sample interface. These results are used to characterize the corrosion and materials compatibility. These plutonium exposure results are compared to molten cerium and gallium exposure results to evaluate if they are suitable surrogates for molten plutonium.

To address the failure of the Inconel 718 when exposed to molten plutonium metal a tantalum coating was applied onto the Inconel 718 and reevaluated with molten plutonium. This follow up was used to identify if tantalum deployed via molten salt electrodeposition would protect the Inconel from the Pu. Molten salt electrodeposition is a premier coating technique that can apply tantalum metal onto a substrate. A molten fluoride salt system with K_2TaF_7 as the precursor was oriented as an electrochemical cell at 700-900°C to apply the tantalum coating. The exposed samples are characterized with SEM/EDS to examine the survivability of the tantalum coating when exposed to molten plutonium metal. These results are compared with the exposure tests of molten plutonium on pure tantalum.

LA-UR-26-21851

A Density Functional Theory Study on the Point Defects in PuO_2

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Vibrational spectroscopy, particularly Raman, is an important technique when applied to plutonium dioxide (PuO_2), as it can provide valuable insights into a given sample ageing history as radiation defects develop over time. To underpin this spectroscopic feature of PuO_2, we are simulating vibrational spectra of PuO_2 containing defects.

Factor group analysis of the space group of PuO_2 ($Fm\bar{3}m$, face-centred cubic; O_h^5 point group) predicts the observation of one Raman active phonon (T_{2g}), experimentally observed at ~480 cm^{-1}, however alpha decay and formation of defects can cause the T_{2g} band to appear at lower wavenumbers, broaden and decrease in intensity. Radiation damage in PuO_2 generates new peaks in the Raman spectrum, such as those seen in the experimental literature in the 530-660 cm^{-1} region, attributed to the formation of various defects. These include various point defects, anionic Frenkel pairs, cationic Frenkel pairs and alpha helium. The Raman-inactive mode (1LO2) is thought to become Raman active through symmetry lowering and lattice disorder and appears in the region ~580 cm^{-1}, possibly following the formation of Frenkel oxygen defects generated through alpha decay.

Computational modelling of this subject provides a more fundamental understanding of the self-irradiated PuO_2 Raman spectra. This study employs Density Functional Theory including spin orbit interaction to investigate the vibrational fingerprint of pure and defective PuO_2, focusing on the IR and Raman signatures. The defects investigated in this study are various point defects, due to their relevance in the experimental literature.

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Divergent Mechanisms of Hydrogen and Oxygen Consumption on Calcined PuO_2: Implications for Long-Term Storage

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The safe and long-term storage of plutonium dioxide requires reliable prediction of headspace gas composition, yet mechanisms governing post-calcination gas consumption remain poorly understood. Radiolytic decomposition of adsorbed water produces both H_2 and O_2 within sealed containers, and the subsequent consumption of these gases by PuO_2 surfaces determines whether conditions evolve toward flammable or pressurized states. Previous work has established that gas generation rates depend on the excess oxygen parameter ($\delta = O/Pu - 2.0$), specific surface area, local radiation field, and adsorbed water loading, but systematic studies isolating the consumption pathways and their dependence on calcination processing conditions have been lacking.

We present a comprehensive experimental investigation of H_2 and O_2 consumption kinetics across anhydrous PuO_2 samples spanning storage-relevant calcination conditions. The sample matrix includes two source materials (BLO and BLO-C) calcined at 400 °C and 950 °C under both air and argon atmospheres, with specific surface areas ranging from 0.8 to 5.0 m^2/g. Small-scale reactors (SSRs) were charged with known quantities of H_2 or O_2 and monitored over periods of months *via* gas chromatography. This geometry delivers ~403× higher radiation dose per gram than full-scale storage containers due to alpha range limitations, enabling accelerated mechanistic studies while maintaining chemical relevance.

Despite occurring on the same PuO_2 surfaces, hydrogen and oxygen consumption proceed through fundamentally different pathways. Hydrogen consumption follows Langmuir chemisorption kinetics characterized by pressure-dependent equilibrium, first-order rate dependence on H_2 partial pressure, and a strong correlation with specific surface area. Calcination temperature is the dominant control parameter: 950 °C processing produces approximately 7× higher adsorption site density than 600 °C, attributed to oxygen vacancy generation and surface defect formation. Reversible surface activation was observed, with a 4× capacity increase upon sequential refilling experiments. No correlation with specific power was detected within constant-power sample sets, establishing that radiolysis *does not drive hydrogen uptake*. The mechanism is best described as $H_2(g) + \square_surface \rightleftharpoons H_2\text{-}\square_ads$, where □ represents oxygen vacancies and coordinatively unsaturated Pu^{4+} sites.

Oxygen consumption exhibits markedly different behavior: first-order exponential decay kinetics no meaningful surface area dependence, and dramatic sensitivity to calcination atmosphere. A critical mechanistic reversal was identified in which air calcination enhances O_2 consumption at 400 °C (+11%) while argon calcination dominates at 950 °C (+56%, corresponding to a 3.3× difference in half-life). This crossover is consistent with a dual-mechanism model: at low calcination temperatures, surface the dose to the surface provides reactive oxygen sites that consume headspace O_2 through chemical reduction, while at high temperatures, oxygen vacancy defects created during calcination serve as irreversible sinks for molecular oxygen. Bi-exponential fitting reveals distinct fast and slow site populations, supporting the coexistence of these two pathways.

These mechanistic distinctions carry direct implications for storage optimization. Because H_2 consumption is predictable from surface area while O_2 consumption depends on defect chemistry

imparted by processing history, a single calcination protocol can be identified that simultaneously addresses both gases. Air calcination at 950 °C produces material with low specific surface area (minimizing hydrogen uptake capacity and generation from adsorbed water) while suppressing oxygen vacancy concentration (extending the O_2 half-life to ~349 days versus ~105 days for argon-calcined material). This work provides the mechanistic foundation for optimizing calcination protocols and predicting long-term headspace gas evolution in PuO_2 storage configurations.

Speciation of Plutonium in Nitrate Media: Effect of Fluorides

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The CEA DAM operates actinides recycling processes in nitrofluorhydric media. In this context, understanding and controlling the complexes formed by plutonium in these media is of critical importance for optimising existing processes and designing novel and efficient process schemes. This study aims to investigate the coordination chemistry and formation of these complexes (speciation) in nitrate media and in the presence of fluoride ions.

For the Pu(IV) oxidation state and varying nitric acid concentrations from 5 to 12 M, three species are identified via visible-near infrared absorbance spectroscopy: the dinitrato complex $Pu(NO_3)_2^{2+}$ predominates at 5 M while the hexanitrato species $Pu(NO_3)_6^{2-}$ is major at 12 M; and a tetranitrato complex $Pu(NO_3)_{4(aq)}$ coexists between with both the latter [1, 2].

This speciation study deals with the determination of the complexation constants (β) of these complexes. After studying Pu(IV) in nitric acid alone, the influence of other species, such as fluoride ions, is also examined. At a concentration of 12 M nitric acid, the addition of fluoride results in a decrease in the intensity of the characteristic band at 491 nm of the plutonium hexanitrato complex $Pu(NO_3)_6^{2-}$ leading to the appearance of a band at 476 nm (Figure 1). Spectral decomposition reveals a signal that may correspond to a mixed complex $PuF_x(NO_3)_y^{(4-y-x)}$, where x = 1 or 2.

Preliminary results highligth the significant impact of fluoride ions in the coordination chemistry of Pu(IV). The formation of several mixed plutonium/nitrate/fluoride complexes is observed and is currently under investigation and characterisation. Taking into account these species into the chemistry of current or future processes could provide substantial benefits, including the optimization of fluorine usage through controlled, on-demand addition.

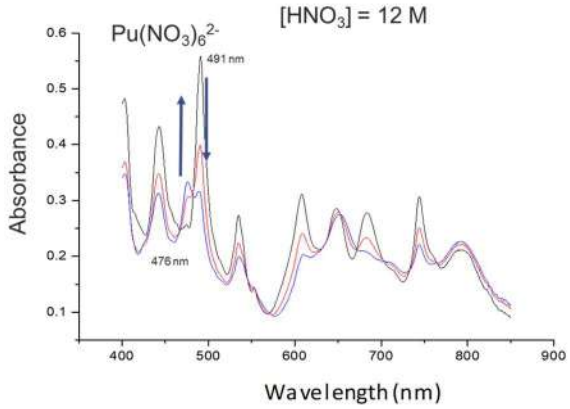


Figure 1. Absorption spectrum of a Pu(IV) hexanitrato complex in 12 M nitric acid (black: [F-] = 0 M) and spectral evolution upon fluoride addition (red: [F-] = 0.03 M; violet: [F-] = 0.06 M).

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Developments in HiZ Physics Package Fabrication

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The High Energy Density (HED) Materials Physics program consists of a series of subcampaigns to investigate the structure and strength of relevant materials at high pressure regimes. Within the program there are a series of targets using Plutonium as the material of interest. The fabrication and assembly process we typically use for the production of lower-Z physics packages require several adjustments to accommodate safety requirements of working with Plutonium.

Improvements in the machining capabilities allow increased precision in thickness of the sample, and improvements in coating robustness allow removal of the sample/pusher glue bond which has allowed the physics drive to maintain better control and probe new regions of the equation of state. We have also successfully implanted helium into the metal matrix, based on an adjusted procedure used for Pb samples previously, which will allow investigation into the effects of aging plutonium in material structure and strength. We will discuss HiZ HED target requirements and how these have been met with current target design, fabrication, assembly and metrology.*

**Representative abstract from closely related work, final abstract pending internal document review*

Prepared by LLNL under Contract DE-AC52-07NA27344. LLNL-POST-867206

Ion Milling and Sputter Deposition for the HEDS of Oxide-free Plutonium

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In Pu research, the creation of an oxide free surface is notoriously difficult to produce; but more so, it is difficult to maintain. The importance of oxide-free surfaces increases as the ratio of potential oxide layer thickness relative to the bulk grows, with oxide layers typically stabilizing at hundreds of nanometers thick in dry air, and increasing exponentially with relative humidity. In High Energy Density Science (HEDS) research with Pu, where analytical samples well below 10um are commonplace, an oxide layer of any measurable thickness can have significant downstream effects on data accuracy and reliability. To this end, development efforts utilizing Ar ion milling and DC magnetron sputter deposition have now allowed for the production Pu samples with minimal oxide layers. Additionally, this work has allowed these samples to be transported, assembled on the benchtop, and processed for data, in air and outside traditional containment barriers (e.g. gloveboxes), all while maintaining that minimal oxide layer.

The research to be presented will cover the development, implementation, and practical results of the ion milling of Pu surfaces, as well as the DC magnetron sputtering of both Al and Cu onto Pu surfaces. With a focus on the characterization of the interface between the Pu and the coating element, the current and future analysis results will include FIB/SEM cross-sectioning (Figure 1 below) and EDS. Additionally, X-ray diffraction examining lattice mismatch, white light interferometry for surface roughness, and lifetime studies of samples in air will be discussed as permitted.

