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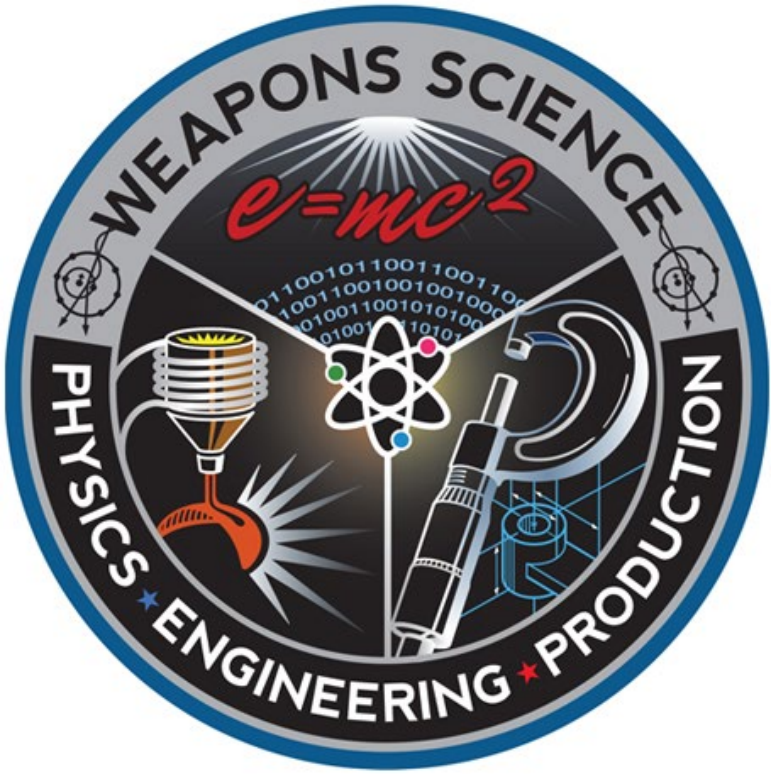
Nuclear Fundamentals Orientation Module 2

Nuclear Weapons Production- Materials Cycle

Managed by Triad National Security, LLC for the U.S. Department of Energy's NNSA

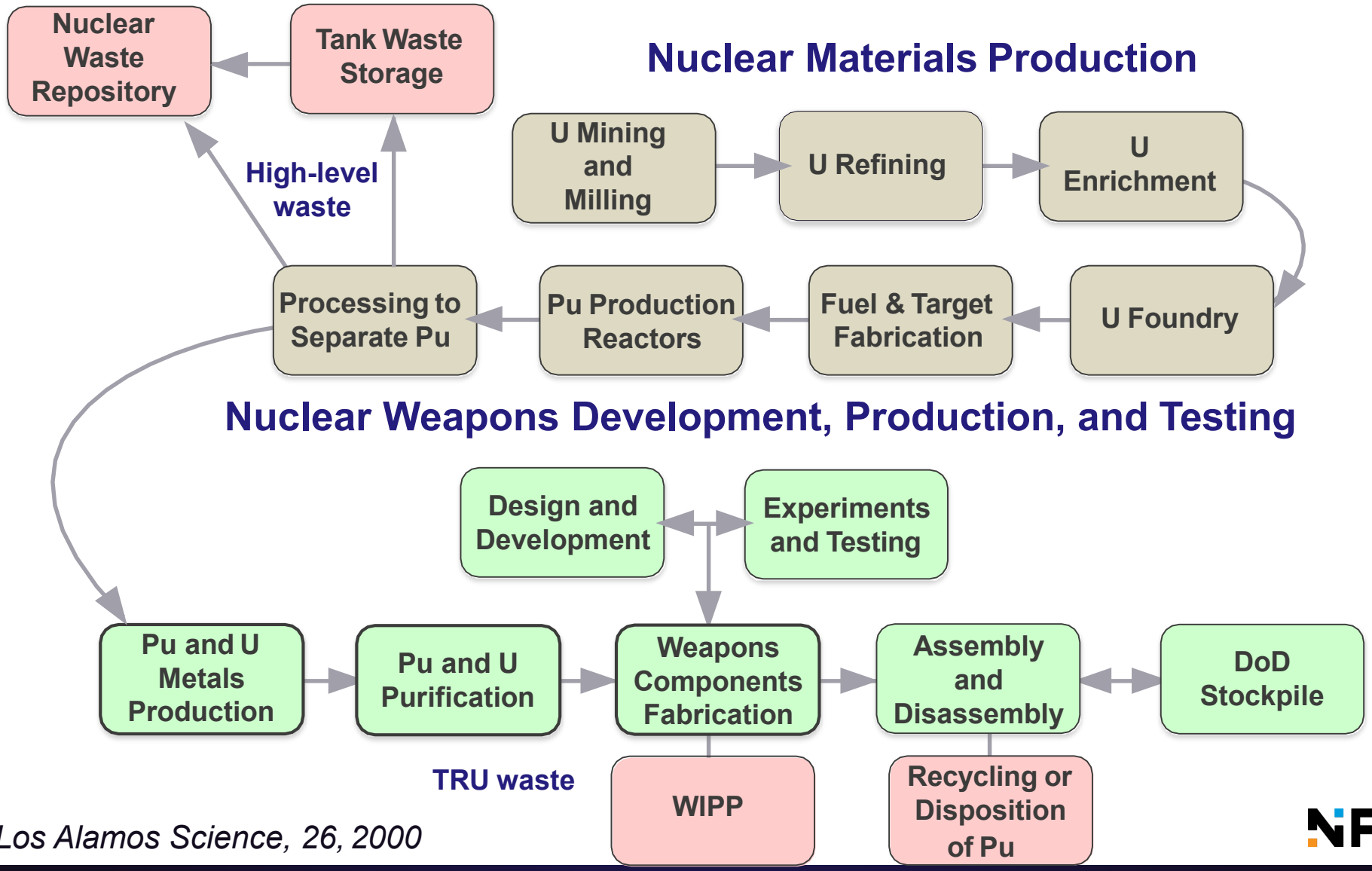
Nuclear Weapons Production- Materials Cycle



Presentation Overview:

- This presentation will familiarize participants with the production of plutonium, its separation, conversion to metal, casting, machining, and assembly of special nuclear materials common to nuclear weapons, and the disposal of nuclear waste
- Focus in this talk is plutonium (in future briefings we will broaden to include other materials)
- Questions and Answers

U.S. Nuclear Weapons Cycle Required a Massive Infrastructure



Los Alamos Science, 26, 2000

Plutonium in 2019

- > 2800 metric tons (tonnes)
  - Spent fuel
  - Nuclear weapon's components
  - Various nuclear inventories
  - Legacy materials
  - Wastes
- 515 tons separated Pu
- There is a continuing obligation to understand and manage this material responsibly in all applications.
- Los Alamos serves a key role as a consolidated Center of Excellence for Pu activities against this national obligation

Abright, Global Stocks of Nuclear Explosive Material, 2015
Plutonium Handbook, Chapter 5, 2019

Pu Button

Rocky Flats Plant
SNM Vault x-y retriever

Plutonium Challenges

Nation faces new challenges due to dramatic changes in the nuclear weapons programs of the nuclear powers

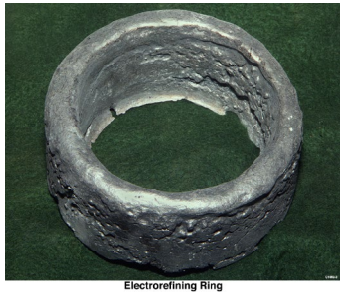
- Ensure weapons are safe & reliable
- No nuclear testing
- No new cycle of development & deployment
- Remanufacture of plutonium components
- Stabilize & store excess nuclear materials
- Secure against theft & diversion
- Burning as fuel in nuclear reactors
- Disposition of radioactive waste
- Emerging threats of nuclear terrorism
- Environmental cleanup of Cold War Legacy
- Training the next generation

ICBM launch from Submarine

Nuclear Waste in TRU Packs delivery at WIPP

Plutonium Science & the Los Alamos Mission

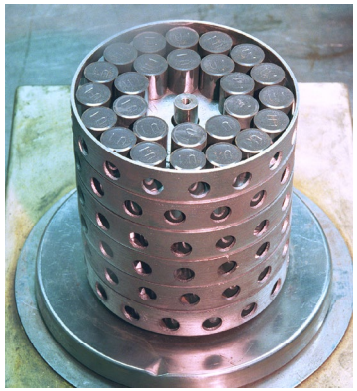
- Process, separations & conversion chemistry
- Metal preparation
- Metallurgical & physical properties of actinide alloys
- Fundamental condensed matter physics of actinide compounds
- Nuclear fuels production
- Nuclear waste isolation and storage
- Heat source fabrication for interplanetary exploration
- Nuclear materials stabilization
- Characterization and transport modeling in homeland defense



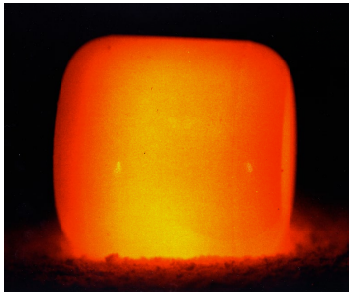
Electrorefined Pu Ring

Direct Oxide Reduction salt charge and Pu button (bottom)

Storage can swelled by Pu hydriding reaction



Trays of Clad Heat Sources

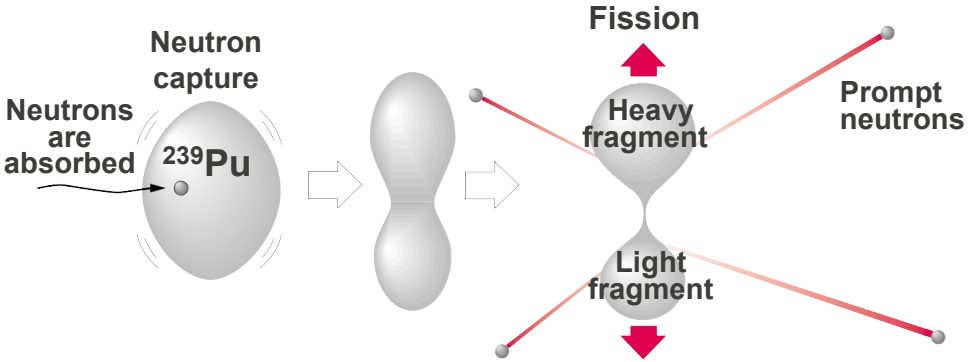


Heat Source: Pu-238 oxide

What Makes Plutonium So Complex and Difficult?

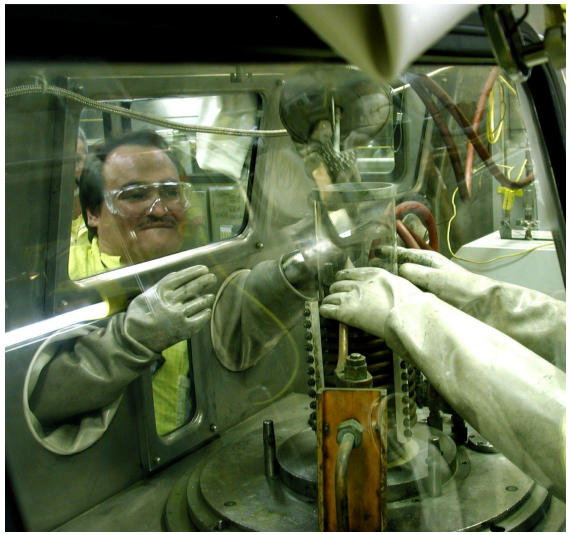
The science is complex - the 5f electron story

- Neutrons provide a “factor of millions” in energy
- But its the nature of the electrons that makes plutonium one of the most complex of all elements



The rest is difficult

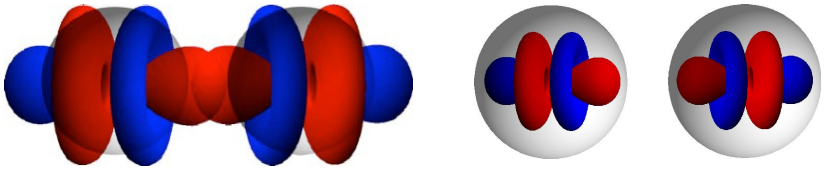
- Radioactivity, toxicity - handling challenges
- Regulatory environment
- Cost – specialized facilities
- Secrecy - limits talent and peer review
- Programmatic priorities



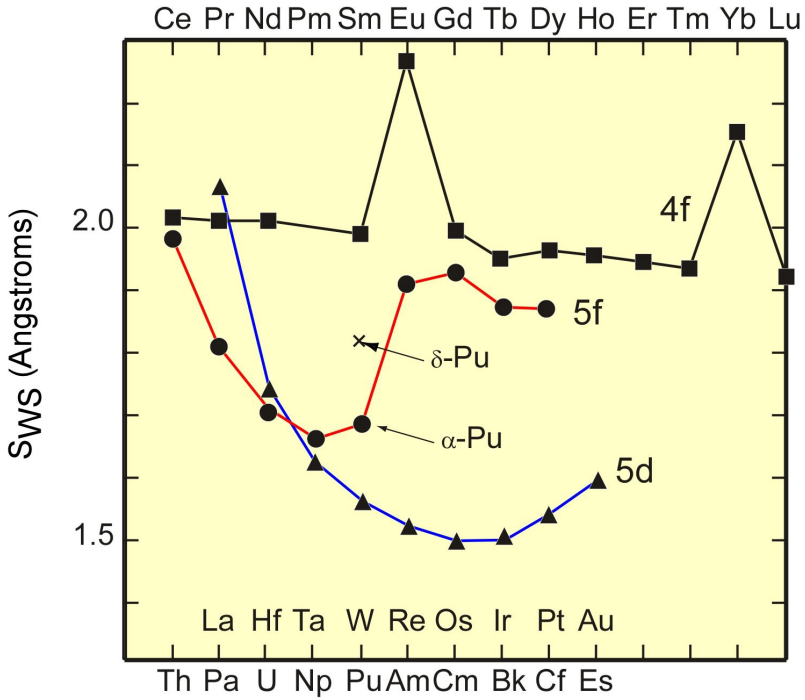
Pu Casting Preparation

What is it about the electrons that makes Pu so different?

- Plutonium has *5f* electrons in the valence shell (an actinide)
- Roles of *5f* electrons can vary from being localized (nonbonding) to delocalized (bonding)
  - In pure phases, alloys, compounds and molecules
- Delta phase plutonium has an exotic electronic structure
  - 5f electrons are neither fully localized or delocalized
- Exotic electronic structure gives rise to unusual and puzzling behaviors
  - Apparent in the rich phase diagram elemental plutonium. Satisfactory electronic structure



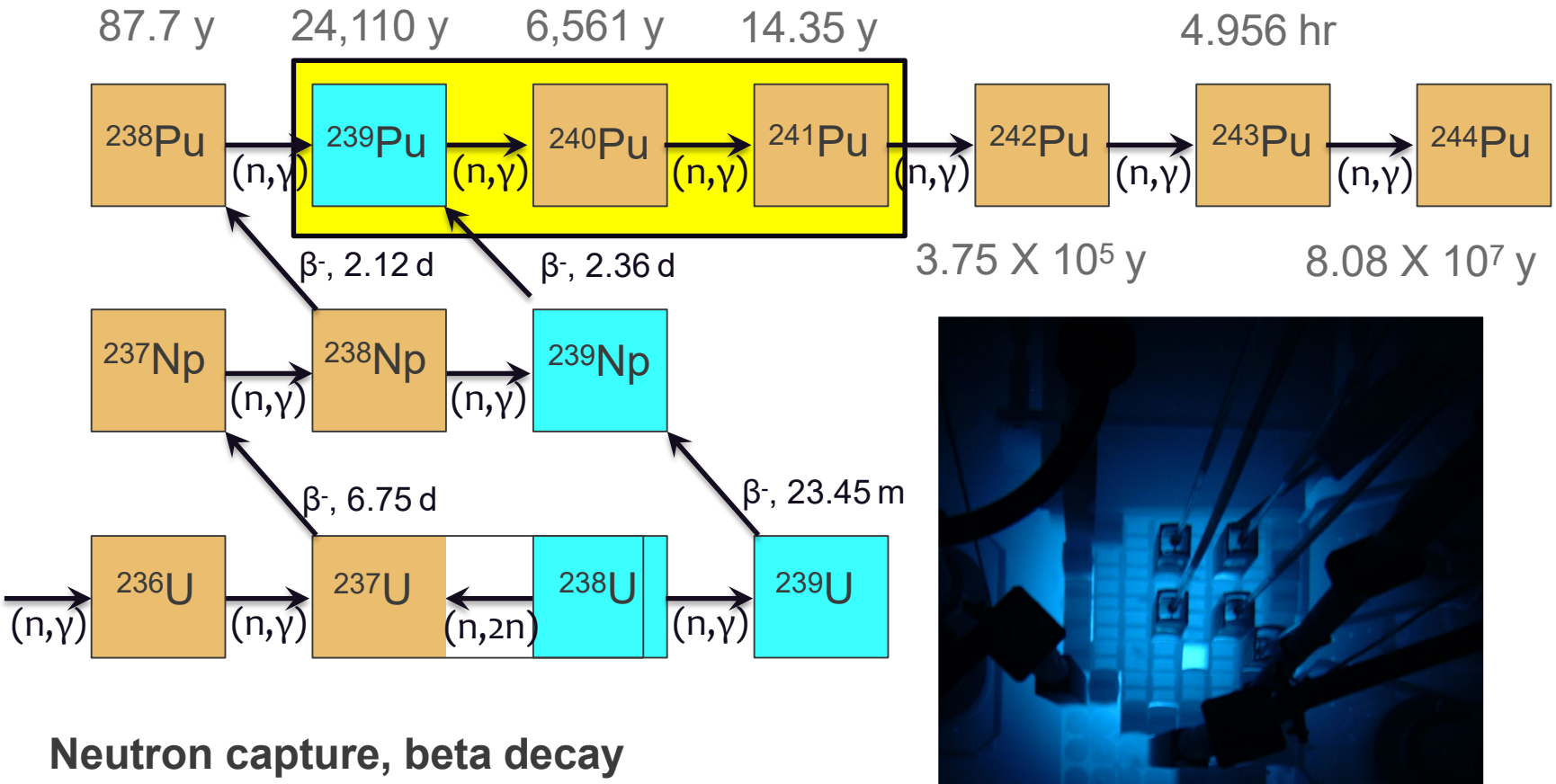
Metallic electrons **Atomic electrons**



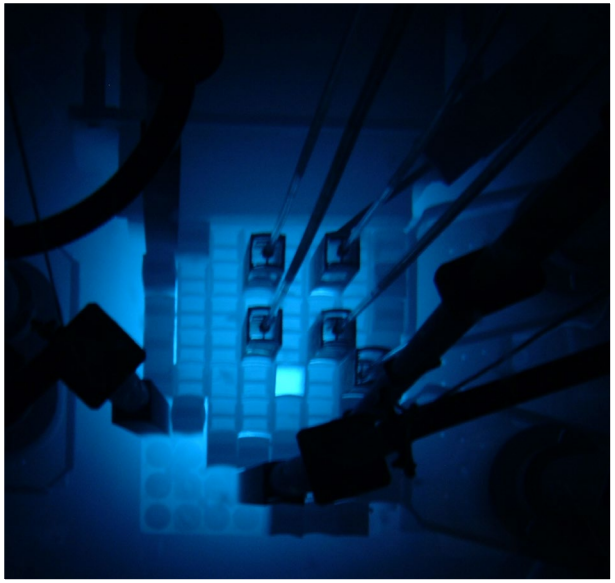
Savrasov, Kotliar, Abrahams, *Nature*, **2001**, *410*, 759
Wills, Eriksson, **2000**, *Los Alamos Science*, 26, 128

Preparation of Plutonium

Plutonium Buildup in Nuclear Reactors



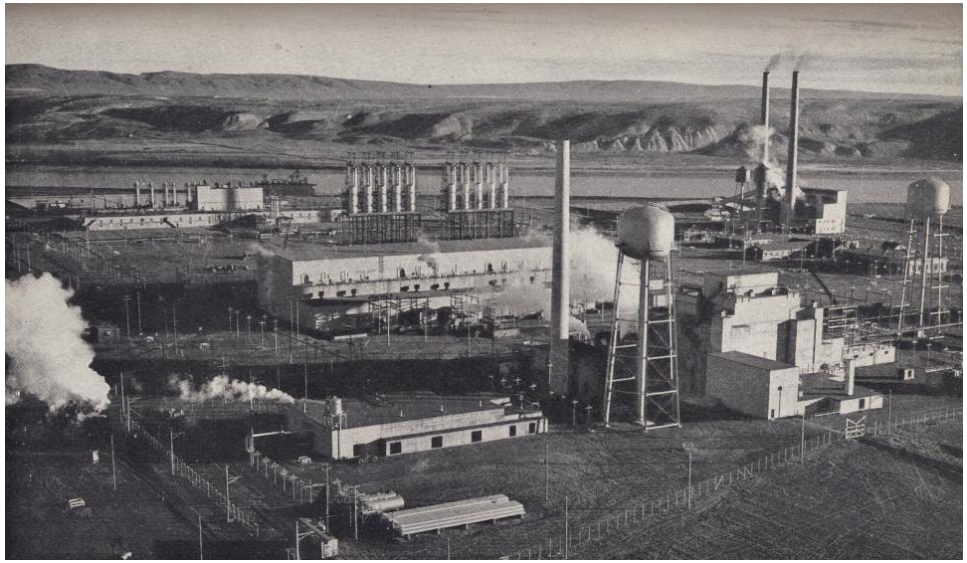
Neutron capture, beta decay



Cherenkov radiation glow of a nuclear reactor

Clark, Hecker, Jarvinen, Neu, *Chem. Act. and Transact. Elements*, Chapter 7, **2006**

Production of Uranium and Plutonium, 1943



Hanford Nuclear Reactor Facility

Plutonium (^{239}Pu):
"B" reactor construction began in March of 1943 and came on line in September of 1944!!

Uranium (^{235}U):
The K-25 gaseous diffusion plant, at the time the largest single factory under one roof in the entire world

Oak Ridge Uranium Enrichment Facilities

The Pu Separations and Purification Challenges

- Find a means of separating Pu in high yield and purity from the many **tons of U** in which Pu was present at a **maximum concentration of 250 ppm**.

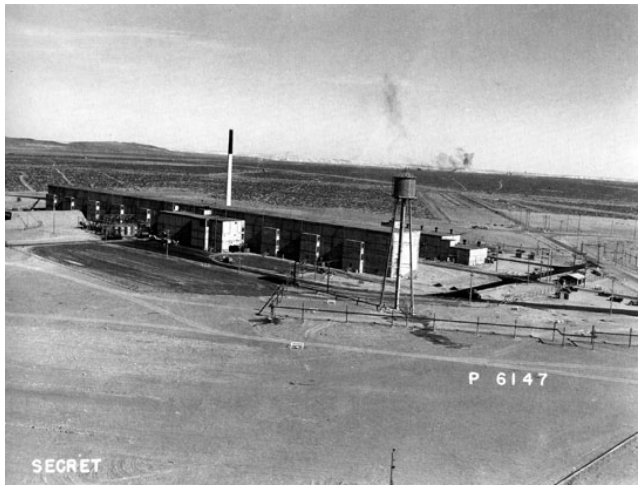
- Because of this low conc., compounds of **Pu could not be precipitated directly**, and any precipitation-separation process had to be based upon **coprecipitation phenomena**, that is the use of so-called **"carriers"** for Pu.

- The radioactive **fission products** had to be separated to less than **one part in 10^7** parts of Pu originally present. This requirement was necessary in order to make it **safe to handle the Pu**.

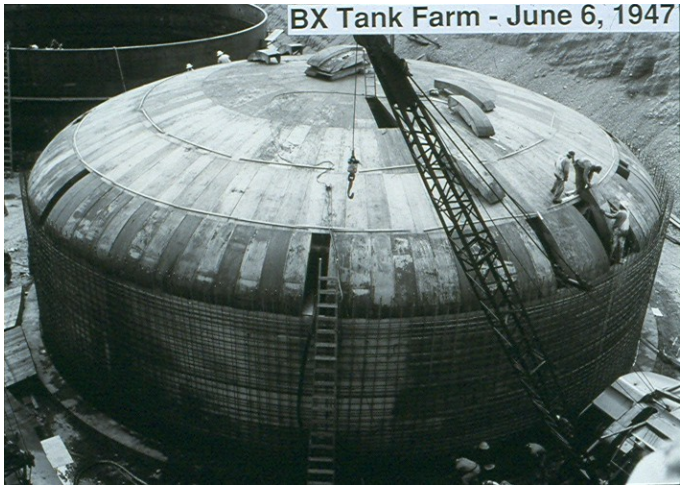
- **Without separation** from the fission products, the Pu from each ton of uranium would have more than **10^5 Ci of energetic gamma** radiation associated with it.

Original Plutonium Separation: Bismuth Phosphate

- Seaborg first separated **microgram** quantities (2.77 µg PuO_2) 1942
- Process scaled to **kilogram** production at Hanford in 1944
- ***Scale-up factor of 10^9 !!!***
- 1 - 1.5 tons of fuel per day, whose Pu content was **~ 300 ppm**, resulting **in 300 to 450 g Pu/day**.
- During the 1940s and 1950s the T and B Plants at Hanford generated an average of **30 cubic meters of waste per metric ton** of spent fuel processed
- **U not recovered**



Hanford T-Plant (1944)