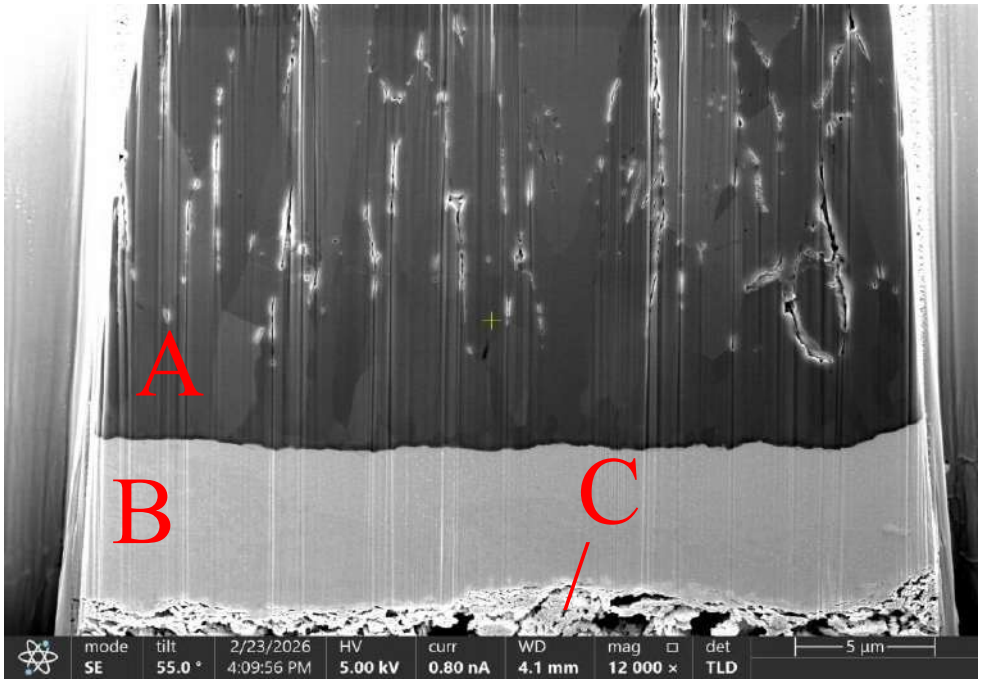


Figure 1. Vertical cross-section SEM image of an Al-coated Pu sample, showing A) the dark Al coating, B) the light Pu substrate, and C) the oxidized Pu encroaching from the unprotected face below

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Hot Isostatic Pressing for Plutonium Immobilisation

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The UK has approximately 140 tonnes of civil separated plutonium dioxide (PuO2) powder following completion of reprocessing operations. The management strategy for this inventory has recently been reviewed and, to reduce the long-term safety and security burden associated with it, UK Government policy has changed to immobilisation as the preferred way forward to put the material beyond reach. UKNNL has been supporting this mission by developing one of the credible options for Pu immobilisation, Hot Isostatic Pressing (HIP) technology, over more than two decades from inactive development to plutonium active sample fabrication and demonstration plant installation. The advantages of HIP and zirconolite are that together they produce a very robust ceramic material that is specifically designed for immobilising radioactive materials, with high loadings and very low leach rates. It is, however, less technologically mature than alternative options and requires development and demonstration. In 2020, a small-scale glove box capability was installed in Central Laboratory to allow plutonium zirconolite pellets to be fabricated and analysed. The subsequent samples produced have validated the Pu-zirconolite system with UK PuO2 and successfully shown the system's chemical flexibility with respect to incorporation of neutron absorbers and variations of PuO2 feed characteristics, such as age and chloride content. The current focus of work is commissioning the 'HIP100' facility; a laboratory-scale alpha-active HIP facility within UKNNL Central Laboratory. When complete in 2027 it will be the UK's first HIP capability able to manufacture PuO2 containing products.

He bubble nucleation and growth rates in naturally aged δ-Plutonium

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The self-irradiation through α-decay in Pu-Ga alloys remains an outstanding issue as the development of He bubbles through this decay negatively affects the mechanical properties of these alloys. Both experimental and modeling studies have quantified changes in Pu-Ga mechanical properties, but they require knowledge of He bubble development with respect to time. Previous studies show that He atoms are generated at an approximate rate of 41 at ppm/yr[1], and He bubble number densities from 0.6-2.0x10^{17} cm^{-3} are associated with this generation rate [2]. However, this rate has not yet been used to predict He bubble number densities in samples older than 42 years[2].

Recent studies using scanning transmission electron microscopy have provided new He bubble sizes and distributions in three naturally aged Pu-Ga alloy samples[3,4]. Here we apply kinetic models, including crystal growth rate[5] and crystal size distribution (CSD)[6] equations to assess He bubble development as a function of time. CSD calculations for He bubble size distributions are found to be within the range of previously published He bubble number densities[2], suggesting the viability of applying CSD equations to He bubbles. Based on over 1,100 He bubble measurements from the three aged samples, we calculate nucleation and growth rates to provide realistic forecasting on He bubble growth rates, as well as bubble size and abundance in older Pu-Ga alloys.

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LA-UR-26-21834

Preparation of Plutonium Arene Complexes Through Oxidative Dissolution of Plutonium Metal

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Oxidation and dissolution are important parts of reprocessing and purifying plutonium metal. Chlorination of plutonium metal using chlorine gas is a particular challenge due to its toxicity and handling challenges. Our group has been interested in advancing milder chlorinating reagents, such as $GaCl_3$, which can chlorinate *f*-element metals under a variety of conditions. Previous work by our group has established ethereal solvents as a possible media to effect this $GaCl_3$ mediated oxidation of plutonium metal. Here, we report that aromatic solvents are also competent solvents to effect this oxidation. Here, the $GaCl_3$ reagent acts as an oxidant, a chloride source, and as a capping ligand to form the final product. Bond metrics and spectroscopic data will be presented to elucidate periodic trends across the lanthanide and actinide series for these complexes. Notably, this is one of the few isostructural and isomorphous series of complexes that span the lanthanide and actinide series. Further reactivity studies will be presented to establish a more organometallic understanding of the complex.

Developing Dynamical Mean-Field Methods for Actinide Materials Simulation at AWE

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Electronic structure simulations for actinide materials are complicated by the propensity of the 5f electrons to form strongly correlated ground states. To model these states accurately, a combination of efficient density functional theory (DFT) and many-body methods, chiefly Dynamical Mean-Field Theory (DMFT) is required. Several questions of industrial importance require models to describe the influence of local perturbations on the electronic structure. Examples include the surface chemistry of a solid, or predicting structural relaxations due to a lattice defect or dislocation. To simulate these effects in conjunction with strong correlations requires scalable and accurate DMFT implementations that can work with large unit cells, even when neglecting non-local correlations. Ongoing work to develop such a capability at AWE is presented.

A Density Functional Theory Study on Induced Covalent Short Bond Maximization on U in Pu-Ga Alloys and Extrapolation to the ζ-Pu-U Phase

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Understanding the effects of the radioactive decay of 239-Pu due to its daughter products, U and He, on the phase stability of δ-Pu has been a long outstanding problem. While many Pu aging studies have focused on He ingrowth and its effects of void swelling, minimal studies have focused on U, even though significant amount of lattice damage will occur from the U recoil energy. It is well-known that U is a δ-phase destabilizer and thus using density functional theory, a systematic study of U defect calculations has revealed the fundamental role of U impurities in affecting the overall electronic structure of δ-Pu-Ga alloys and the complexity of Pu-U alloys in general (Figure 1). The calculations imply that U induces short bonds that enhance covalency in the δ-Pu lattice which competes with the Ga stabilization mechanism [1]. In addition, the migration of U within the δ-Pu-Ga alloys induces local distortions thus migrating as a multi-complex defect regardless of diffusion mechanism. U-vacancy migration yielded an activation energy of 1.27eV, where U-substitutional with neighboring Pu interstitial to a U octahedral interstitial site was slightly lower at 1.25eV. Therefore, the DFT results imply that the U atom can diffuse via either mechanism, although the formation energy to form the U-substitutional with neighboring Pu interstitial is 1.08eV lower in energy than a U-substitutional with first nearest neighboring Pu vacancy with formation energy of 0.04eV and 1.12eV, respectively. Interesting, the energy difference is the same energy required to form a Pu monoclinic monovacancy defect [2], where the U-vacancy defect resembled the same local distortions (Figure 2).

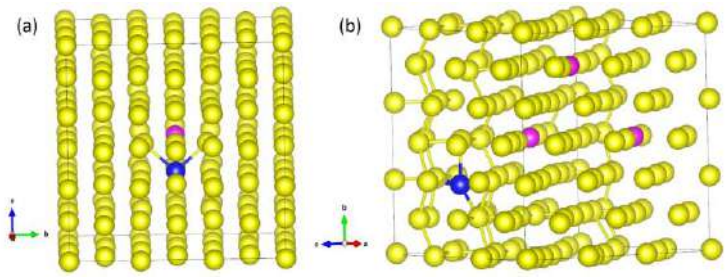


Figure 1: Ionic relaxed structures of (a) 0.926 at.% Ga of the U atom at first nearest neighbor distance from Ga and of (b) 2.778 at. % Ga of U octahedral at third nearest neighbor to the Ga atom. Pu atoms are yellow, U atoms are blue, and Ga atoms are purple.

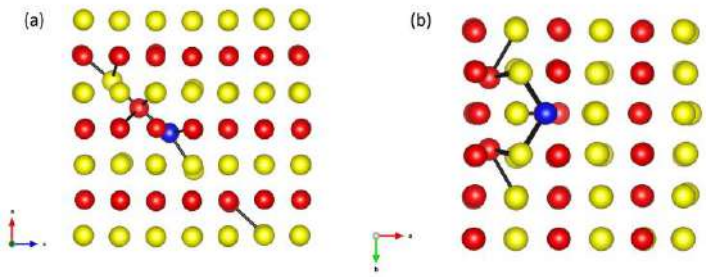


Figure 2: Migration of (a) interstitially mediated and (b) vacancy mediated U migration. The Pu anti-ferromagnetic arrangement is shown by spin-up (yellow) and spin-down (red), and the U atom is blue. Short Pu-Pu bonds <3.0Å are denoted by black.

We use these conclusions to extrapolate our findings to the ζ-Pu-U phase as defined by Lawson et. al [3] to distinguish the atomic lattice sites of the U and Pu atoms within this structure (Figure 3). Supporting evidence comes from random alloy modeling suggesting a significant U-U bonding interaction energy relative to weaker Pu-Pu interaction energy and a nearly nonexistent U-Pu interaction across the ζ-Pu-U composition range, but no evidence for short range order. Spin-polarized DFT calculations for the extrapolated ζ-phase were performed where the Pu atoms were treated in an anti-ferromagnetic arrangement, which was based on two criteria: (1) The first criteria that needed to be satisfy was that the spin-up and spin-down are determined when the Pu-Pu bond was >2.8Å, followed by the second criteria of the Pu-Pu bonds at 2.8Å < x < 2.95Å. After volume and ionic relaxation, where the cell shape remained constrained, the crystal structure underwent a percent difference of 3.35% in volume (a_0 = 10.566Å versus a_{expt} = 10.685Å) and some bonds where slightly expanded or contracted (Figure 3). Therefore, this extrapolated version of the ζ-phase based on the induced covalent short bonding by a U defect observed in the δ-Pu-Ga alloys appears to be stable according to density functional theory calculations. Heat of formation and the electronic structure will be discussed further.

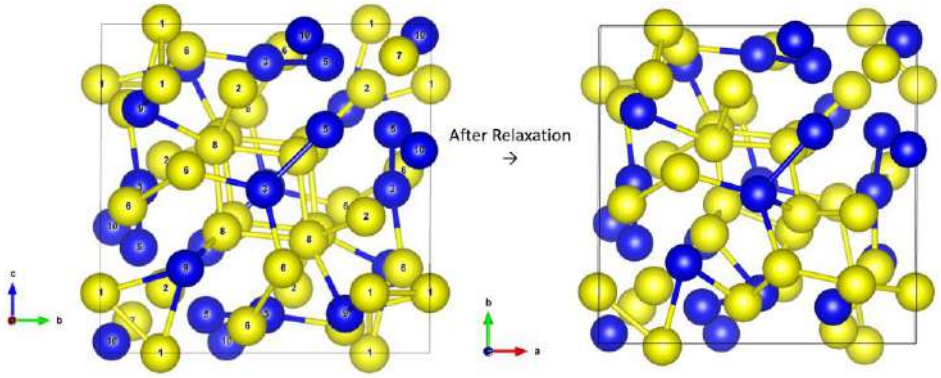


Figure 3: Left image is the hypothesized ζ-Pu-U structure as defined by Lawson et al. [2], where the U atoms are denoted in blue and Pu atoms are denoted in yellow. Right image shows the DFT volume and ionic relaxed structure.

Acknowledgement to the LDRD program at Los Alamos National Laboratory for supporting the research associated with projects #20150057DR: "Aging in Delta Plutonium Alloys: A Fundamental Approach," #20230202DR: "Understanding Pu Corrosion on Machined Surfaces," #20240478CR-GTS: "Advancing Actinide Science, Technology and Engineering for National Mission," and #20260038DR: "Development of a Thermodynamic Framework for Predicting Plutonium Alloy Microstructures." This research used resources provided by the Los Alamos National Laboratory Institutional Computing Program, which is supported by the U.S. Department of Energy National Nuclear Security Administration under Contract No. 89233218CNA000001.

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Porphyrin-Based Plutonium Metal-Organic Frameworks: A Versatile Platform for Investigating Elusive Plutonium Cluster Chemistry

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Plutonium coordination chemistry has evolved rapidly over the last decade, transitioning from fundamental studies of 1D and 2D coordination polymers to the realization of complex 3D metal-organic frameworks (MOFs). Following the landmark synthesis of Pu-UiO-66 [1], interest intensified in leveraging these porous scaffolds to stabilize elusive coordination environments and high-coordination nodes that are typically inaccessible via traditional high-temperature solid-state methods. Because MOFs offer a modular platform for nanoscale control, they are uniquely suited for investigating the distinct relativistic effects and *f*-orbital participation in bonding that characterize the tetravalent actinides.

Porphyrins serve many important roles in biological systems, such as enabling photosynthesis and oxygen transport. They are highly conjugated, aromatic molecules that possess many resonance structures. Porphyrins and porphyrin derivatives are usually intensely colored, with a strong absorption band (B band) around 420 nm and weaker bands (Q bands) around 520-650 nm. Due to their remarkably poor solubility even in nonpolar solvents, porphyrins can be functionalized with benzoic acid groups to create tetrakis(4-carboxyphenyl)porphyrin (TCPP) linkers.

Herein, we report the solvothermal synthesis of a plutonium-based MOF that incorporates the TCPP linker. The synthesis involved a 9:1 molar ratio of plutonium(IV) nitrate to TCPP in dimethylformamide (DMF), with water and trifluoroacetic acid (TFA) added as modulators. The solution was then heated at 120 °C for four days, slowly cooled to room temperature over two days, and stored in a freezer. Characterization of the resulting microcrystalline material via powder X-ray diffraction (pXRD) confirmed it to be isostructural with the zirconium-based PCN-222 framework [2]. Ancillary analysis using physisorption measurements and absorption and vibrational spectroscopy supported this assignment.

While initial crystal dimensions were restricted to the microscale (10-50 μm), we observed a significant morphological and structural transformation over 14 months. This aging process, likely driven by radiolysis, resulted in the growth of significantly larger crystals reaching 250 μm. Analysis of these larger crystals using single-crystal X-ray diffraction, pXRD, and Raman revealed a phase transition from the original porous framework into a high-symmetry plutonium compound that is tentatively assigned as a plutonium oxyhydroxide.

This work, supported by a comprehensive synthetic roadmap developed across the lanthanide series, establishes a new transuranic architecture for MOFs and provides further insight into the impact of self-radiolysis on the stability and transformation of actinide-based porous materials.

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Title

Bulk vs. Nano-Indentation Stress-Relaxation of δ-Phase Plutonium Alloys: Methodology, Parameter Extraction, and Compliance Corrections

-Roger Jaramillo, Zachary Levin, Isaac Hernandez, Todd Martinez, Veronica Anghel

Abstract

The mechanical performance of δ-phase plutonium (Pu) alloys is governed by time-dependent relaxation processes that are difficult to capture across length scales. This work presents a comparison between macroscopic stress-relaxation experiments on bulk specimens and high-resolution nano-indentation relaxation tests on the same alloys. Bulk samples were subjected to compression deformation in a 10 kN screw-driven mechanical test frame and held at constant displacement for 2 to 3 minutes. The resulting stress-relaxation curves were fitted to a logarithmic decay model ($\Delta\sigma(t) = C1 \ln[1+(t/\tau)]$) to extract a characteristic relaxation time (τ) and the material constant (C1). A compliance-correction scheme was applied by measuring the machine stiffness with a reference specimen and subtracting it from the raw load-displacement data, yielding true material compliance.

In comparing to nano-indentation stress relaxation measurements; key differences between the two approaches are highlighted: (i) bulk tests probe homogeneous, uniaxial stress states whereas nano-indentation samples a confined, triaxial field; (ii) parameter extraction in bulk data relies on direct strain control, while nano-indentation demands indirect strain estimation from contact area; (iii) compliance correction in bulk experiments is a non-linear subtraction, whereas nano-indentation demands a geometry-dependent, non-linear correction.

Despite these methodological disparities, the extracted τ and C1 values show reasonable agreement for all investigated δ-phase compositions, demonstrating that nano-indentation can reliably reproduce bulk relaxation behavior when proper compliance corrections are applied. The results provide a robust multiscale framework for assessing time-dependent mechanical properties of Pu alloys and inform future modeling of their long-term performance.

Keywords: δ-phase plutonium, stress relaxation, bulk compression testing, nano-indentation, compliance correction, logarithmic decay model.

Exploring the Influence of Variable Hydrogen Pressure on the Early Stages of the Plutonium–Hydrogen Reaction

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The early stages of the reaction between plutonium and hydrogen were investigated under both constant and variable pressure conditions to better understand factors relevant to the storage of plutonium metal. Interest in the formation of plutonium hydride has increased in recent years due to its relevance for ensuring the safe, storage of plutonium metal [1–8]. Hydrogen can accumulate within sealed storage vessels during extended storage and conditions favourable for hydride formation may develop [9]. Hydride formation raises safety concerns due to the pyrophoric nature of plutonium hydride, which poses a risk of dispersing plutonium beyond appropriate containment [10]. The dependence of initiation time, nucleation rates, and radial growth velocities of plutonium hydride corrosion sites on hydrogen pressure have been established [1]. However, unlike controlled laboratory experiments, the pressure of hydrogen during storage would not remain constant and therefore, exploring the reaction under varied pressure regimes is of interest.

Using an *in situ* hot stage microscopy capability, the influence of hydrogen pressure on the initiation stages of the reaction between hydrogen and a plutonium alloy have been examined (Figure 1). A set of standard constant pressure experiments that measured initiation times, nucleation rates, and radial growth of plutonium hydride corrosion are compared to variable pressure experiments conducted using ramped pressure control or by varying the positioning of the gas inlet.

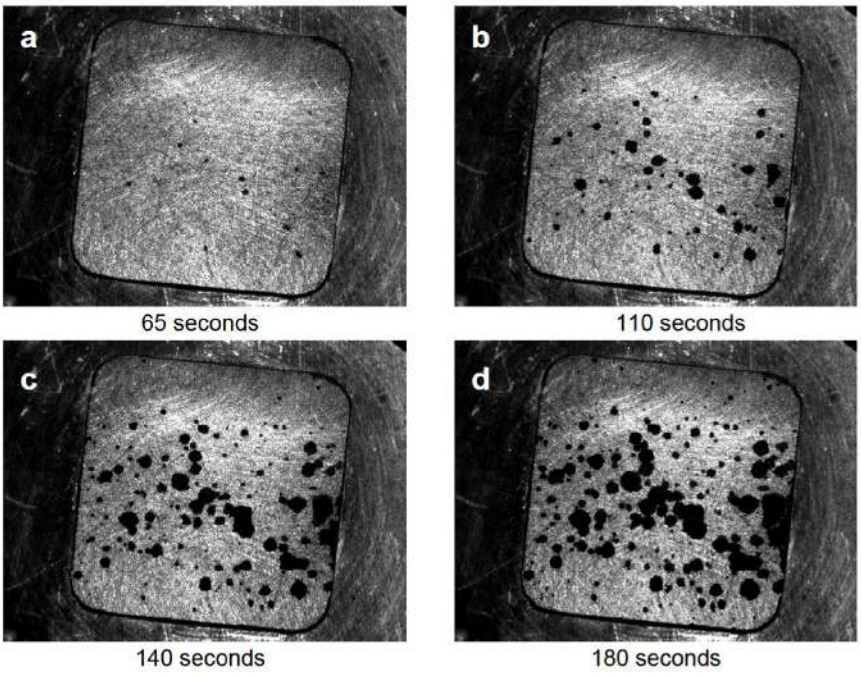


Figure 1 - Time resolved images illustrating the nucleation and growth behaviour of plutonium hydride reaction sites.

Variable hydrogen pressure conditions exert a measurable influence on the initial stages of the plutonium–hydrogen reaction. Although The kinetics observed under constant and ramped pressure conditions differ, they appear to correlate in terms of the hydrogen exposure ($bar \cdot s^{-1}$). The positioning of the gas inlet causes short-lived variations in local pressure which influence kinetics (Figure 2).

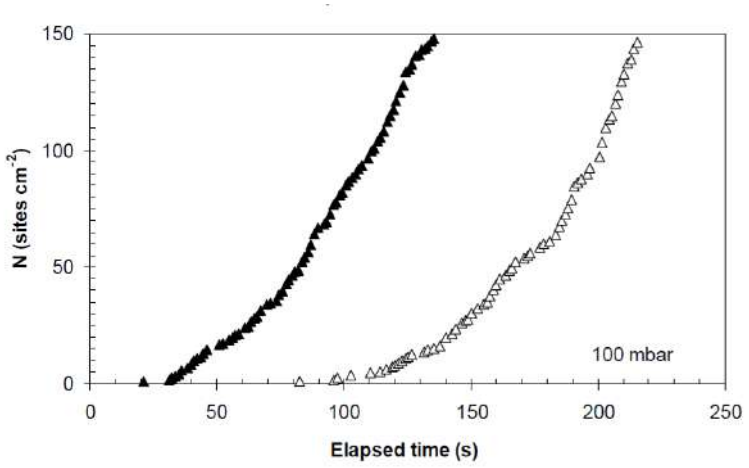


Figure 2 - Nucleation profiles for experiments conducted under 100 mbar of hydrogen. This study made use of two reaction vessels which were identical apart from the location of the hydrogen inlet. When the hydrogen inlet is directed at the sample the nucleation rates are greater.

This study has observed variations in hydrogen pressure, even over short timescales, influencing the initiation behaviour and early kinetics of the plutonium–hydrogen reaction. By comparing constant pressure experiments with those conducted under ramped and spatially varied pressure conditions, it is demonstrated that hydride formation is sensitive not only to the absolute pressure but also to the dynamic nature of hydrogen delivery. These findings highlight the importance of considering realistic, pressure regimes when assessing hydride formation.

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Synthesis of Actinide Fluorides for Advanced Nuclear Fuels and National Security

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Development of new protocols for the synthesis of high-purity, anhydrous actinide fluoride materials is driven by increasing ramp-up of advanced nuclear reactor systems including Molten Salt Reactors (MSRs), which in parallel require the development of safeguards and nonproliferation strategies. Furthermore, these compounds are essential building blocks in fundamental chemistry. Synthetic routes towards isolated actinide fluorides are dominated by the use of hydrofluoric acid (HF) using pressure or flow reactors at high temperatures, with significant infrastructure investments required to mitigate hazards these conditions pose to workers and the environment. We demonstrate the capability to generate anhydrous actinide fluorides at low temperatures, with minimal investment, using easy-to-handle main group fluoride reagents in reaction with actinide chloride coordination compounds. Described in this presentation are new synthetic routes to ThF_4, UF_4, and UF_3 materials bypassing the use of HF, with progress towards the synthesis of analogous Pu materials discussed. Efforts towards utilizing common oxide starting materials in the generation of actinide halides will also be discussed.