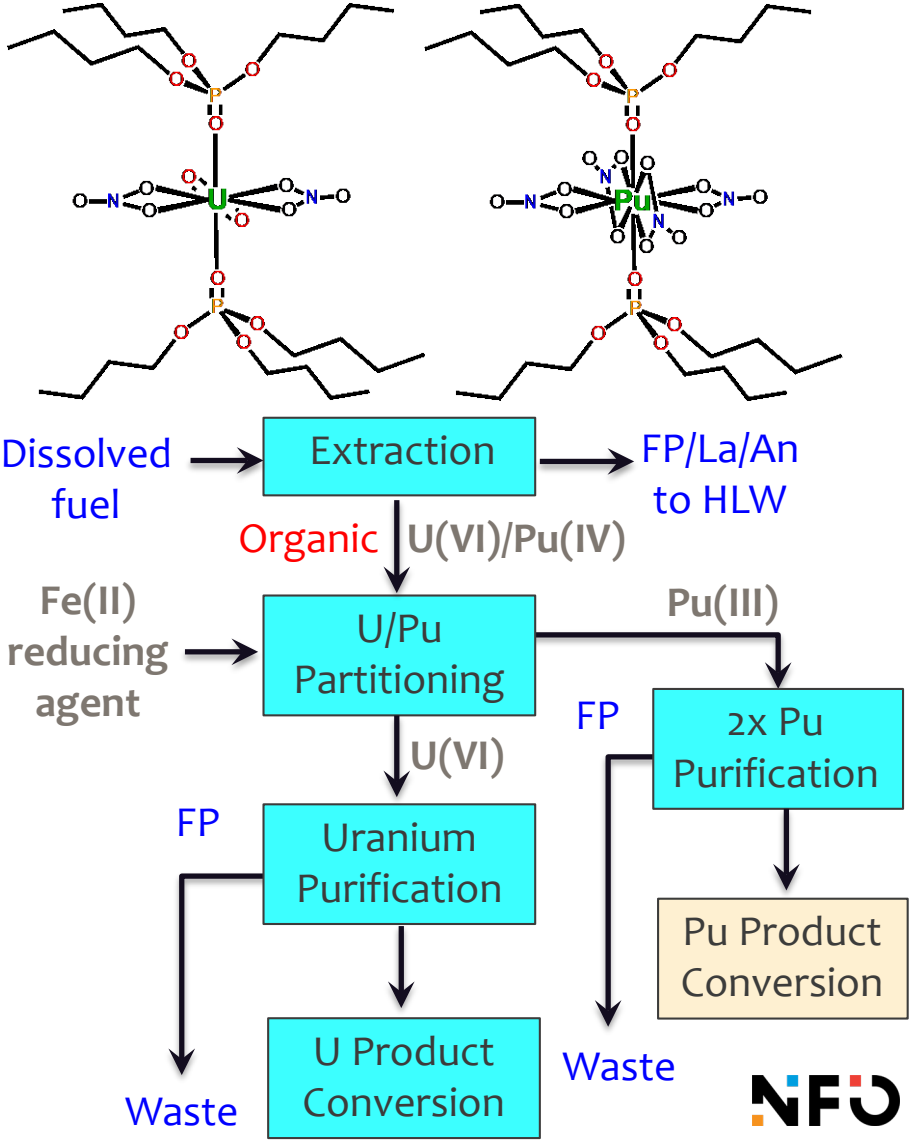
BX Tank Farm- June 6, 1947

Plutonium Was Separated Using PUREX

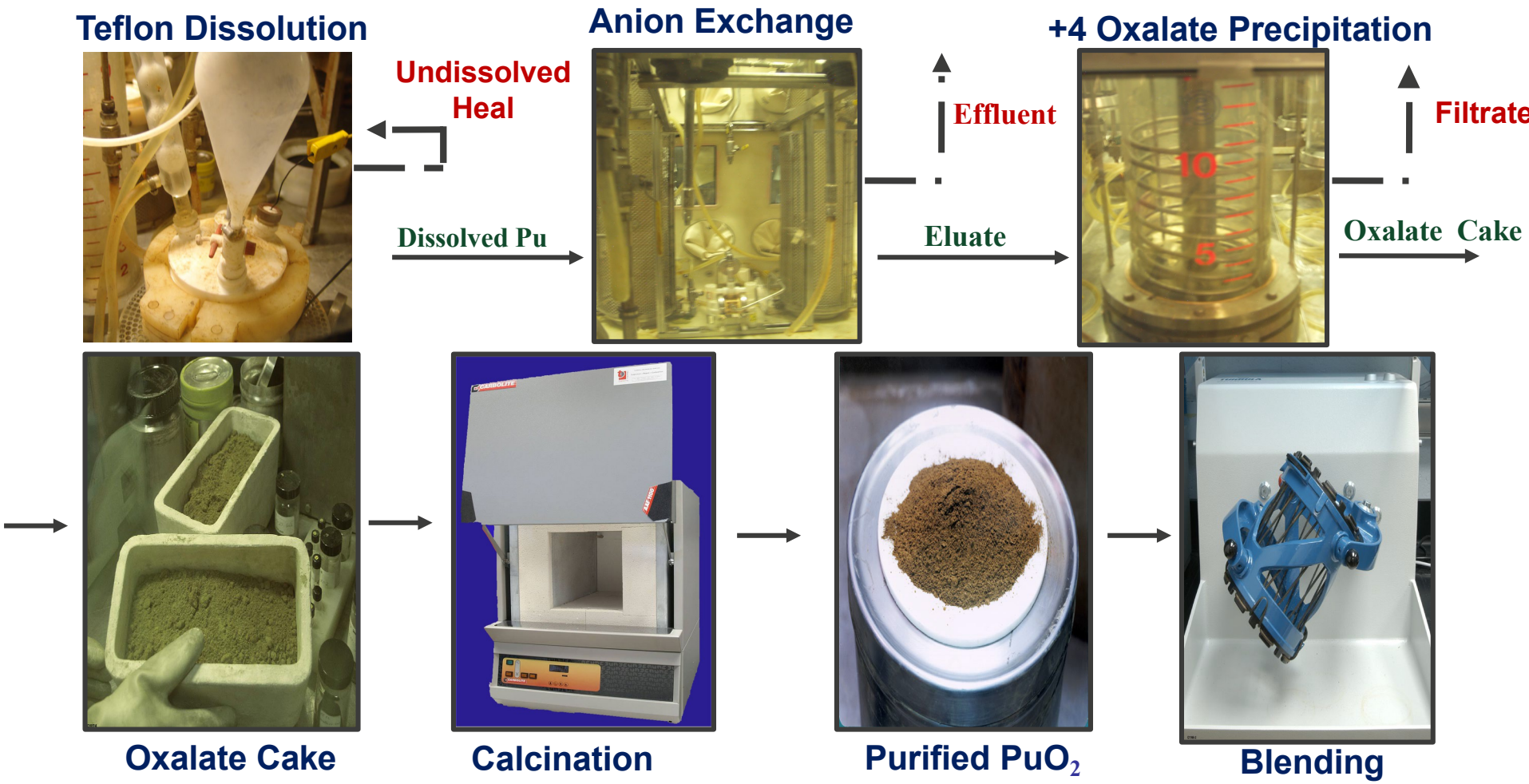
PUREX Plant construction authorized, 1953, operations 1956

- Capacity of 800 MT/U month by 1961
- 30% TBP in kerosene or n-dodecane
  - In high acid, U and Pu are extracted into the organic phase.
  - Conducted in a bank of mixer/settler contactors or in a pulsed column

PUREX construction,1954



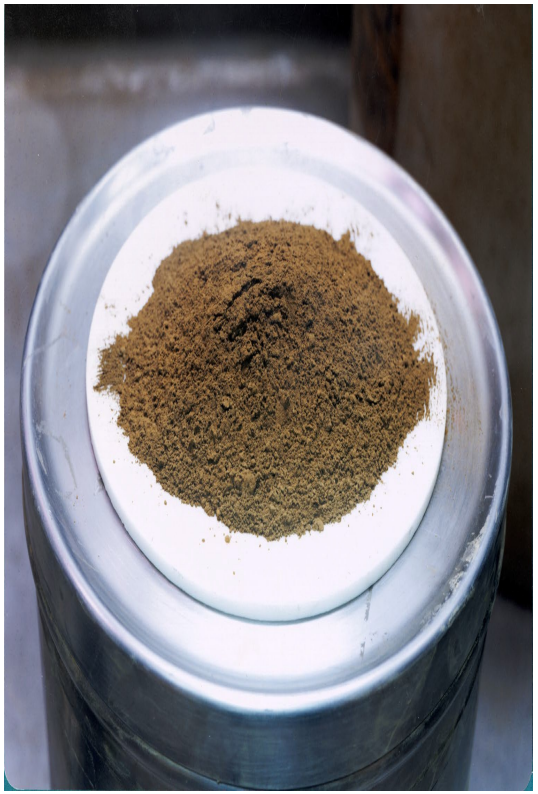
Conversion Chemistry: Aqueous Nitrate Process Flow Diagram