5f-in-Core Pseudopotentials and Applications to Electronic Structure of [Pu_{38}] Cluster

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Abstract: Computational modeling of actinide complexes are challenging because of the $5f^n$ open shell. Theoretical calculations of actinide clusters with multiple actinide atoms are expensive and often difficult to converge in self-consistent iterations. To solve this issue, relativistic pseudopotentials (PPs) and basis sets provide an possible solution for theoretical modeling of heavy actinide elements. The norm-conserving Goedecker–Teter–Hutter (GTH) pseudopotentials offer distinct advantages for investigations of actinide molecular and condensed-phase systems, due to their excellent transferability and reliable accuracy. Recently, we have developed a set of benchmarked scalar-relativistic GTH-type 5f-in-core pseudopotentials and corresponding Gaussian basis sets, for the widely studied trivalent and tetravalent actinide ions [An(III), An(IV); An = Pa–Lr].[1] The 5f-in-core formalism adopted in these GTH PPs effectively mitigates the computational challenges associated with the open 5f valence shell in actinide elements. In this work, the structural optimization, analyses of electronic structure, chemical bonding, cluster stability and formation mechanism of $[Pu^{IV}_{38}O_{56}Cl_{54}]^{14-}$ cluster (Figure 1) were performed by using our 5f-in-core PPs and basis sets in the CP2K package. This new PPs can enable large-scale quantum-chemical computations and *ab initio* molecular dynamics simulations of actinide systems in gas phase, solutions, molten salts, bulk solids and surfaces.

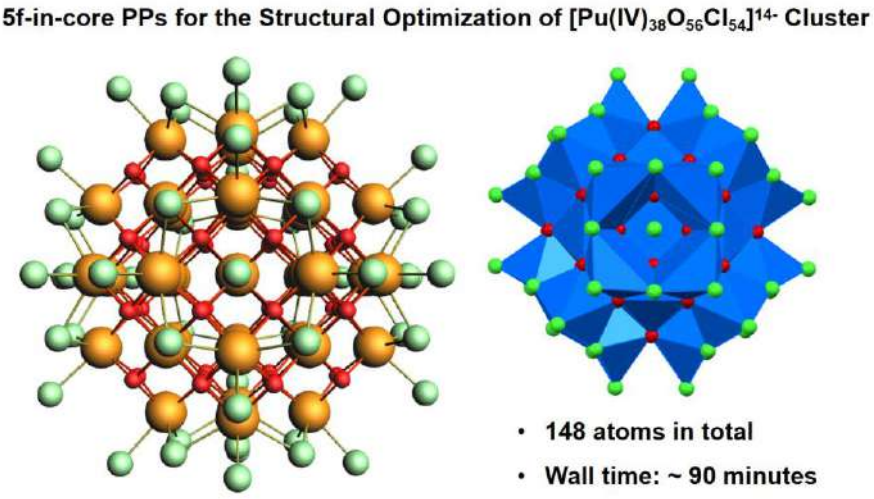


Figure 1. The structure of $[Pu^{IV}_{38}O_{56}Cl_{54}]^{14-}$ cluster optimized by using the 5f-in-core PPs.

Keywords: 5f-in-core, pseudopotential, actinide, electronic structure, Pu_{38} cluster

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Measuring the Density of Plutonium Surface Oxides

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Plutonium is known to have an extremely high oxygen affinity, growing a surface oxide in even low oxygen content atmospheres [1,2,3]. There are some measurements on the densities of mechanically packed oxide powders [4], for example for investigating powder sintering behaviour [5], and also on x-ray diffraction measured lattice parameters to calculate ideal oxide densities. However, there is very little data on *in-situ* grown surface oxides, necessary to fully understand growth kinetics.

Stanford et al [6] created a range of oxide thicknesses on bulk metallic plutonium, ranging from 35 nm to 400 nm as measured from focussed ion beam cross-sections. For each sample they used electron probe micro-analysis (EPMA) to measure the oxygen Kα x-ray intensity relative to pure plutonium oxide reference materials, termed the O k-ratio to generate O k-ratio versus oxide thickness correlation curves.

The authors of this study applied thin film analysis algorithms on Stanford et al's comprehensive published data to calculate the densities of their grown oxides. Normally these algorithms are used to calculate layer thickness and/or composition from EPMA measured element k-ratios, using an assumed layer density. Utilising the known thicknesses and O k-ratios, and assuming stoichiometric PuO_2, the calculations were 'inverted' to output densities. The results from 3 different software packages, PENEPMA, DTSA-II and BadgerFilm, were compared and produced values ranging from 4.0 to 5.0 g.cm^{-3} (Table 1). This falls within the 1.5 – 5.8 g.cm^{-3} range of values published for mechanically compacted oxide powders [4] but is only half that of the 10.2 – 11.5 g.cm^{-3} expected from XRD-measured lattice parameters [7]. The authors discuss the potential causes for this discrepancy.

Table 1 Calculated mean densities and standard deviations.

Software	Mean density (g.cm^{-3})	Standard Deviation (g.cm^{-3})
PENEPMA	4.1	0.5
BadgerFilm	4.7	0.5
DTSA-II	5.0	0.6

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Using Principal Component Analysis on Compositional Measurements of Multi-component Pu-alloys to identify potential minor phases

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Identifying and measuring the composition of minor phases in multi-component Pu-alloy is an important part of understanding the phase equilibria behavior and material properties of these alloys. Using wavelength-dispersive X-ray spectrometry (WDS) on the electron microprobe (EPMA) results in precise quantitative compositional data with a high spatial resolution (~ 1 micron), making it an ideal technique for this application. Analyzing the composition of small (< 5 microns) minor phases in various plutonium alloy compositions is a difficult task however, for reasons related to the generation and detection of characteristic X-rays using a focused electron beam. First, the excitation volume of the characteristic X-rays of interest can exceed the size of the grain, while X-rays from relatively lighter elements can be excited within the grain by heavier, higher energy X-rays coming from outside the grain (fluorescence), both of which lead to spurious signals that are difficult to quantify. There may also be peak interferences in the minor phases that are not present in the bulk material due to differing compositions and are therefore not accounted for. Short counting times (commonly used for broad area map surveys due to practical analysis time considerations) lead to poor counting statistics and detection sensitivity that can make phase identification difficult. Each of these difficulties can be overcome however with a dedicated analytical campaign designed specifically for a minor phase that has a particular composition. But this can be very resource intensive approach, particularly when the identities of the target phases are unknown.

We present here the results of applying principal component analysis on quantitative WDS map data that identifies correlations of the elements across varying bulk alloy compositions. This statistical method leverages the large number of data points in a map to overcome the difficulties listed above, and to point to the potential compositions and identity of minor phases present within the sample. This information can then be used to design a targeted analytical procedure that minimizes instrument time and analyst effort.

Handheld LIBS Analysis in Nuclear Material Processing

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Laser-Induced Breakdown Spectroscopy (LIBS) utilizes short, high-energy laser pulses to ablate material off a sample surface and generate a high-temperature plasma. As the plasma cools down, the excited species within emit a characteristic optical spectrum that is recorded by a spectrometer, and the elemental constituents of the sample under investigation can subsequently be identified. Due to its simplicity and ability for multi-elemental analysis, LIBS has been a very attractive spectroscopic tool for analytical chemistry for a broad range of samples with unknown compositions, that is especially suitable for harsh environments where laboratory setting and/or proper sample preparation are not readily available.

In this work, LIBS has been introduced in a nuclear production facility in Los Alamos National Laboratory (LANL) to perform analytical chemistry assays on actinide containing materials. Portable Handheld (HH-) LIBS analyzers (Z903, SciAps Inc.) allow for high-quality analytical measurements with no sample preparation, even in a radiologically contaminated environment. We demonstrate that the limit of detection of HH-LIBS analyzer can be as low as tens of ppm inferred from Fe impurity in Ce-host matrix (as Pu surrogate) that features highly complex LIBS emissions. This implies that, along with fast outcome (less than 10 minutes), LIBS can not only provide rapid identification of impurities on various stages during nuclear processing but also offer complementary approaches on, for example, go/no-go decisions to filter out the condition that requires further examination via ultrahigh sensitive ICP-analysis. We will also discuss the possibility of developing quantitative analysis.

Atomic-Scale Mapping of Pu Oxidation and Hydrogenation with Environmental Atom Probe Tomography

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Predicting the long-term stability of plutonium (Pu) requires mechanistic insight into how surface oxides and hydrides nucleate, grow, and evolve under storage-relevant environments. To address this knowledge gap, we have begun implementing the advanced Atom Probe Tomography capability of environmental-Atom Probe Tomography (eAPT) [1]. eAPT experiments were conducted on δ-Pu specimens to observe in real-time the interaction between native oxide and deuterium. Within the inherent electric field environment of the eAPT experiment, introduction of pure D_2 into the analysis chamber initiates hydrogenation and oxide-reduction events that were captured directly in reconstructed 3D atom distribution maps. This direct visualization provides quantitative insight into hydrogen uptake rates and reduction kinetics at the atomic scale, revealing strong crystal orientation dependences on hydrogenation behavior. Using PNNL's ball-model reconstruction tool we identified specific surface facets, particularly (111), (001), and (011) orientations that are observed as loci of increased reactivity. The facet-resolved maps also demonstrate grain-to-grain variability in the oxidation and hydride formation, underscoring the importance of microstructural context when considering corrosion susceptibility. Recent eAPT studies on iron [1] and associated quantum mechanical models on ruthenium [2] show that tens of V/nm electric fields (EFs) polarize and attract gas molecules, effectively increasing the local partial pressures at the specimen apex and lowering adsorption energy barriers. In this context, the PuO_x reduction observed under D_2 likely includes an EF contribution, making it necessary to develop field-ramp control and pulse-parameter sweeps to separate EF-driven effects from intrinsic reaction dynamics and enable facet-specific rate extraction. These eAPT results provide the first time-resolved evidence of plutonium oxide reduction and hydride formation by integrating atomic-scale composition mapping while recognizing EF-dependent effects. This framework is leading to an improved interpretation of aging behavior and supports the development of predictive models for long-term stability of plutonium materials.

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Critical Review and Thermodynamic Assessment of the Pu-Ni System

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A thermodynamic assessment of the Pu-Ni system has been performed using the CALPHAD (CALculation of PHAse Diagrams) method, leveraging both experimentally available data for phase equilibria and new density functional theory (DFT) calculations for heat of formation and heat capacity predictions. Heat of formation for the PuNi, $PuNi_2$, $PuNi_3$, $PuNi_4$, $PuNi_5$, and Pu_2Ni_{17} intermetallic compounds have been calculated using three different DFT methods and compared to the final CALPHAD assessment. In this work, we include an extensive review of the experimental data to optimize Gibbs energy parameters for the CALPHAD fit. In addition, this work leverages DFT computed heat capacity for the PuNi intermetallic to better assess the Pu-rich portion of the phase diagram. This work highlights the overall lack of experimental data and uncertainty in determined phase boundaries for this system but provides the first thermodynamic assessment of the Pu-Ni binary system grounded by rigorous DFT calculations.

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Title:	Probing the to ’ phase transformation with X-ray Photoemission Spectroscopy
Author(s):	Ponder, William Stephen Boehm, Alexander Matthew Olive, Daniel Thomas Mazza, Alessandro
Intended for:	Meeting: Plutonium Futures, 2026-09-13 - 2026-09-17 (Edinburgh, UNITED KINGDOM)
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Probing the δ to α’ phase transformation with X-ray Photoemission Spectroscopy

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Low-Gallium δ-stabilized Pu alloys exhibit low phase stability and are subject to reversion to the α’ phase when subjected to thermal cycling or mechanical pressure. The α and α’ phases have the same monoclinic structure but have diminutive difference in lattice parameter and included Gallium. Given the identical crystal structures it is expected that α and α’ phases should have nearly identical electronic structure and should produce similar XPS spectra. The α and δ 4f XPS lineshape have clear differences, owning to changes in local bonding and electronic structure. The α’ to δ phase transformation should be observable via X-ray photoemission spectroscopy (XPS) which can directly probe the transition as a function of temperature near-surface (first few nm). This work presents XPS on δ-Pu metal prepared such that the α’ phase is mechanically induced. The sample was polished and was not annealed to retain polishing induced α’. The sample was measured in-situ while heating to monitor the reversion to the δ phase. Experiments inducing α’ phase by cooling to cryogenic temperatures and subsequent reversion by heating will also be discussed. These experiments shed light on the mechanisms of α’ formation and reversion related to temperature.

Quantifying Manual Lapping for Precision Beryllium Target Components

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Beryllium remains a key target material because of its low atomic number, stiffness, and favorable performance in demanding experimental configurations. Its finishing, however, presents persistent challenges due to brittle damage, edge sensitivity, and stringent dimensional requirements. Although manual lapping is often necessary for final sizing and surface preparation, the process is frequently treated as operator dependent rather than quantitatively controlled.

This work examines a precision manual lapping workflow for beryllium target components with emphasis on repeatability, dimensional control, surface quality, and defect mitigation. A staged finishing approach was assessed using routine production metrology to monitor thickness uniformity, flatness, surface finish, and edge condition throughout processing. Particular attention was given to removal behavior across abrasive steps, the role of intermediate inspection, and common failure modes such as chipping, edge rounding, and nonuniform stock removal.

The results show that manual lapping can be structured as a controlled fabrication process when supported by defined inspection checkpoints, disciplined handling practices, and clear transition criteria between finishing stages. Quantitative relationships between lapping strategy and part quality are used to identify practical operating windows that improve yield and reduce rework. These findings provide a manufacturing-relevant framework for converting a traditionally experience-driven operation into a reproducible method for precision target component fabrication.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

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Femtosecond Green Laser Micromachining of Challenging Materials for Target Fabrication

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Precision target fabrication often requires cutting and localized material removal in metallic systems that are difficult to machine by conventional means. Mechanical methods can introduce burrs, edge damage, and geometric constraints, while longer-pulse laser processes may produce significant redeposition and undesirable thermal effects. Femtosecond laser processing offers a non-contact alternative capable of high spatial precision with reduced heat-affected damage.

This poster presents recent work using a femtosecond green laser for cutting and ablation of target-relevant metallic materials. The study focuses on manufacturing outcomes critical to component quality, including feature fidelity, kerf geometry, edge condition, debris generation, and evidence of thermally affected material. Comparative observations across multiple material systems are used to distinguish common process behaviors from material-specific responses in ablation morphology and defect formation. Optical microscopy, electron microscopy, and dimensional inspection are used to evaluate processed features and identify dominant quality limitations.

Results indicate that femtosecond green-laser processing can produce high-quality features in challenging materials while reducing thermal artifacts relative to conventional high-energy removal approaches. Key remaining challenges include controlling redeposition, balancing throughput with feature quality, and tuning parameters to accommodate material-dependent behavior. This work supports the development of robust ultrafast laser workflows for precision target fabrication where cleanliness, repeatability, and dimensional fidelity are essential.

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LLNL-ABS-2018232

Path-dependent Phase Selection in Compressed Cerium: Metastability and Microstructural Memory in an *f*-electron Metal

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Competing low-symmetry phases, sluggish kinetics, and strong hysteresis are recurring challenges in *f*-electron materials, yet they are often difficult to disentangle in chemically complex systems. Here we will show that cerium provides a particularly clean test case for how metastability and retained microstructure govern phase selection under pressure. Using neutron diffraction on large volumes of ultra-high-purity Ce, we tracked structural evolution during controlled pressure-temperature cycling between 85 and 295 K up to 8 GPa. The results reveal two distinct transformation pathways. Slow compression at 295 K produces a strongly textured orthorhombic α′ phase, whereas compression at 100–120 K stabilizes a pure monoclinic α″ phase. The α″ phase persists metastably to near room temperature but transforms irreversibly on heating above ~280 K or after modest pressure cycling. At the same time, a remnant fraction of the lower-pressure γ phase remains trapped far beyond its expected stability field, indicating that inherited parent-phase microstructure helps bias nucleation of the high-pressure state. These findings resolve longstanding contradictions in the cerium literature and show that its phase behavior is governed not by equilibrium thermodynamics alone, but by kinetics, history, and microstructural memory. More broadly, they provide an experimentally accessible framework for understanding competing structures and path-dependent transformations in *f*-electron metals relevant to actinide science.

Developing Machine Learning Techniques for Plutonium Manufacturing Equipment Predictive Maintenance Program

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This work aims to develop a machine learning (ML)–based strategy to support a robust predictive maintenance (PdM) program and maximize electrorefining equipment life. In electrorefining, metal is electrochemically purified by removing impurities to the anode and depositing a pure metal into the cathode. One problematic aspect of electrorefining is the need to continuously stir the molten metal for a long duration and stirring mechanism may unexpectedly fail. The current maintenance program is mainly based on reactive or preventive maintenance concepts, where equipment is used until failure or regularly scheduling maintenance work. Yet, streaming data generated by the equipment during their operation can be extremely valuable in identifying "signatures" of these impending failures buried in the data but otherwise not apparent during normal operation. We have developed a process monitoring and maintenance prediction system that will fuse data from acoustic transducers as well as power, temperature, and rpm and employ machine learning to predict when failure will occur. This document had been reviewed and assigned publication number: LA-UR-26-22886.

Investigating the amorphous oxide growth on plutonium metal

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At room temperature some metals such as aluminium, chromium and tantalum form a thin (<10 nm) amorphous oxide film that is passivating, preventing further oxidation. This is a particularly desirable property for radioactive metals where gross oxidation forms respirable particulate which is extremely hazardous when ingested. The formation of an amorphous oxide on plutonium has been suggested by the absence of an X-ray diffraction pattern before and after the sample was observed to visibly tarnish [1]. The possible existence of an amorphous oxide on plutonium metal is investigated further here using three different approaches. First, thermodynamic calculations have been undertaken of both crystalline and amorphous oxide films and are compared. Secondly, X-ray diffraction measurements have been performed on monoclinic α-Pu using an environmental cell following oxidation at room temperature followed by heating in high vacuum. The composition is determined from the diffraction patterns using quantitative Rietveld refinement. Finally, an interesting signature in the X-ray photoelectron spectrum is tentatively assigned to amorphous and crystalline oxide formation on plutonium metal substrates.

[1] A. J. Nelson, P. Roussel, J. Vac. Sci. Technol. A **31**, 031406 (2013).

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Helium Implantation Sample Preparation and Handling in the Small-Scale Transuranic Lab within the Materials Characterization Center

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Reliable sample preparation and mounting are critical to the success of helium implantation experiments. Previous runs using silver paint encountered contact-related issues by the time samples reached Center for Accelerator Mass Spectrometry (CAMS), preventing the experiment from moving forward and resulting in significant loss of time, cost, and coordination effort. This poster describes the helium implantation sample workflow carried out through the Small-Scale Transuranic Lab within the Materials Characterization Center (MCC), including machining, loading, transfer, and installation into the vacuum chamber. Because the work is performed in an actinide laboratory environment, the process involves extensive planning, radiological controls, rigging support, lift plans, and coordination across multiple organizations.

In addition to documenting the workflow, this work evaluates silver epoxy as a possible alternative to silver paint for sample mounting. Testing[MAM1.1] is focused on whether the new material improves contact reliability while still allowing sample removal after implantation for downstream use. Evaluation of bond strength through heat cycling will further inform material selection. This poster highlights both the complexity of the helium implantation preparation process and the importance of mounting material performance in improving experiment reliability.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

LLNL-ABS-2018278

Facility Expansion and Capability Growth in the Small-Scale Transuranic Lab within the Materials Characterization Center

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The Small-Scale Transuranic Lab within the Materials Characterization Center (MCC) has undergone a major expansion to better support actinide work for both National Ignition Facility (NIF) target fabrication and condensed matter physics (CMP) efforts. Previously limited to a single constrained workspace, the lab now spans three connected spaces, providing improved room for sample handling, equipment integration, storage, and overall workflow. The lab supports activities such as helium implantation chamber loading, DAC sample preparation, DCS work, and support for Z machine efforts, while also working in close coordination with the HiZ target fabrication group in the Superblock (SBK). Because this work is carried out in an actinide laboratory environment, even routine operations require specialized controls, radiological planning, and coordination across multiple groups.

This poster provides an overview of the motivation for the expansion and the capabilities the upgraded laboratory is intended to support. The increased footprint improves staging and working space for complex laboratory activities and creates room for future capability growth, including additional gloveboxes and new characterization tools. As part of the Small-Scale Transuranic Lab within the Materials Characterization Center, the expanded laboratory represents an infrastructure investment intended to improve current operations and increase long-term flexibility for actinide-related fabrication, preparation, and characterization work.

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LLNL-ABS-2018280

Solubility of U in Ga-Stabilized δ-Pu Shown by Electron Probe Microanalysis

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The behavior of U in Ga-stabilized δ-Pu is important for understanding the ingrowth of U during Pu aging. Traditional characterization methods of prepared alloys of the Pu-U-Ga phase diagram, such as optical microscopy, SEM-EDS, and XRD, either cannot show chemistry or cannot provide quantitative chemistry to show elemental distribution. Electron probe microanalysis (EPMA) using wavelength-dispersive spectroscopy (WDS) can provide quantitative data on grains smaller than 10 µm, with detection limits often below 0.1 wt. %.

We present the results of EPMA analysis on low-U (≤1 at%) alloys in the Pu-U-Ga phase diagram using quantitative mapping over 256x256 µm with a spot size of 1 µm. The data show U concentrations between 0.5 and 2 at. % in Pu grains, co-located Ga, indicating the material is Ga-stabilized δ-Pu. XRD analysis has verified the material as δ-Pu.

This abstract has been approved for publication by Los Alamos National Laboratory, LA-UR-26-21390.

Early-Stage Uranium-Hydrogen Corrosion Kinetics and Mechanism

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Research into the early-stage uranium-hydrogen degradation mechanism lacks unified interpretation and understanding. This is due to inadequate temporal and spatial-characterization resolution required to unify reaction observations. In this study, white-light interferometry was adapted to characterize the uranium-hydrogen reaction at a temperature of 50 °C and hydrogen pressure of 13.8 KPa. The hydride blister was monitored from induction through critical spallation and UH_3 powder release during rapid surface hydriding. The critical spallation dimensions were measured for height, diameter, area, and perimeter of the first blister. Hydriding growth kinetics were assessed by tracking the first hydride site over the reaction time. Analysis shows that the spall-front velocity of the hydride was changing at a rate of 0.91 µm/min which compares favorably with legacy Sievert's experiments in literature. This work highlights the strength of characterizing the early-stage uranium-hydrogen reaction using white-light interferometry, but more importantly unifying the understanding and mechanism of the uranium-hydrogen reaction kinetics.

Prepared by LLNL under Contract DE-AC52-07NA27344.

In-situ, Real Time Detection of Impurity Transport in Plutonium Electrorefining

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Electrorefining is the primary method used to purify plutonium (Pu) metal across the National Nuclear Security Administration complex. Present day operations rely on constant potential control in a two-electrode molten salt system, with periodic open circuit potential (back electromotive force, BEMF), measurements to determine the run endpoint. Limited real-time process visibility necessitates conservative operating conditions to minimize impurity carryover, resulting in long run times to purify a full-scale anode.

Lawrence Livermore National Laboratory developed a new method that can monitor impurity transport in real time during electrorefining while being controlled in a constant current state [1]. Under conditions where Pu is the only species transported, the system response follows Ohm's Law, producing a linear current-voltage relationship with the slope corresponding to the cell resistance. Deviation from the linear behavior provides an in-situ indicator of impurity transport and can be used to adjust current output in real time, enabling constant current operation while maintaining product purity targets. Proof of concept was demonstrated using a surrogate system with tin as the actinide surrogate and copper as an impurity in a LiCl-KCl-$CaCl_2$ molten salt system. A measurable deviation from linear behavior occurred when current surpassed 20 A, which correlates to a change in reaction potential between the electrodes confirming that tin is no longer the only constituent being transported. The newly designed method was implemented in a Pu electrorefining cell. The system was run at a conservative value of 30 A, reaching the endpoint in approximately 40 hours. Future work will focus on parameter optimization and full automation to minimize operator involvement. If proven viable, this approach could be the first step to in-situ real time process monitoring.