Conversion Chemistry

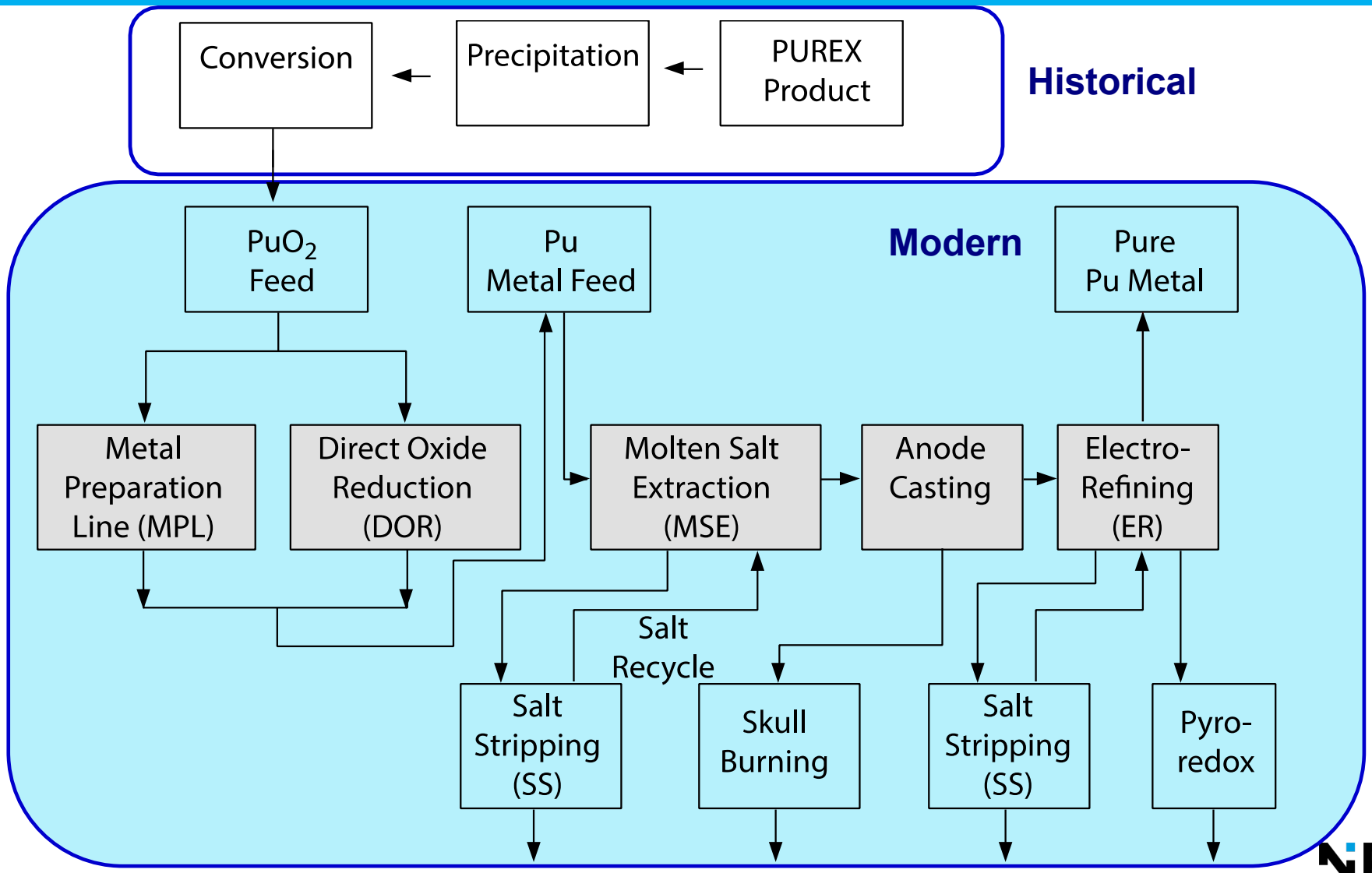
Oxalate Precipitate

Calcination



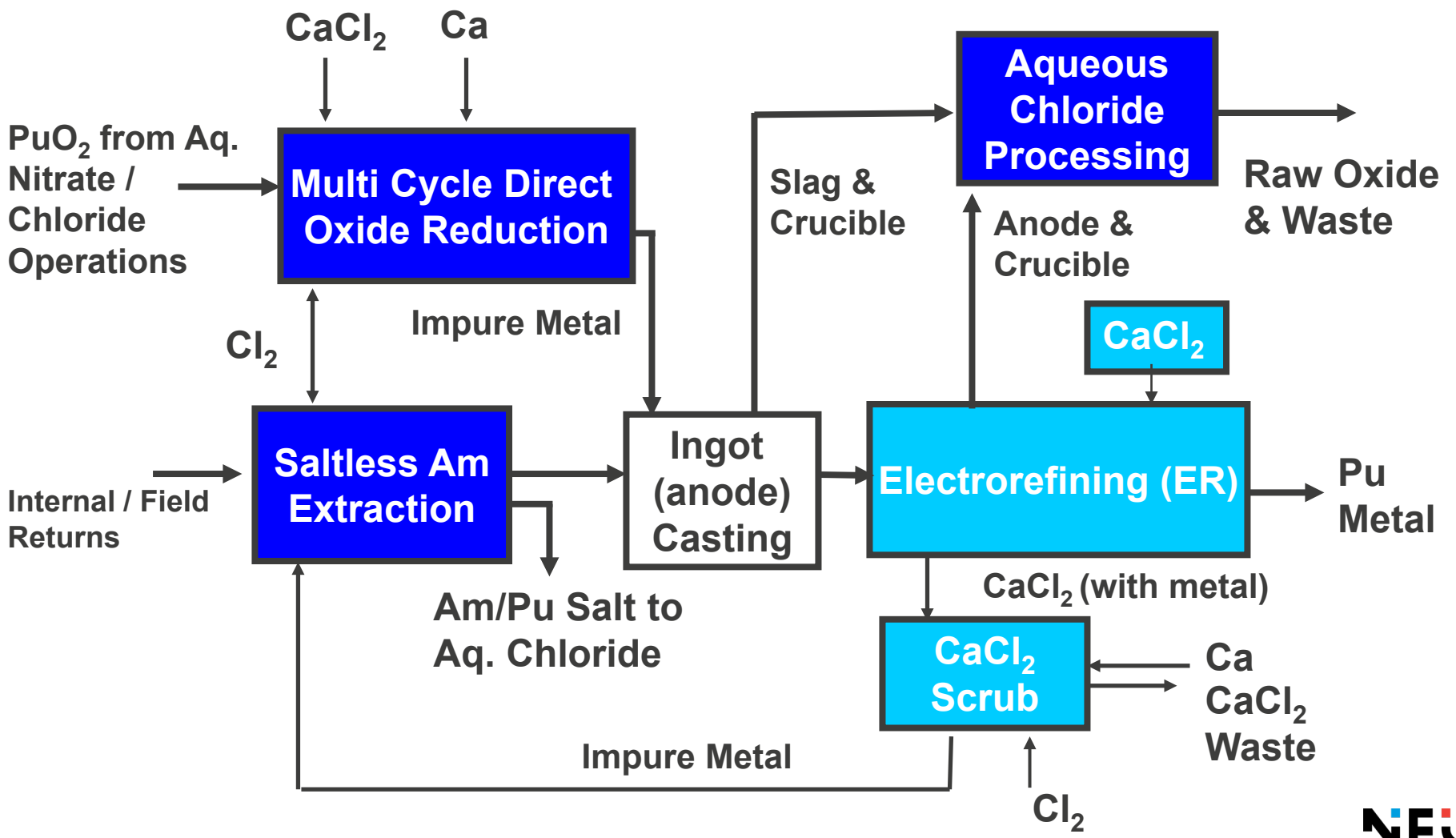
Purified PuO_2

Preparation of Plutonium Metal

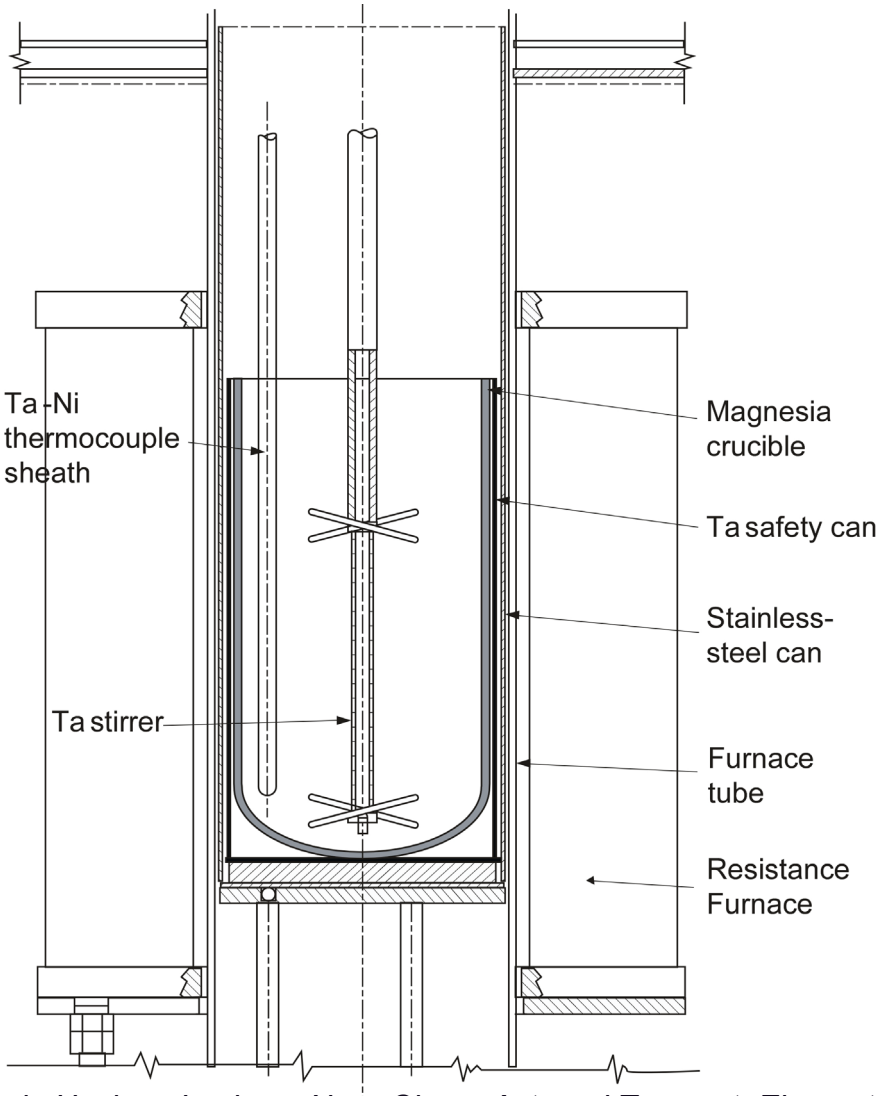


Clark, Hecker, Jarvinen, Neu, *Chem. Act. and Transact. Elements*, Chapter 7, **2006**

Pyrochemical Processing with $CaCl_2$



(Multi-Cycle) Direct Oxide Reduction (DOR)



Metal production

$PuO_2 + 2\ Ca \rightarrow Pu + 2\ CaO$

Salt Regeneration

$2\ CaO + 2\ Cl_2 \rightarrow 2\ CaCl_2 + O_2$

Inverted salt charge and Pu button (top)

Direct Oxide Reduction salt charge and Pu button (bottom)

Clark, Hecker, Jarvinen, Neu, *Chem. Act. and Transact. Elements*, Chapter 7, **2006**

Americium Removal

In-situ Metal Chlorination Molten Salt Extraction (MSE)

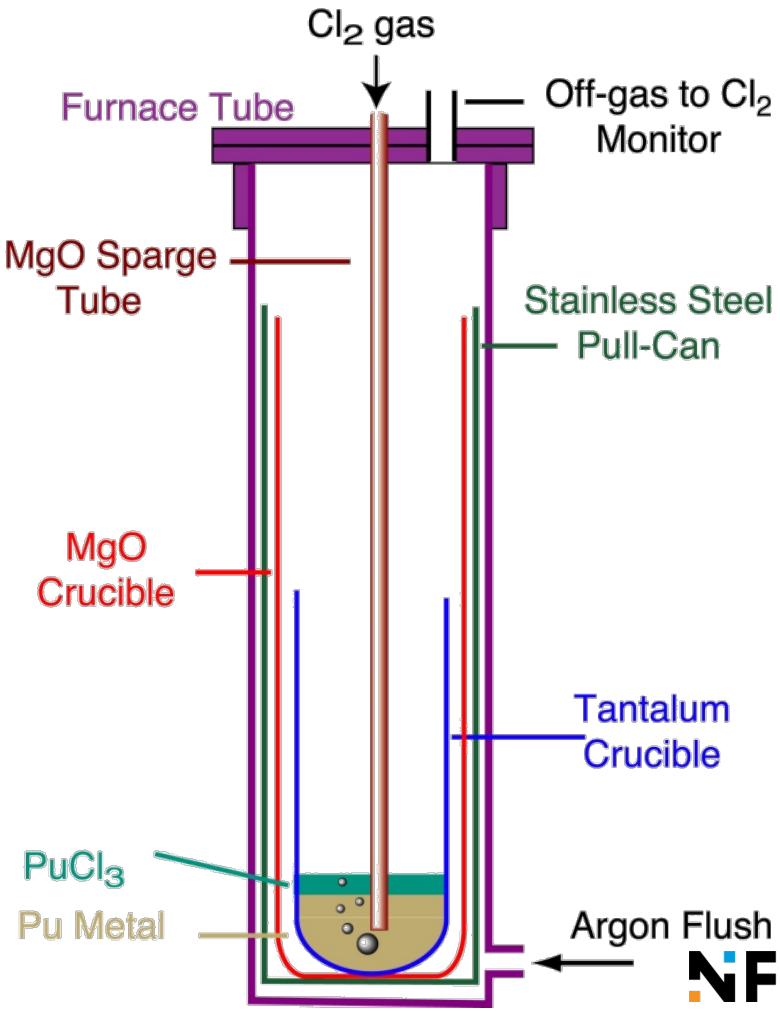
$2\ Pu^0 + 3\ Cl_2 \rightarrow 2\ PuCl_3$

$Am^0 + PuCl_3 \rightarrow AmCl_3 + Pu^0$

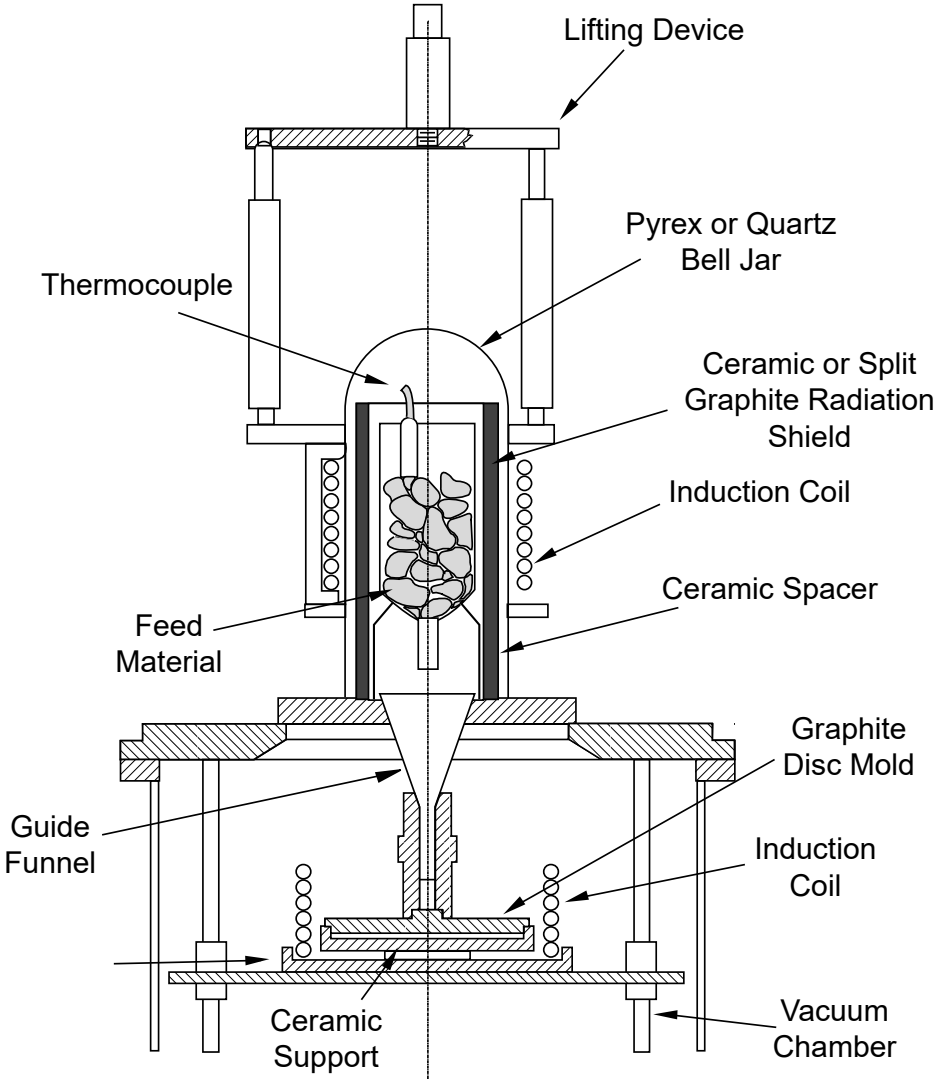
- No cover salt is needed
- Bulk of Am extracted in short time
- 12 hour temp cycle to 750 °C

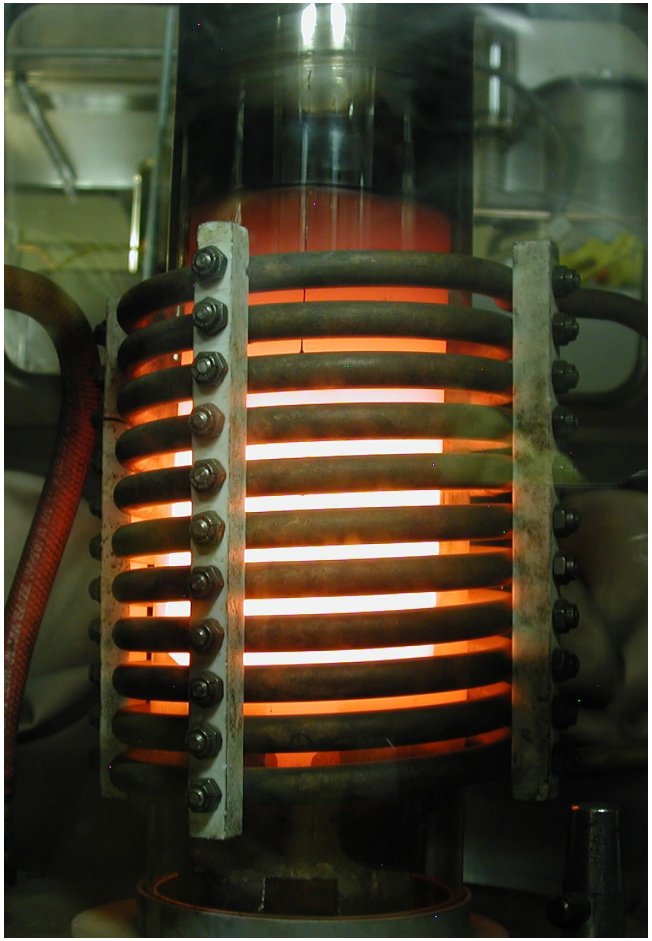
Molten Salt Extraction Button and Residue