[1] Zhang, C., Parkes, A.M. and Holliday, K.S., 2023. First principles optimization of plutonium electrorefining. *Journal of Nuclear Materials*, 577, p.154327.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Transmission Electron Microscopy Characterization of He-Bubble Evolution in Naturally Aged and Annealed δ-Plutonium

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The α-decay process in Pu-Ga alloys leads to significant microstructural damage in the form of vacancies and self-interstitial atoms. As the α-particles come to rest, they acquire electrons to form He atoms, which become trapped at microstructural defects – like vacancies – and accumulate in the microstructure at a rate of about 41 appm/yr per 10^6 atoms of Pu-239[a]. Over time, the trapped He atoms build up to nucleate and grow He bubbles. Recently, He bubble sizes and distributions across naturally aged δ-Pu microstructures have been reevaluated with probe-corrected scanning transmission electron microscopy (STEM)[b, c], and a series of isothermal dilatometer experiments were performed to understand their evolution as a function of time and temperature. With the naturally aged parent material as reference, the present study builds from our STEM results to resolve bubble evolution in the dilatometer samples to specific microstructural features characterized with electron diffraction and STEM electron energy loss spectroscopy (EELS). As in the dilatometer studies of He bubble evolution performed by Wheeler and Kitching – for which TEM results were not published – significant He bubble growth is observed at higher temperatures, and these samples are the primary focus of the STEM characterization presented[d, e].

[a)] W. G. Wolfer, "Radiation Effects in Plutonium," *Los Alamos Science*, vol. 26, p. 274-285, 2000.

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[c)] T. M. Kucinski, D. S. Smith, M. A. Grismer, and G. M. Moore, "Improved Characterization of Helium Bubbles in Naturally Aged Plutonium with Scanning Transmission Electron Microscopy (STEM)," at *Plutonium Futures*, Edinburgh, 2026. Abstract submitted.

[d)] D. W. Wheeler, and P. D. Bayer, "Evaluation of the Nucleation and Growth of Helium Bubbles in Aged Plutonium," *Journal of Alloys and Compounds*, vol. 444-445, p. 212-216, 2007.

[e)] Unpublished work referenced in: D. W. Wheeler, and P. Roussel, "Review of the Behavior of He Bubbles in Aged Plutonium and their Influence on Material Properties," *Journal of Vacuum Science and Technology*, vol. 43, no. 020802, 2025.

Effects of Temperature, Ionic Strength, and pH on Aqueous Plutonium(VI) Speciation in NaCl, $MgCl_2$, and $Na(CO_3)_2$

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Plutonium solubility and speciation in environmental systems are controlled by oxidation state, the presence and concentration of complexing or competing ions, and the geochemical boundary conditions of the system, which ultimately determine mobility and sorption behavior. This behavior is of particular interest to performance assessments for geologic repositories, where the release of plutonium is a critical performance measure. These assessments rely on the availability of experimental data across a wide range of variables to describe all pathways of potential release. Historically, experiments have been performed at 25 ℃ and relatively low ionic strength ($I \leq 0.1$ m); the high temperatures and ionic strengths expected in geologic repositories are not widely represented in the available data.

Conditions representative of geologic repositories, such as high temperature and ionic strength, will impact actinide solubility and speciation. Previous research shows that actinide hydrolysis and complexation increase with increasing temperature, leading to higher stability ranges of complexes with low or neutral charge.[1] Actinide complexation with chloride and carbonate is ionic strength dependent, with increasing complexation in higher ionic strength systems.[2,3]

The lack of available data in thermodynamic databases and limitations of the models designed to predict the behavior of actinides in extreme environments highlights the need for reliable experimental data on the complexation of actinides in solutions at temperatures, ionic strengths, and pH values expected in a geologic repository. To address the complexity of expanding the range of temperature and ionic strength, UV-vis-NIR spectrophotometry was used to study the effect of temperature (T = 25–95 °C), ionic strength (I = 0.1–10 m), and pH (pHi = 5–12) on Pu(VI) speciation. Salt solutions of NaCl, $MgCl_2$, and Na_2CO_3 were used for the ionic strength systems.

Pu(VI) complexation in chloride solutions is driven by increased ionic strength, pH, and temperature. In the NaCl and $MgCl_2$ systems, peaks indicative of the mono- (837 nm), di- (843 nm), and trichloro (849 nm) species were observed. Additional peaks were observed at 851, 857, and 861 nm, which may indicate the formation of the tetrachloro species, or products related to hydrolysis or carbonate complexation. Increased complexation with chloride was observed with increasing pH, ionic strength, and temperature in both systems. However, the impact of temperature on the Pu(VI) speciation in the $MgCl_2$ system was significantly less than in the NaCl system. The speciation of Pu(VI) in the presence of equimolar Na_2CO_3 was studied as a function of temperature to determine contributions from carbonate species in the chloride systems.

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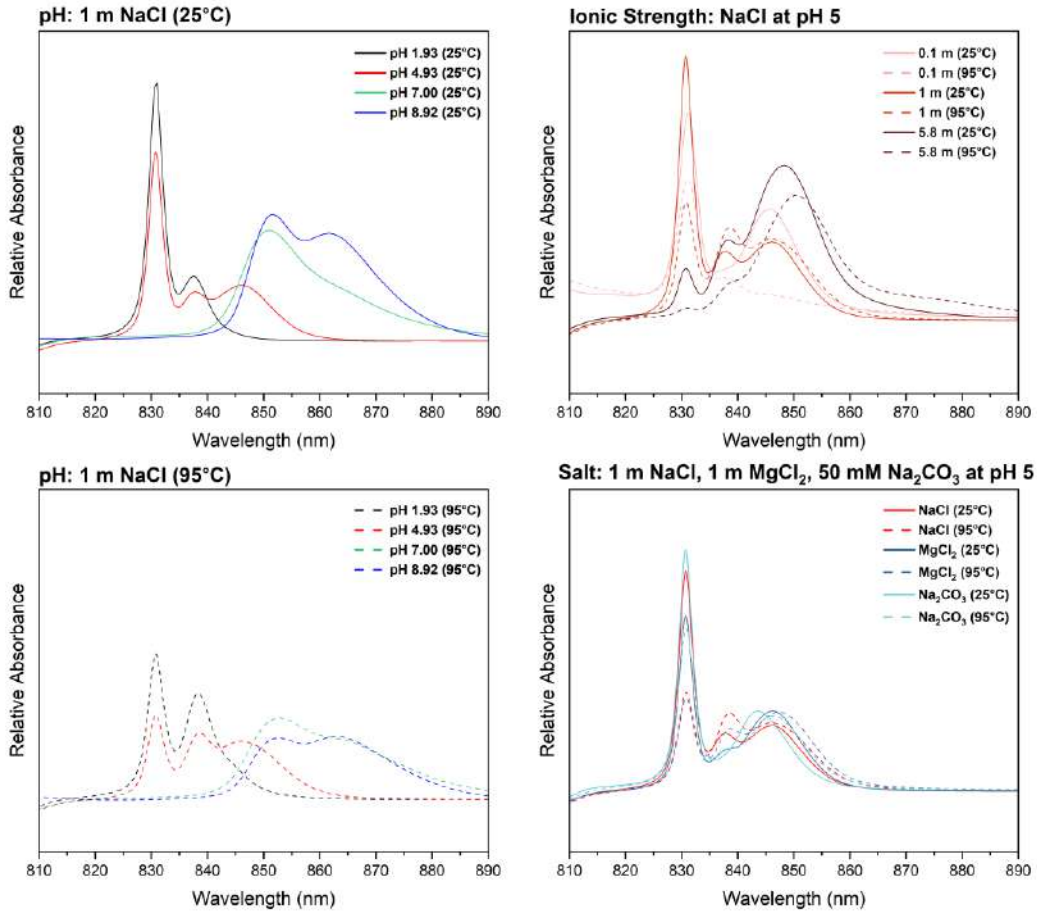


Figure 1: UV-vis-NIR data for Pu(VI) comparing speciation dependence on (Top, Left) pH in 1 m NaCl (25 °C), (Bottom, Left) pH in 1 m NaCl (95 °C), (Top, Right) ionic strength in NaCl at pH 5, and (Bottom, Right) salt content, including 1 m NaCl, 1 m $MgCl_2$, and 50 mM Na_2CO_3 at pH 5.

Actinide Research Quarterly

Owen Summerscales[1*], Franz Freibert, Ping Yang

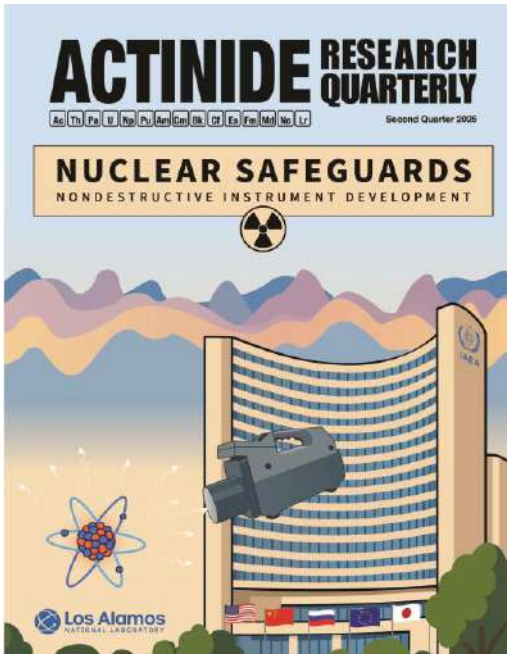
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Actinide Research Quarterly (ARQ) is a publication of the Glenn T. Seaborg Institute for Transactinium Science at Los Alamos National Laboratory (LANL). Established in 1994, it highlights research in actinide science in such areas as process chemistry, metallurgy, surface and separation sciences, atomic and molecular sciences, actinide ceramics and nuclear fuels, characterization, spectroscopy, analysis, and manufacturing technologies. **ARQ is the only publication in the world that covers all aspects of actinide science in one magazine**.

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Non-Destructive Rapid Turnaround Characterization to Support Plutonium Experimentation

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Plutonium-based experiments are necessary for advancing our understanding of this exotic element. Characterization of related experimental products, materials, and processes, often can introduce additional challenges especially when plutonium and other radioactive contaminants are present. Therefore, introducing non-destructive and rapid turnaround characterization techniques and methods, such as X-Ray Computed tomography (XCT), to plutonium-based experiments will increase safety and ensure deadlines and deliverables are met. One area that is currently benefiting from this characterization technique is the application of protective coatings for plutonium environments. These protective coatings are prepared with a molten salt electrodeposition method. Advancing molten-salt electrodeposition as an efficient fabrication methodology requires comprehensive multi-scale characterization to assess coating uniformity, defect evolution, and long-term performance. In this work, coatings produced in a controlled inert-atmosphere molten-salt environment were characterized using a suite of electrochemical, microscopy, and tomography techniques. These characterization methods are presented and evaluated against each other to examine the molten salt electrodeposition method as well as the plutonium and other metal exposure assessments.

Surface morphology and microstructure were evaluated using digital microscopy, profilometry, and scanning electron microscopy (SEM), while chemical uniformity was examined with energy-dispersive X-ray spectroscopy (EDS). Subsurface structure was resolved using XCT, which enabled non-destructive three-dimensional quantification of coating thickness, uniformity, and overall defect characterization. Presented results will include details on protective refractory metal coatings on nickel alloy substrate as well as molten plutonium exposure assessments on the protective coatings. These characterization methods were used to evaluate the hermeticity and materials compatibility of the coatings for application in plutonium environments, where chemical stability, defect tolerance, and barrier integrity are critical. The presented work and related dataset presents a framework and comparison when evaluating experimental procedures involving plutonium.

Modelling Heat Transfer, Diffusion, Water Adsorption and Radiolysis in PuO_2 Powder under Repacking and Storage Conditions

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During retreatment and repacking of PuO_2, glovebox atmospheres are inerted to prevent moisture adsorption; however, under fault conditions, this may not be achievable. Radiolysis of the water results in the generation of hydrogen, with the potential for the formation of a flammable atmosphere and/or pressurisation of a sealed container. The ability to model the rate of water adsorption, as well as predicting the consequences of long-term storage, plays an important part in establishing process design and safety.

A 2D model of PuO_2 powder in a cylindrical container was developed to simulate heat transfer, diffusion, adsorption and radiolysis. This model has been applied to open containers to determine the rate of moisture ingress and adsorption, and to sealed containers to predict pressure, atmospheric composition and humidity changes within a container during storage. During validation, the model was found to predict temperatures accurately to within 4% of experimental values. Mass gain due to adsorbed water was overpredicted by between 32-38% for Magnox inner cans by the end of each experiment, which makes for a reasonable and conservative estimate.

Further development is planned to include containers in alternative configurations, as well as convection, radiative heat transfer, liquid water accumulation and additional species and reactions. Expansion of the physical property data used by the model is also planned.

Structural and Thermophysical Properties of Pu Intermetallics

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Plutonium is arguably the most complex element in the periodic table. It sits near the middle of the actinide series, precisely in between where the 5*f* electrons are involved in bonding, as in the transition metals and light actinides (Th-Np), or are localized close to atomic core and do not contribute to bonding, as occurs in the lanthanides or heavy actinides (Am-Lr). This itinerant to localized crossover can be observed in the trends of the Wigner-Seitz radii of the transition metals, lanthanides, and actinides, where plutonium, particularly its cubic δ-Pu phase, lies in between localized and itinerant behavior. The near degeneracy of these two bonding configurations leads to a variety of unusual behavior, including six different allotropic phases, mechanical properties ranging from extremely brittle to ductile, and the largest Sommerfeld coefficient of all the elements. These interesting properties are derived from the 5*f* electrons of plutonium. An understanding of the subtle changes in electronic structure at this localized/itinerant crossover of Pu's 5*f* electrons is still lacking. Experiments capable of revealing the electronic structure, such as angle-resolved photoemission or de Haas van Alphen measurements, have not been possible because single crystalline samples of Pu are not easily synthesized. However, single crystals of plutonium compounds, which exhibit a range of behavior from localization to itinerancy of Pu's 5*f* electrons and are suitable for these measurements, may shed light on the physics near the localization/delocalization transition.

Pu_6M (M = transition metal) compounds crystallize in the U_6Mn-type with the tetragonal space group *I*4/*mcm* (No. 140). The building blocks that compose the Pu_6M structure are Pu1 distorted octahedral units and Pu2 face-shared square antiprisms that have transition metal atoms preferentially occupying their centers. The Wyckoff positions of the Pu1, Pu2, and M atoms are 8h, 16k, and 4a, respectively. The Pu_6M compounds can be easily prepared by arc melting. The rich structural chemistry, microstructure, and thermophysical properties of these compounds are exemplified through a suite of techniques including X-ray diffraction, heat capacity, and electron microscopy.

Plutonium Casting Modelling – Beyond the Present

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Casting modelling has become widely used for various industrial applications. In general, modelling techniques such as Computation Fluid Dynamics (CFD), Finite Element (FE) and Meso-Scale (MS) modelling are utilised as a research tool to aid optimisation of manufacturing processes; therefore, it has become a powerful ally. These techniques have helped to better understand the complexities and natural behaviour of molten metal flow, assess turbulence generated during mould filling, helped to develop an in-depth understanding of the complex interactions between molten metal flow and mould material, predict casting defects, segregation, thermal stress behaviour and assess the average cooling rate experienced during the casting processes, which all have a profound effect on the metallurgical properties of the casting being produced.

Casting plutonium alloys is both complex, time-consuming affair involving multiple stages, these include; Vacuum Induction Melting (VIM) and annealing. By their very nature, studies involving plutonium alloys are inherently difficult and therefore, future studies are likely to be limited due to a number of factors which include; high associated costs and health and safety considerations. As such, development of the plutonium casting processes has been limited and the fundamental casting process design has seen little improvements in the last 20 years. The casting and homogenisation processes are simple by design, gravity VIM casting and isothermal annealing however; when considering the number of complex metallurgical and physical phenomena that occur, some of which occur simultaneously, developing a validated process model encompasses all the phenomena, becomes a true challenge.

Modelling the heating cycle of the casting process is currently beyond the capability of the modelling software used (Keysights ProCAST), as it involves modelling the complex interactions in the form of eddy currents and hence, several thermal examinations using the non-active area casting furnace were conducted. Numerous thermocouples were attached to the various components inside the furnace and the temperatures were recorded as a function of time. It was concluded that prior to pouring being initiated the majority of components inside the furnace reached a thermal steady-state. These steady-state temperatures as a function of mould temperature are plotted in Figure 1.

In developing the initial thermal flow FE model the steady-state temperatures associated with each component were extracted and used as a starting point. The simulated results from the initial thermal flow FE model were compared to thermocouple data obtained from actual castings. Thermocouple data revealed that the casting experiences an average cooling rate of between 3 – 3.5°C per minute through the $\varepsilon + \delta$ mixed-phase region. Results from the FE model showed an average cooling rate of 3.2°C/ minute and these can be found in Figure 2.

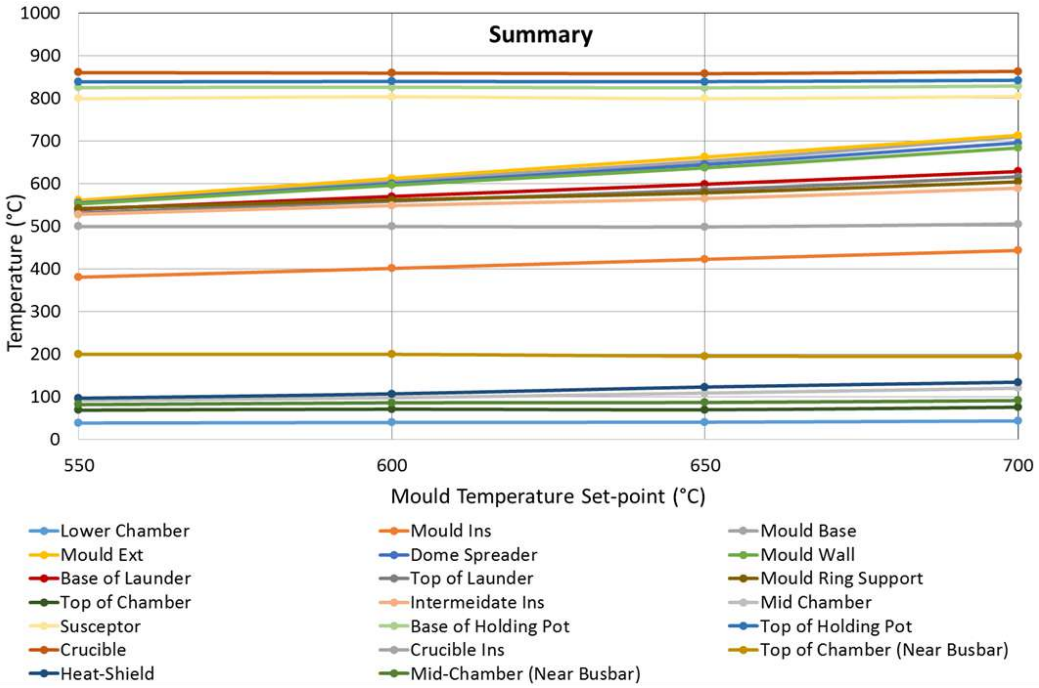


Figure 1: A summary of the thermal examinations carried out using the non-active area furnace for various heating cycles.

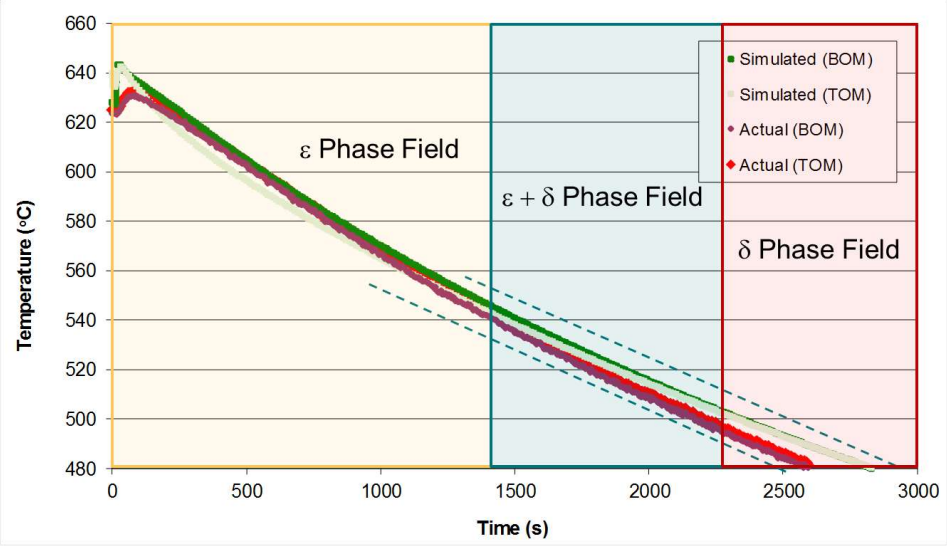


Figure 2: shows the process thermocouple data for plutonium alloy castings compared against nodal temperatures found at a similar thermocouple location from the model.

In conclusion, most of the physical phenomena can be modelled using commercially available software; the challenge of predicting the metallurgical phenomena i.e., grain size, segregation behaviour and solid-state phase transformation at the length scales of interest to engineers still remains. The thermal-flow FE model still requires further development and validation, a time-line for model development has been proposed, once validated this model will be utilised to guide future process improvements and aid in designing a future plutonium casting capability.

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Carbothermal reduction sintering of MOX fuel for fast-neutron reactors

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1. Introduction

The oxygen-to-metal (O/M) ratio of MOX pellets for fast-neutron reactors is imposed during sintering in a hydrogen-rich atmosphere. In mass-production sintering furnaces, the O/M ratio of MOX pellets rarely falls below 1.98 due to poor gas flow. This study experimentally evaluated the carbon thermal reduction method, which involves sintering MOX powder with added graphite powder, as an alternative to the reducing gas method for achieving low O/M ratios.

2. Experiments

MOX powder containing 30% Pu (prepared by mixing UO_2 powder with 50% Pu-MOX powder without prior heat treatment) was blended and ball-milled with graphite powder at concentrations of 0, 3000 ppm, and 6000 ppm, and hand-pressed into 10 mm high specimens using a 3 mm press die. These compacts were then heated at a rate of 10°C/min to 1650°C using a thermal balance (TG) connected to an oxygen-control system, held at that temperature for two hours, and cooled at a rate of 30°C/min. The sintering atmosphere consisted of Ar + 4.5% H_2 gas (without humidification).

3. Results

The variations in the O/M ratio of MOX were evaluated based on the weight changes obtained using the TG setup. Figure 1 shows the behaviour of the O/M ratio of a MOX sample (A), to which 3000 ppm of graphite was added, and sample (B), to which no graphite was added. The rate of the change in the O/M ratio slowed at around 400°C. At temperatures between 1300 and 1400°C, the O/M ratio of sample (A) was lower than that of sample (B). After cooling, sample (A) achieved an O/M ratio of 1.979(1), whereas sample (B) had a higher value of 1.984(1). An O/M ratio of 1.906(1) was obtained with a graphite addition of 6000 ppm, but the density of the sintered body decreased significantly, leading to the exclusion of this concentration due to poor mechanical properties.

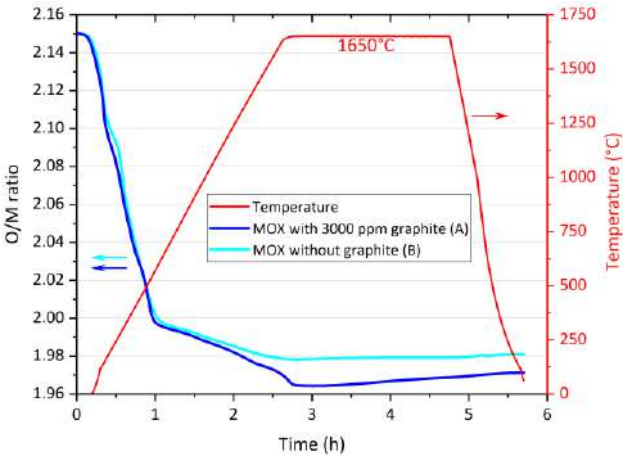


Figure 1. Variations in the O/M ratio of MOX containing 3000 ppm graphite (A), and without graphite (B), as a function of time and temperature during sintering in Ar + 4.5% H_2

4. Conclusions

It has been found that adding 3000 ppm of carbon powder can effectively reduce the O/M ratio during the sintering process of MOX. For mass-production-scale sintering furnaces with poor gas flow, adding graphite to the feedstock powder is a promising method of reducing the O/M ratio of MOX for fast-neutron reactor applications.