MSE Apparatus



Vacuum Melting and Casting

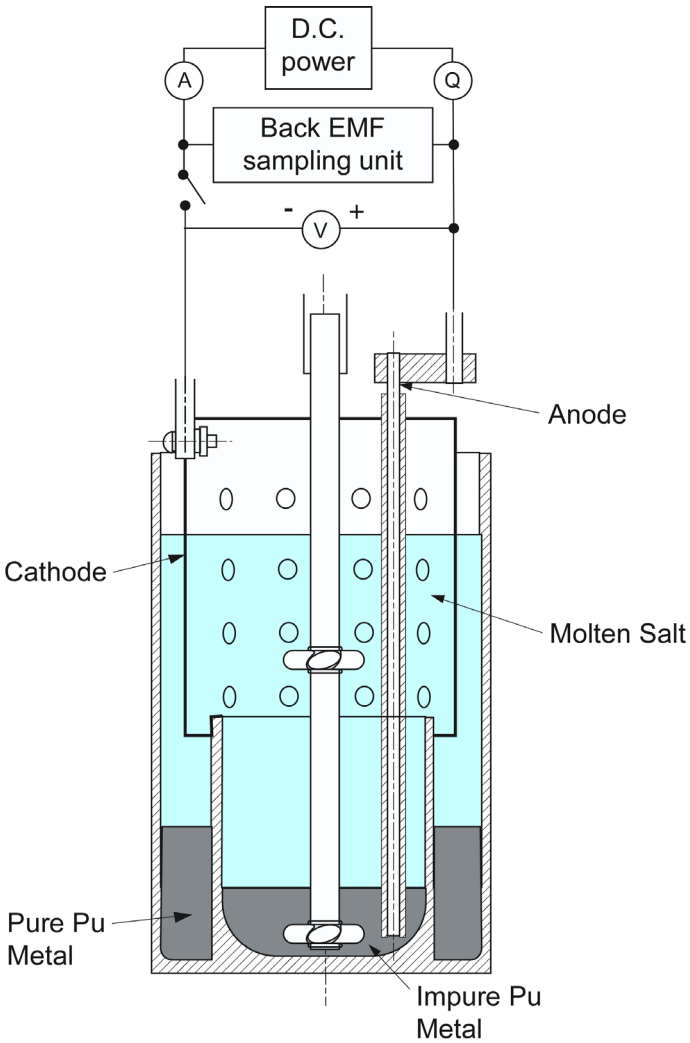




Pu Metal Casting

Clark, Hecker, Jarvinen, Neu, *Chem. Act. and Transact. Elements*, Chapter 7, **2006**

Electrorefining (ER)



Electrochemical Cell

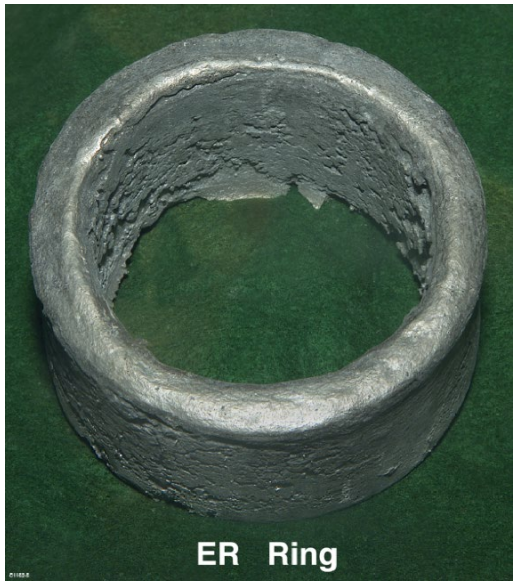
Anode: Pu (impure) $\longrightarrow$ $Pu^{3+} + 3\ e^-$

Cathode: $Pu^{3+} + 3\ e^-$ $\longrightarrow$ Pu (99.9% pure)

Net reaction: Pu (impure) $\longrightarrow$ Pu (pure)

Electrolyte: NaCl-KCl + $PuCl_3$

Pu Ingot



Electrorefined Pu Ring

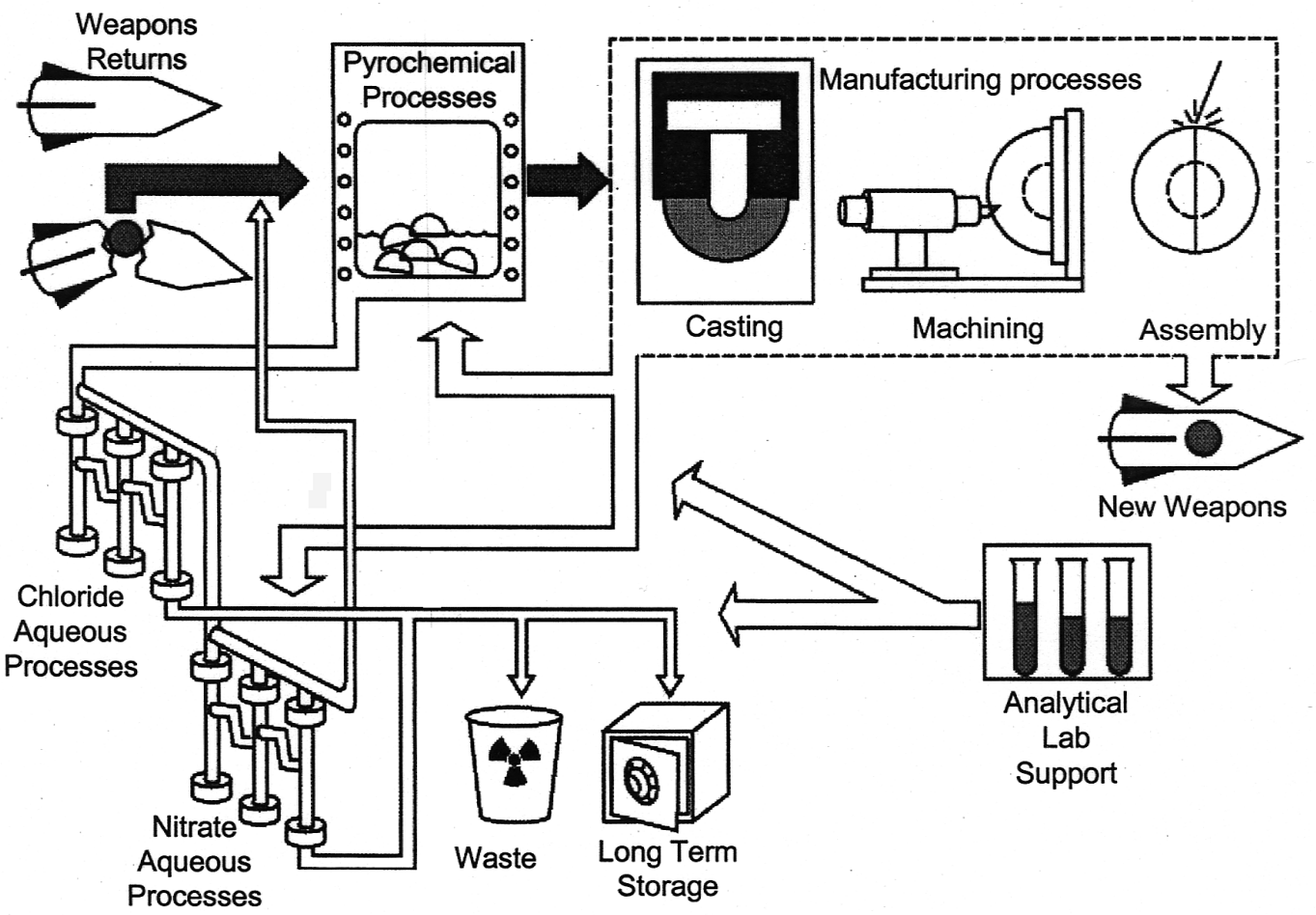
Clark, Hecker, Jarvinen, Neu, *Chem. Act. and Transact. Elements*, Chapter 7, **2006**

Plutonium Processing & Purification

UCNI Video

Plutonium Metal Processing and Reuse

Metal Processing and Reuse Cycle



(K. Staudhammer, 2002)

Unalloyed Plutonium Has Unusual Properties

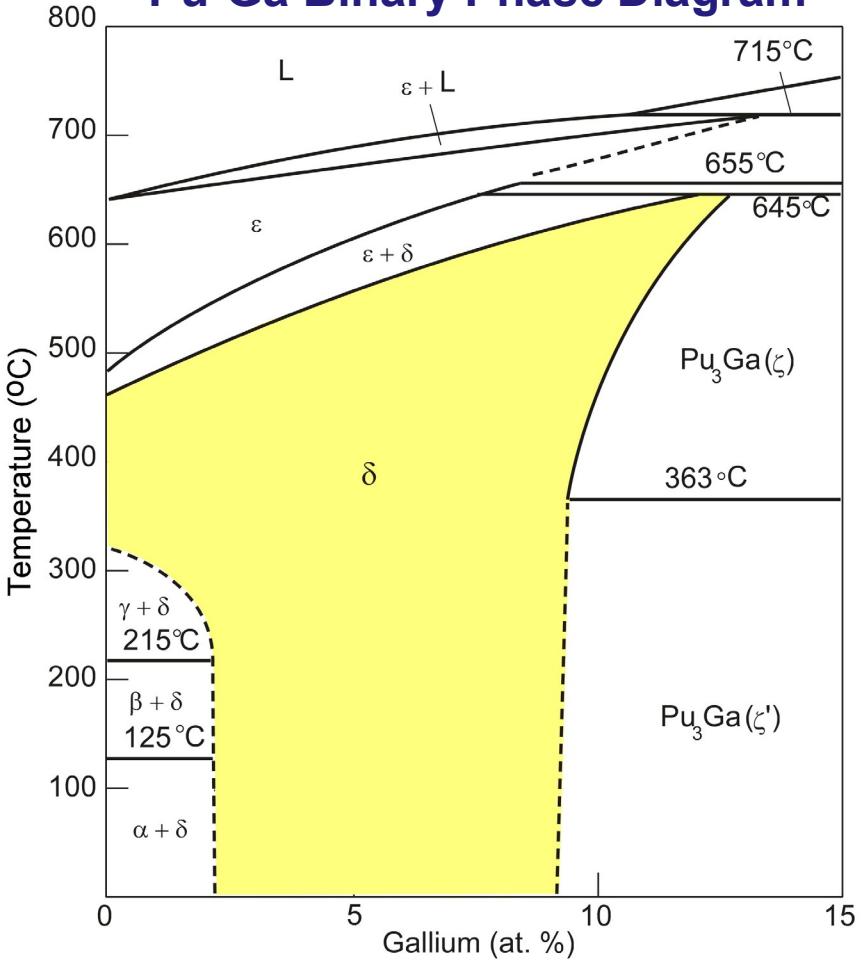
- Unique low-symmetry crystal structures
- Six allotropic phases (seventh under pressure)
- FCC phase is least dense and highly elastically anisotropic
- Dramatic volume changes
- Extreme sensitivity to alloying
- Low melting point
- Low cohesive energy
- Large specific heats
- Volume decrease upon melting
- Anomalies in low-temperature transport properties
- Dramatic variation in mechanical properties
- Very high self-diffusion in bcc epsilon phase
- Highly unusual properties of the liquid phase
- Great affinity for oxygen and hydrogen
- Very large thermal expansion coefficients
- Negative thermal expansion in FCC phase
- Self-irradiation damage because of radioactive decay

(S. Hecker, 2005)

Alloying With gallium stabilizes the face-centered cubic δδ-phase which has many attractive properties.

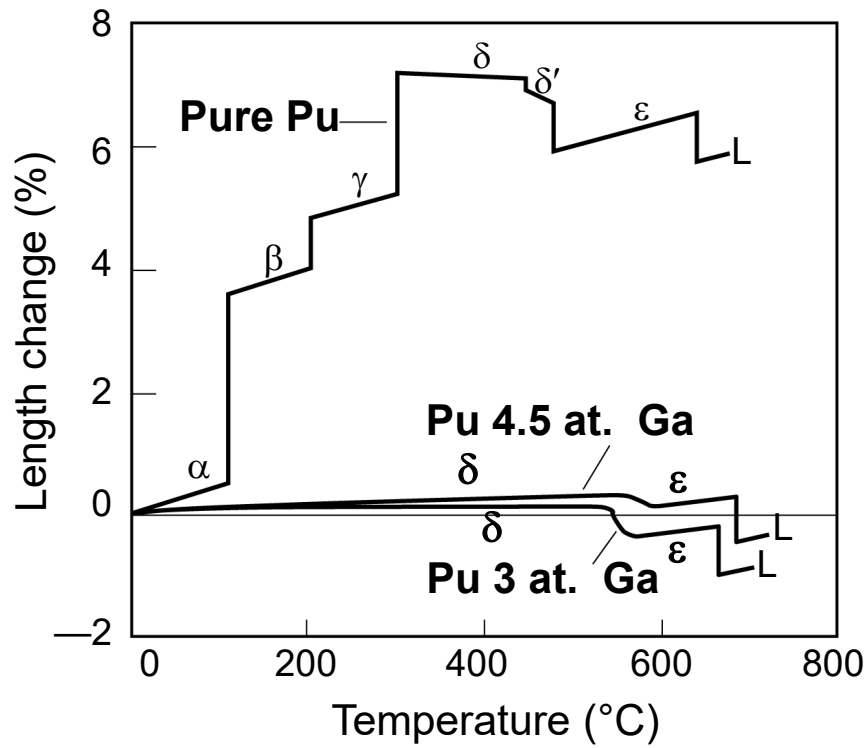
Clark, Hecker, Jarvinen, Neu, *Chem. Act. and Transact. Elements*, Chapter 7, **2006**

Pu-Ga Binary Phase Diagram



F. H. Ellinger, *J. Nucl. Mater*, **1964**, *12*, 226.

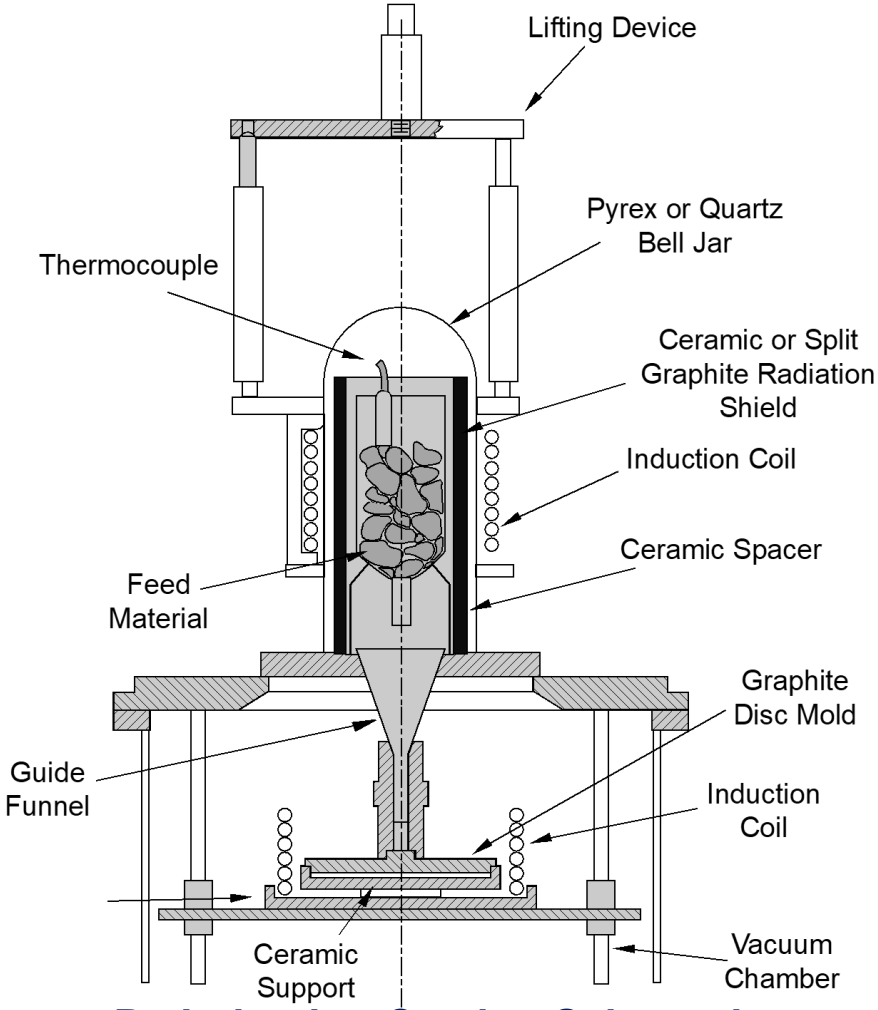
Plutonium Thermal Expansion



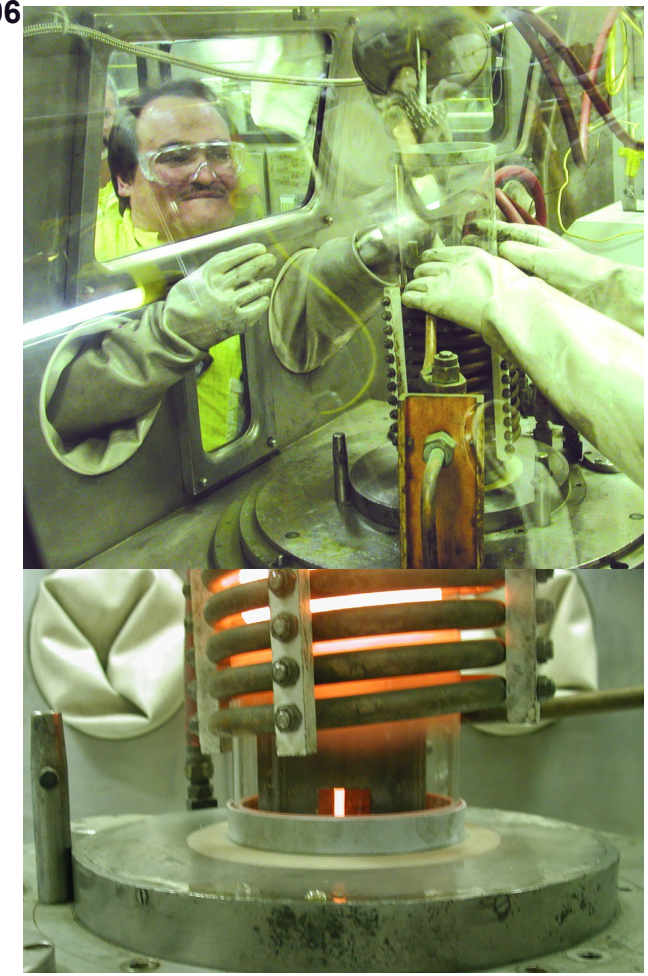
Hecker, Harbur, Zocco, Prog. Mater. Sci. **2004**, 49(3-4), 4295

Plutonium Components Are Formed By Induction melting and casting into shaped molds.

Clark, Hecker, Jarvinen, Neu, *Chem. Act. and Transact. Elements*, Chapter 7, **2006**



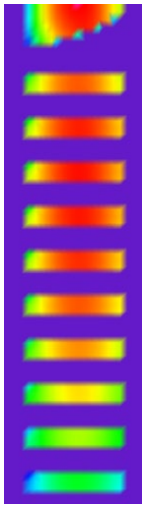
Pu Induction Casting Schematic



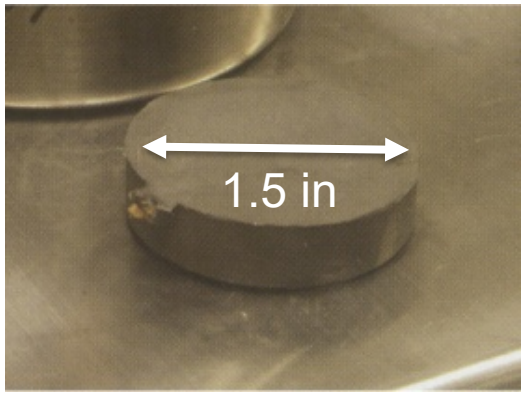
Pu Casting Preparation and Molten Pu Pouring

Popular shapes for Pu ingots include plates, pucks and rods as feedstock for experimental samples.

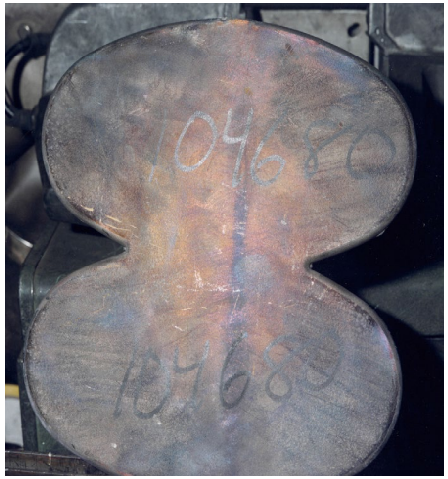
Modeling mold thermal response to molten Pu metal is important to predicting microstructural characteristics and properties. (Cooling rate determines grain size.)



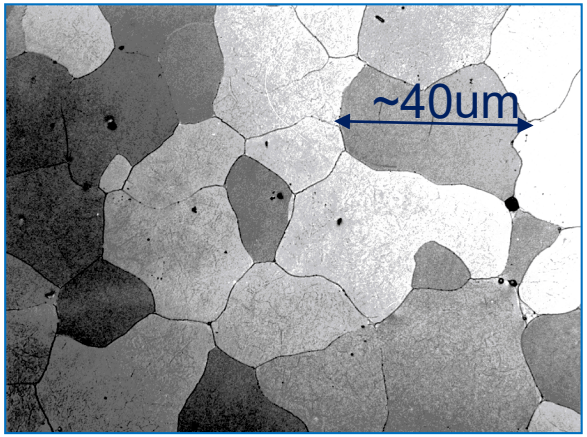
Puck Mold Model



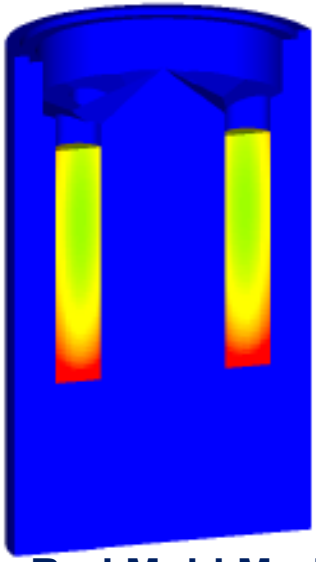
Cast Pu Puck



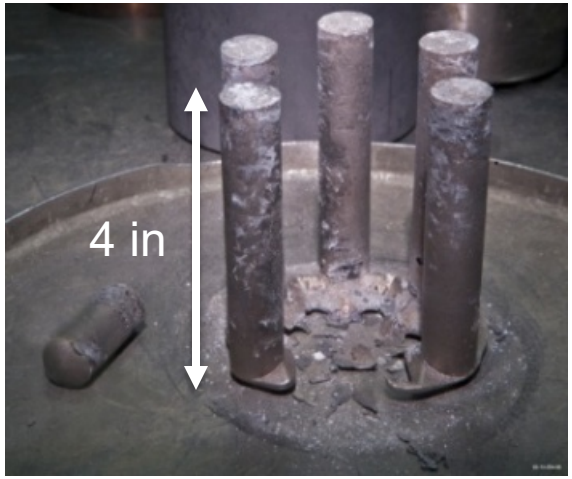
Cast Pu Ingot



Grain Structure (F. Freibert, 2010)

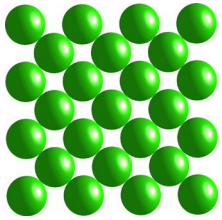
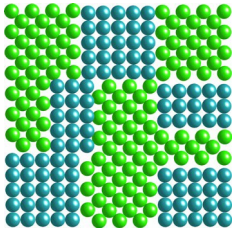
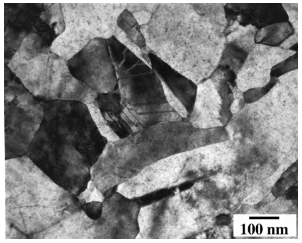


Rod Mold Model



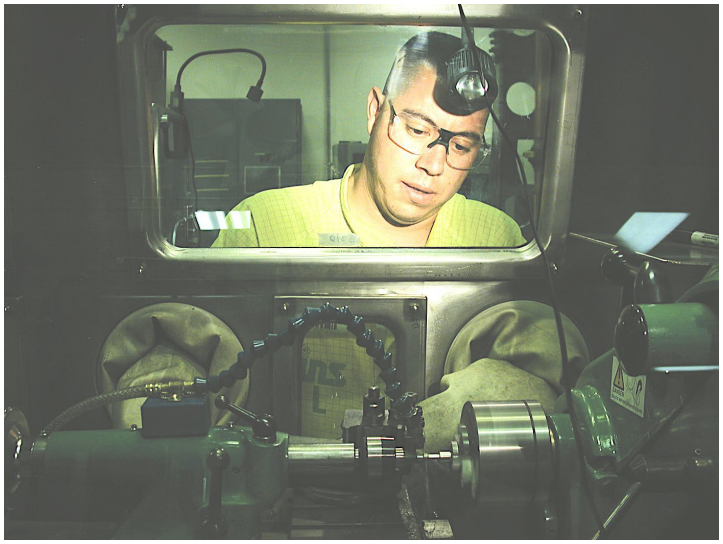
Cast Pu Rods

Length scales from atomic to macroscopic correlate with important properties.

Atomic scale ~ 10 Å	Nanoscale ~ 100Å	Microscale ~ 100μ	Macroscale ~ 100cm
			
atomic structure	nano structure	bulk properties	component processes
Fundamental structure & properties of homogeneous materials	Bulk properties of average structures, and heterogeneous materials	Controlling effects of heterogeneities, process coupling (chem, phys, mech)	
- Ga distribution - impurity effects - lattice parameter - local & micro-structure - defect structure - self-irradiation damage - phase stability - Ga, Pu diffusion	- lattice damage - void swelling / He ingrowth - He bubble formation - Ga distribution (coring) - elastic properties - density changes - grain size - dislocation structure	- mechanical behavior - dynamic properties - stress-strain response - strength, embrittlement - weld stability - corrosion behavior - spall strength - dimensional stability	

(D. Clark, 2002)

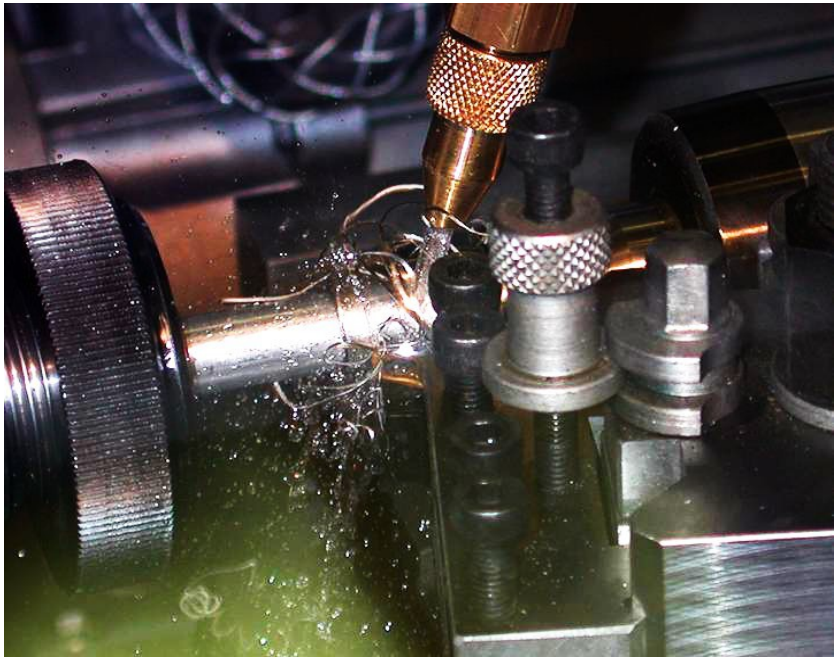
Machining and forming plutonium is required to achieve finished state.