The Plutonium-Hydrogen Interaction Studied via X-ray Photoemission

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Hydrogen has been shown to greatly increase the corrosion of plutonium resulting in adverse implications for safe storage and stability. Therefore, understanding the nucleation and growth of plutonium hydriding is critical in helping to understand this corrosion phenomenon. In this study, hydrogen gas dosing is performed on alloyed plutonium at different temperatures and probed with X-ray photoemission spectroscopy to better understand the early kinetics and thermodynamics of plutonium hydriding. Dosing is completed at a rate of 0.5 Langmuir (L) per second with total dosing ranging from 5-400 L. Depth profiles, probing the propagation of the nucleated hydride are completed at 200 L for each temperature studied. These results are discussed to provide context to the interplay of hydrogen solubility in the metal, hydride nucleation, and the hydride growth rate.

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Pu L_{III} and Fe K Edge X-ray Absorption Spectroscopy of Bulk Pu_6Fe Synthesized via Arc Melting

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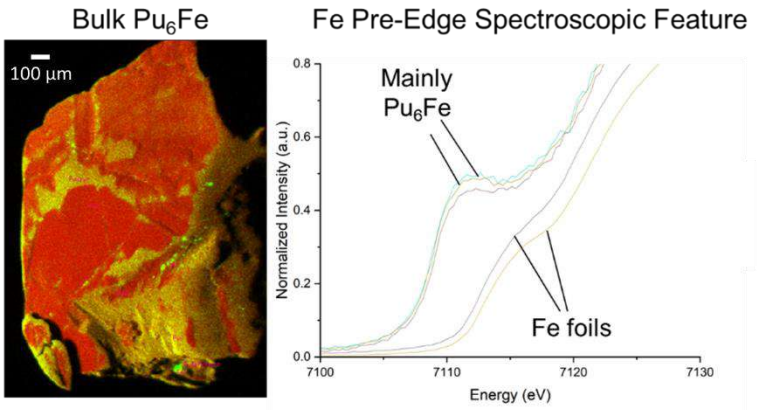


Figure 1. Elemental map (Pu – red, Fe – green) of an arc melted piece of Pu_6Fe without surface preparation. Maps were collected in fluorescence mode at beamline 2-3, SSRL, X-ray spot size ~5 µm.

Plutonium has the most complicated phase behavior of any single element, exhibiting no less than 6 crystalline phases between room temperature and melting. This inherent complexity of Pu is only exacerbated when it is alloyed, either intentionally or incidentally as with impurity elements. Among others, systems containing Pu and Fe were studied early in the history of Pu materials science, where the Pu_6Fe eutectic alloy was used in the first metallic plutonium fuel elements, which powered the Los Alamos Molten Plutonium Reactor in the late 1940s. The presence of the Pu_6Fe has also been identified in Pu-Ga alloys containing Fe impurities via TEM by scientists at Lawrence Livermore National Laboratory (2002). Synthesis of Pu_6Fe has been executed recently at Los Alamos National Laboratory by fusion of powders, and then for this work via arc melting of metallic Fe and Pu components in stoichiometric proportions. Although the phase space of the Pu-Fe system has been generally explored, atomic scale structural characterization, including manifestation of impurity phases as a result of processing conditions, has not been fully realized for this compound. X-ray absorption spectroscopy has proven viable for characterizing a wide range of local structural effects in general, as well as in Pu alloys specifically, hence our application of this technique to Pu_6Fe. Herein we present the first fluorescence mode X-ray absorption spectroscopy of this material, including Pu L_{III} edge and Fe K edge fluorescence data for two separately synthesized and processed pieces of Pu_6Fe. We seek to establish X-ray absorption signatures for this material and to make a start at associating these signatures with processing conditions, thereby providing a basis for future study of this system.

Two pieces of Pu_6Fe, each synthesized separately, were analyzed using fluorescence mode X-ray absorption spectroscopy at beamline 2-3 within the Stanford Synchrotron Radiation Light source (SSRL) at SLAC National Accelerator Laboratory. This analysis resolved the Pu_6Fe phase itself, as well as various other phases such as alpha plutonium, $PuFe_2$, and various degrees of oxidation exhibited in both the Pu and Fe spectroscopic data (see elemental map in Figure 1). While oxides were observed each set of data, plutonium oxide was present more often and with higher scattering intensities. This could suggest that the plutonium within the Pu_6Fe or $PuFe_2$ phases may oxidize more readily than the iron. Somewhat unexpectedly, the Fe pre-edge features in the from both pieces provide an interesting signature for Pu_6Fe and its associate phases (Figure 1). Point spectroscopy associated with Pu_6Fe, $PuFe_2$, or other phases shows

a clear difference, in the form of an intense single shoulder feature, in the pre-edge compared to metallic Fe or Fe_2O_3. Pre-edge features are influenced by a variety of factors, from simple symmetry considerations to electronic density of states and bonding interactions. While deconvoluting or modeling those features are out of the scope of this work, we nevertheless call attention to transition metal pre-edge features, within the X-ray absorption spectra, as a possible probe for bonding and/or hybridization in actinide intermetallics.

Study of the hydriding behaviour of cerium containing iron-rich impurities

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Cerium is used here as a non-radioactive surrogate for plutonium to demonstrate and optimize devices in a new glovebox dedicated to plutonium studies. The glovebox contains devices with camera and a PM-IRRAS (Polarization-Modulation InfraRed Reflection-Absorption Spectroscopy) device.

Hydriding reaction of cerium is studied. H_2-cerium interaction initiates with the formation of localized hydride sites as for plutonium.

The objective of this work is to investigate the morphology of these hydride sites and to identify the nature of the hydride nucleation sites. Focused ion-beam scanning electron microscopy (FIB-SEM) associated with energy dispersive X-ray spectroscopy (EDS) was applied for this purpose.

Figure 1 shows a hydride site and the corresponding EDS iron map. As for many other hydride sites on the sample, an iron-rich zone is observed near the centre of the site. Some of the Fe-rich inclusions present in the cerium used in this study could play a role in the hydriding behaviour. Figure 2 shows the cross-sectional morphology of this hydride spot exhibiting a complex microstructure with large cracks.

Characterisations are still in progress to elucidate the nature(s) of nucleation sites on cerium and similar studies are also underway to investigate the plutonium hydriding behaviour (nucleation and growth, nucleation sites).

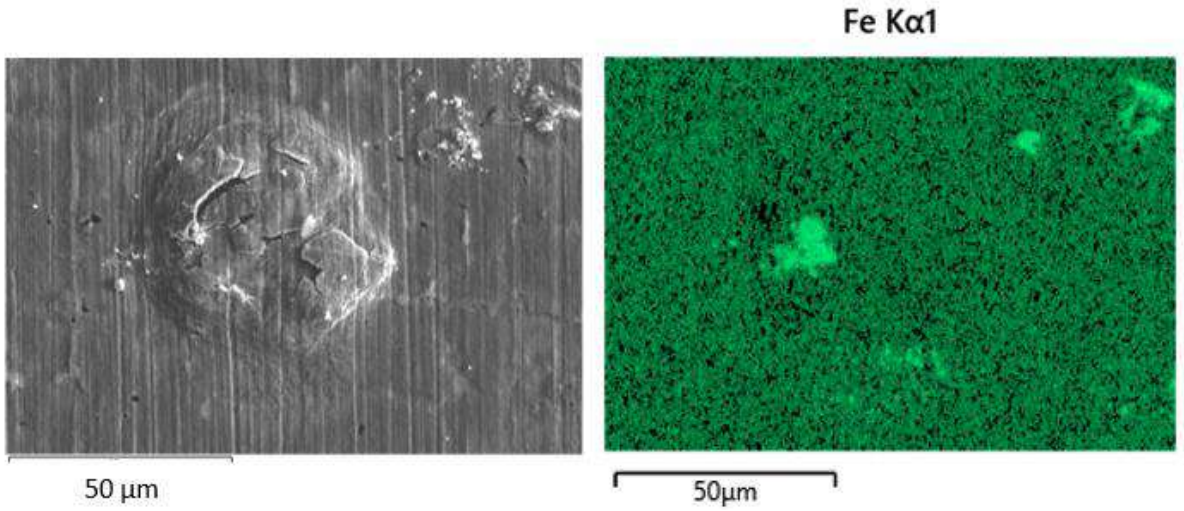


Figure 1: Cerium hydrided at ambient temperature under 530 mbar H_2:
(a) Secondary electron image of a hydride site, (b) EDS map of Fe K-edge of the same area.

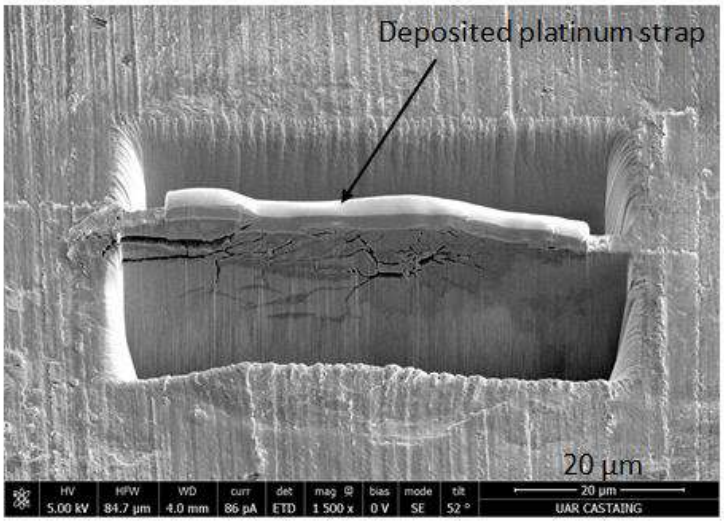


Figure 2: Secondary electron image of the FIB cross-section of the hydride site shown in Figure 1.

A Multiphase Equation of State for Ce

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We present the development of a multiphase equation of state (EOS) for Ce that includes five distinct phases, namely the γ, α, α' and ε solid phases, together with the liquid phase. The EOS has been constructed to provide a consistent thermodynamic description over a broad range of temperatures and pressures, and it reproduces a wide set of experimental and theoretical constraints. In particular, it captures the ambient thermal expansion behavior, static isothermal diamond-anvil-cell measurements, shock Hugoniot response, and shock melting behavior with high fidelity. In addition, it matches first-principles full-potential linear muffin-tin orbital (FPLMTO) calculations of compressibility beyond the experimentally accessible regime, thereby extending confidence in the model to higher pressures. The EOS further predicts a liquid-vapor critical point near 9000 K, offering insight into the expansion behavior of Ce.

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Beyond Hardness: Measuring Elastic and Dissipative Properties by Nanoindentation

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The nuclear materials community is frequently constrained by limited material availability and challenging experimental conditions. Advanced nanoindentation provides a powerful approach for characterizing mechanical behavior at the nanoscale while minimizing required sample volume. In this work, a single plutonium specimen is used to measure hardness, elastic modulus, and internal friction.

Unlike conventional microhardness testing, hardness is determined from the load–displacement response rather than optical measurement of the residual indent. Analysis of the elastic unloading segment of the curve further enables extraction of the elastic modulus. In addition, the nanoindenter is operated as a dynamic mechanical analyzer (nanoDMA) to measure frequency-dependent mechanical response, allowing quantification of dissipative behavior, or internal friction.

This approach enables characterization of bulk regions, small-scale microstructural features, and intermetallic phases within the same specimen, eliminating the need for multiple samples and extensive preparation for different testing platforms. Nanoindentation results are compared with bulk measurements obtained using complementary techniques, including microhardness testing and resonant ultrasound spectroscopy (RUS), to establish consistency across length scales.