Machining Pu

Rolling Pu

Important in lathe machining is depth of cut, cooling and chip formation.



Machining Pu and Turnings

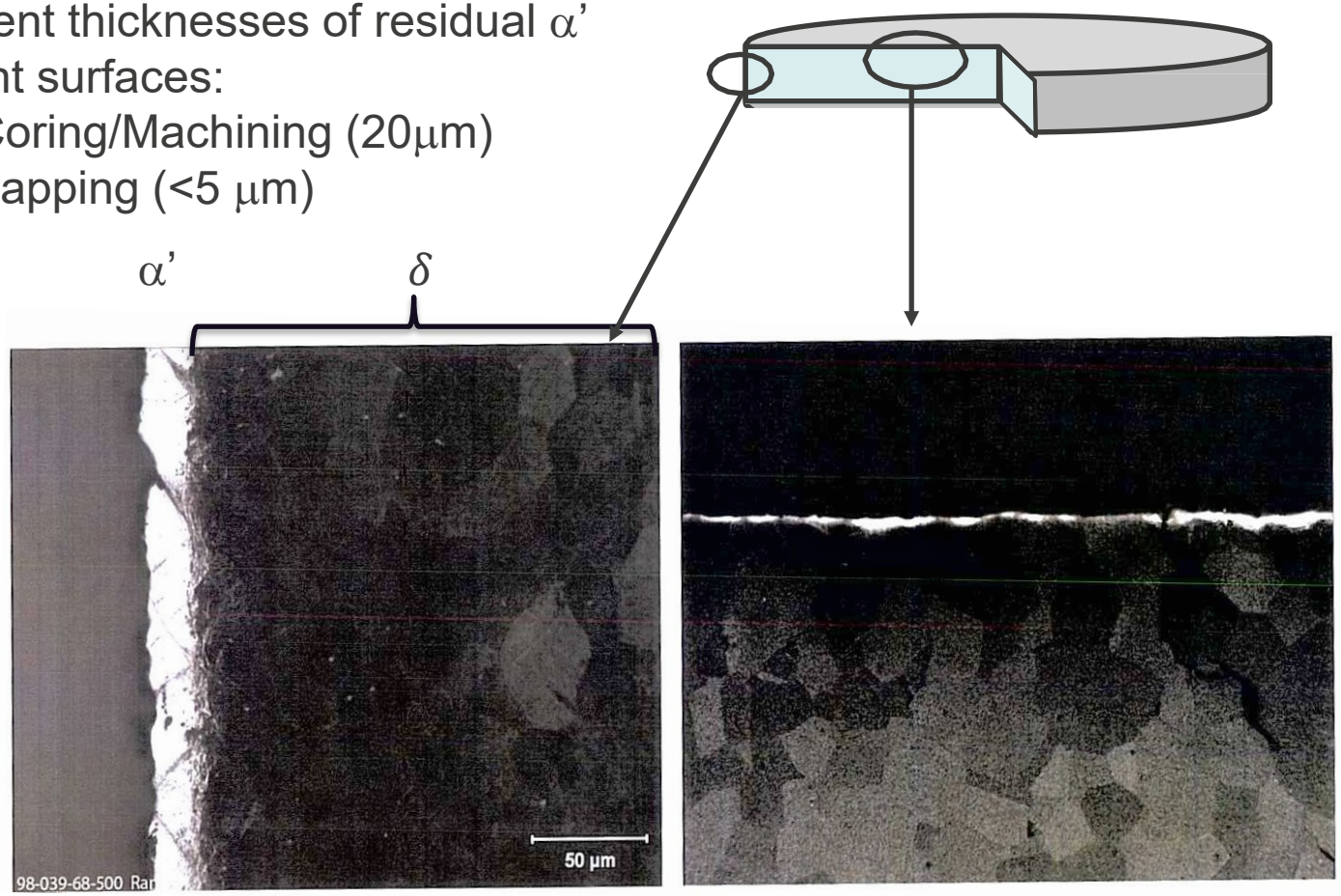
Other materials forming operations may include rolling and hydroforming.

(F. Freibert, 2002)

Heavy cuts during machining operations drive underlying microstructure unstable to mechanically induced phase transformations.

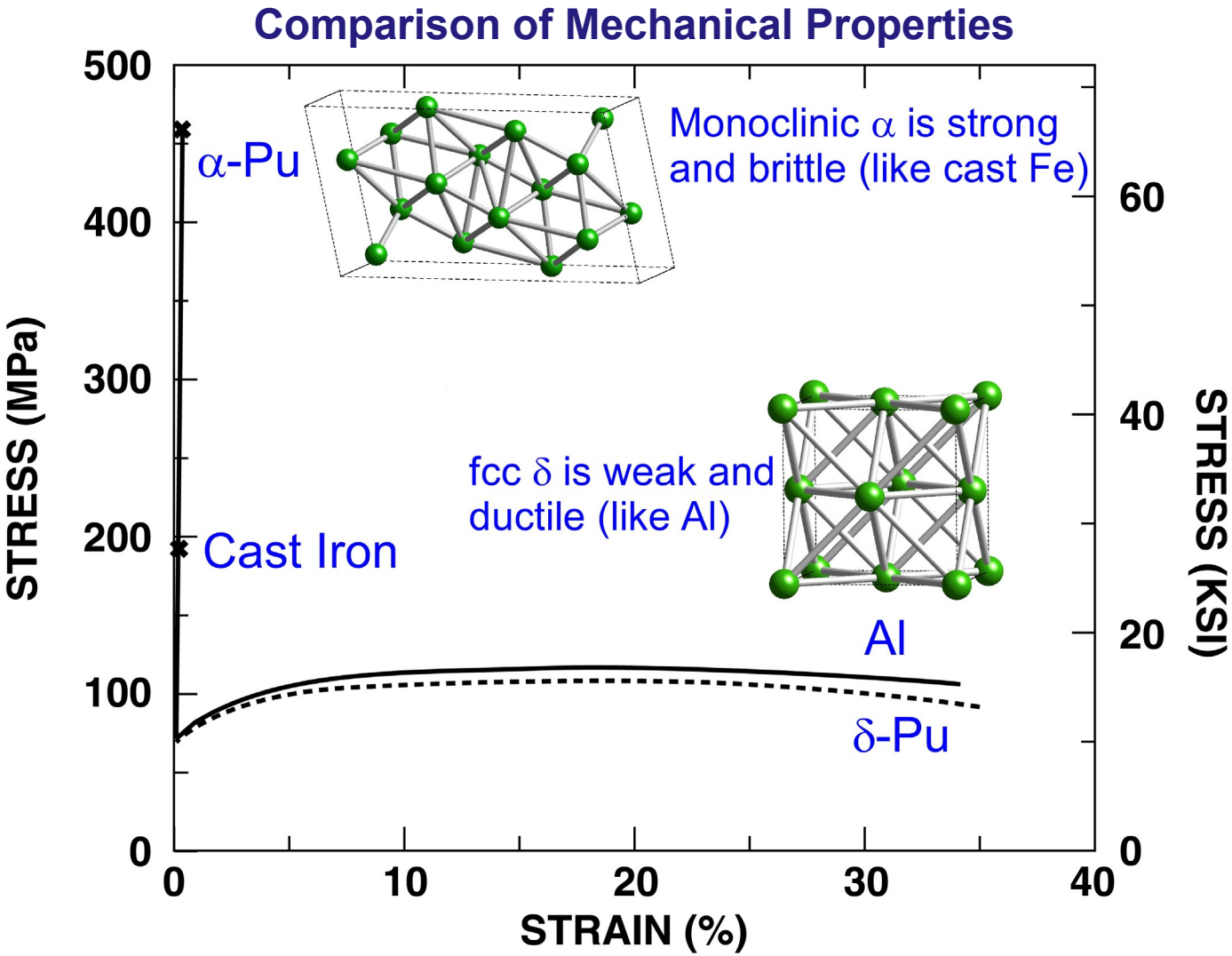
Machining producing gas gun samples with different thicknesses of residual α' on different surfaces:

- Edge Coring/Machining (20μm)
- Hand Lapping (<5 μm)

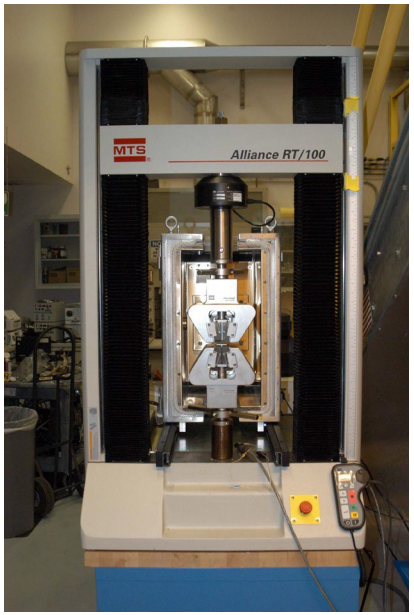


Microscopic cross-section of machined Pu showing α' layer on machined δ metal parts.

Crystallographic phases ($\alpha\alpha$-Pu vs. $\delta\delta$-Pu) show different mechanical properties.



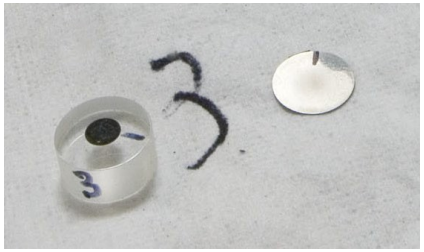
Mechanical Test Frame



Assembly of components, subcritical experiments or Pu Z Machine test assemblies requires similar skills and competencies.

Combining precisely fabricated components into assemblies with exacting tolerances is a challenging role in ensuring the US nuclear deterrent.

Subcritical Assembly (2015)



Pu Sample and Window for Z Machine

Z Machine Sample Panel Assembly

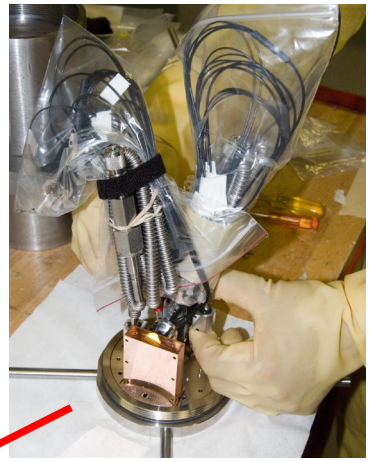
Z Machine Sample Assembly Readied for Shipping

Z Machine Sample Panels

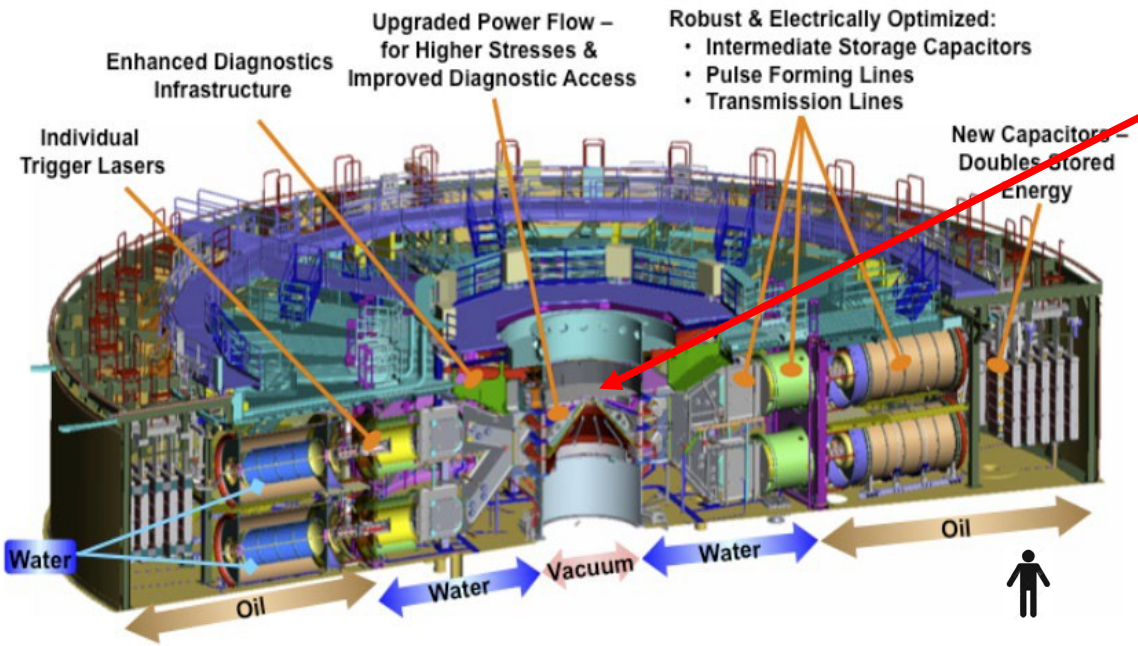
We no longer perform nuclear explosive testing, so we conduct experiments (i.e., Pu compression) for Stockpile Stewardship .

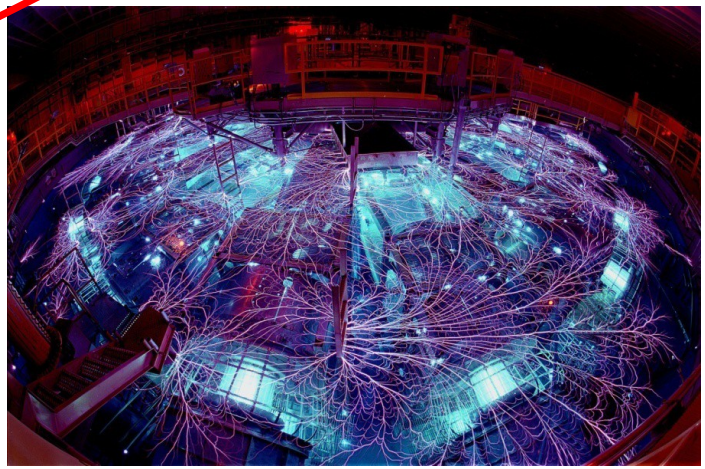
Trinity Pu Compression Test (1945)

The SNL Z Machine is 33 m in diameter and 3 stories high and combines 25 mega-amps into magnetic compression of Pu in less than 1 microsecond. These experiments provide important data for use in weapons physics.



Z Machine Pu Sample Assembly





Z Machine LANL/SNL Pu Compression Test (2015)

Massive amounts of high level radioactive wastes (HLW) were generated from plutonium

- U.S. stored high level radioactive wastes (**HLW**) in shielded, underground tanks
- SRS and Hanford tanks contain 575 million curies in 91 million gallons of sludge, liquid and solid waste stored in 226 underground tanks.
- The costs for managing the tank farms are enormous with about $1 million per day for tanks at SRS..
- Estimates for life-cycle costs reach nearly $250 billion with completion of the cleanup of SRS and Hanford tank farms by 2060-2070.

*Environmental management technical expert group (EM-TEG). EM tank waste strategy review, Los Alamos National Laboratory, Report LA-UR-11-02972, (**2011**)*

Hanford S Tank Farm

SRS H Tank Farm

Tank Waste

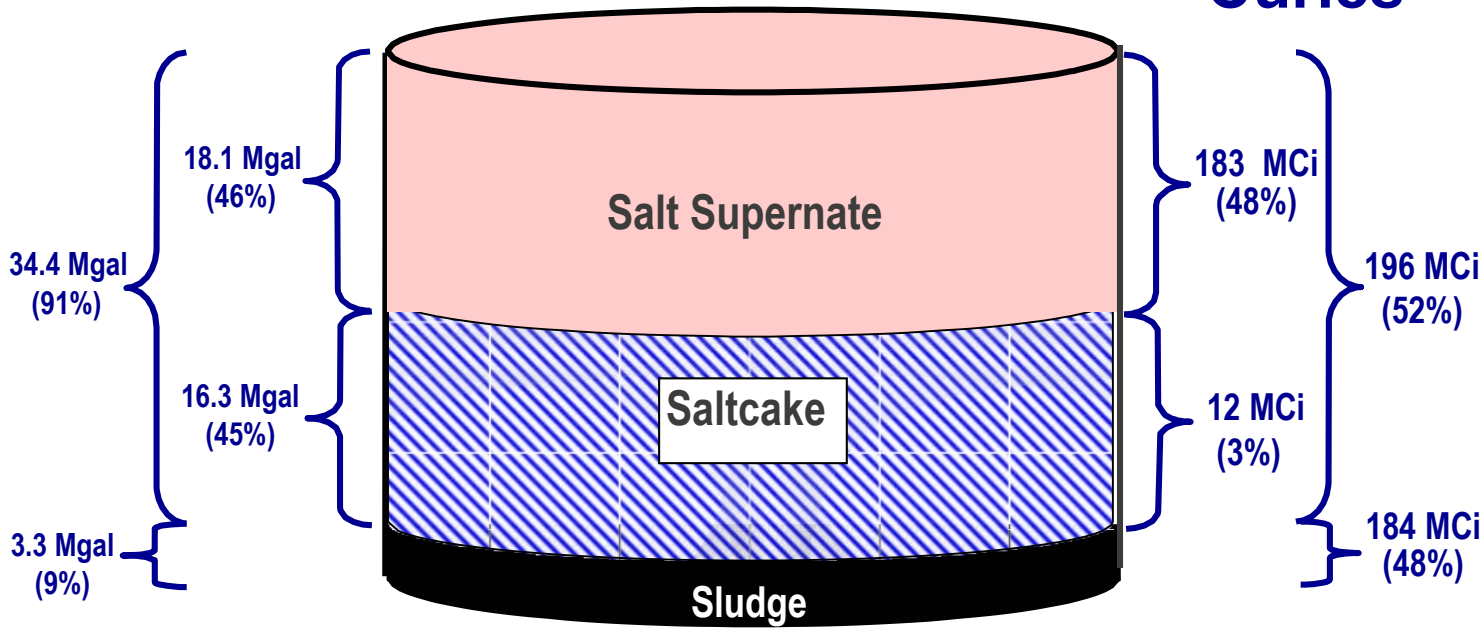
SRS F-Area Tank Farm

Tank Waste Characteristics

Example: SRS Inventory

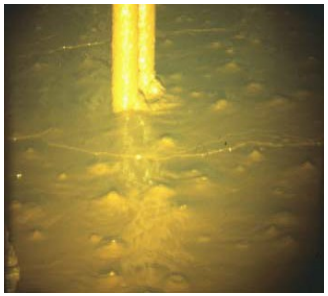
Volume

Curies



Salt Supernate

Saltcake



Sludge

37.7 Million Gallons (Mgal)

Inventory values as of 2008-12-31

National Research Council, **2006**, ISBN: 0-309-65955-8
EM-TEG Report LA-UR-11-02972, (**2011**)

Defense Waste Processing Facility (DWPF)

Savannah River Site – the DWPF receives waste for processing from H Tank Farm. The waste is “vitrified” and poured into stainless steel canisters that are sealed and decontaminated.

DWPF delivered its 4000th cannister in 2016.

DWFP Melter

CAUTION:
MOVE STATION WITH CARE
KEEP HANDS CLEAR OF NEARBY OBJECTS
CDC CONTROL
STATION # 4

DWPF Canisters

Opened the Waste Isolation Pilot Plant (WIPP) For Transuranic (TRU) Waste

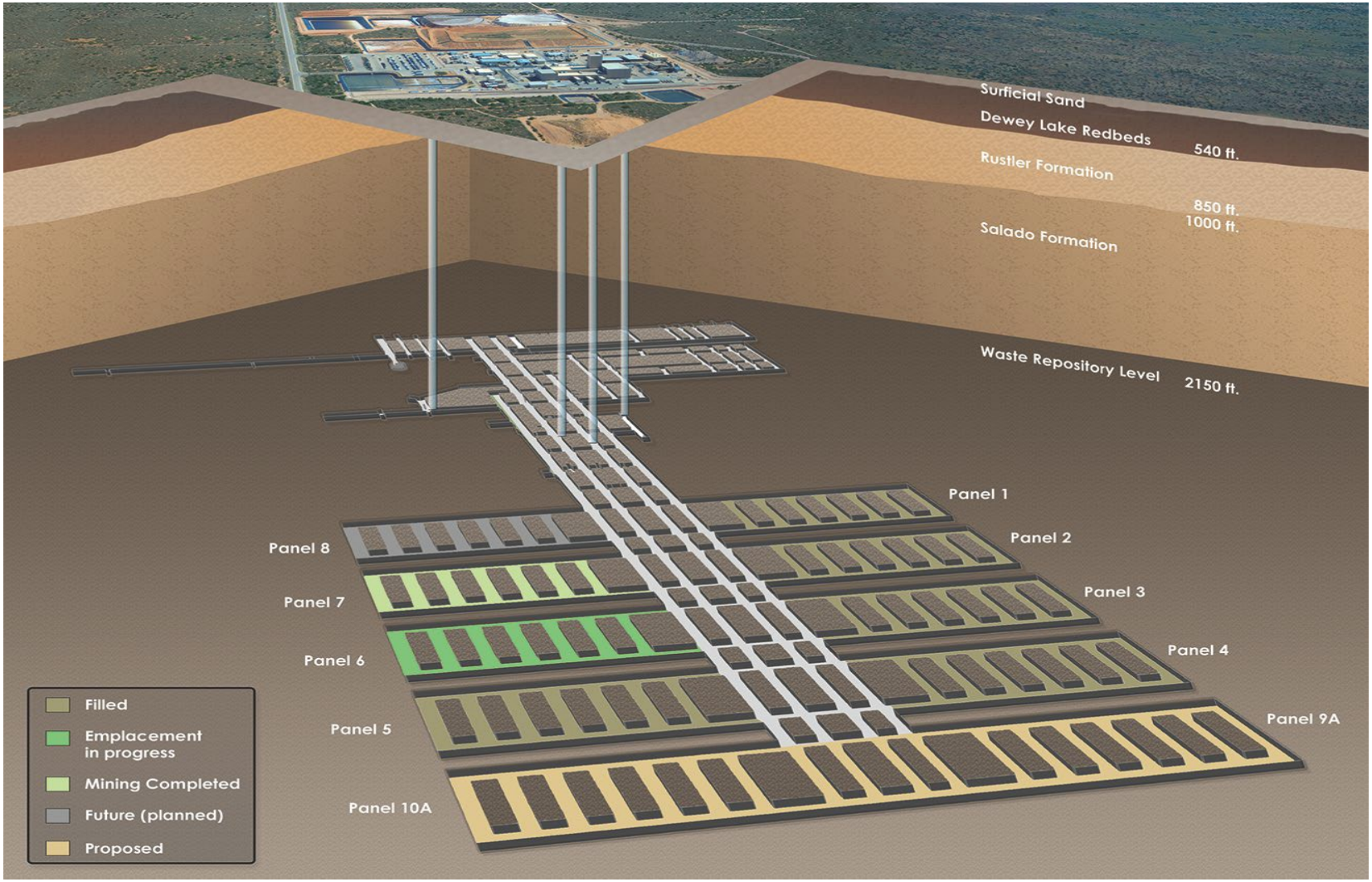
- Deep geological repository – operational in 1999
- 16 mi^2 near Carlsbad, New Mexico
- Disposal rooms mined 2,150 feet underground in a salt formation
- 20 year effort to make scientific case
- Licensed to permanently dispose of transuranic radioactive waste for 10,000 years
- >12,500 shipments, 96,300 m^3 (178,500 containers) waste received by July 2019
- Estimated to continue accepting waste for 25 - 35 years for total cost of $27 billion (2016 dollars)

Physics Today 52.5 (1999)

WIPP Aerial View

TRU Waste in WIPP Disposal Room

Waste Isolation Pilot Plant



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or LANL "Plutonium Science Resources" external website: http://lanl.libguides.com/nuclear

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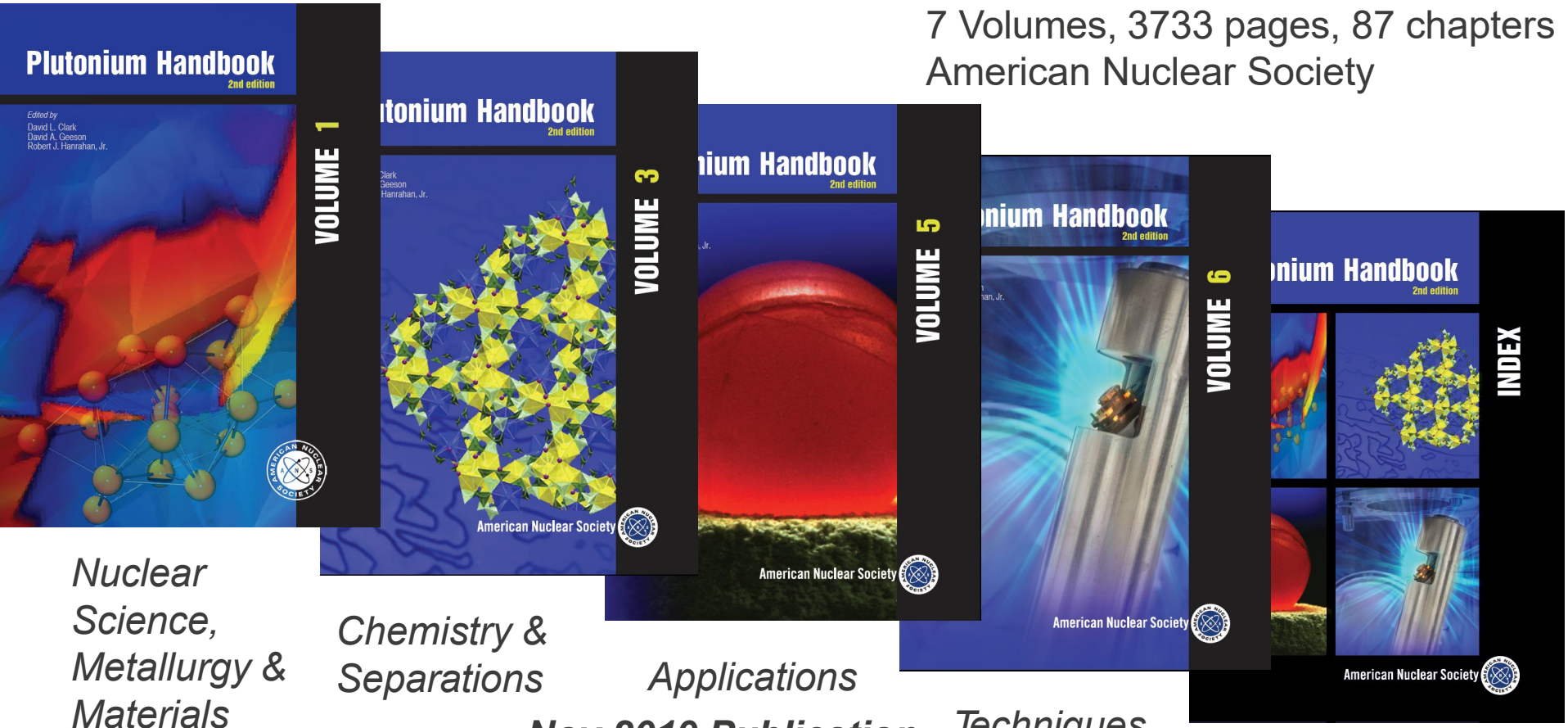
David L. Clark, David A. Geeson, Robert J. Hanrahan, Jr., Editors

7 Volumes, 3733 pages, 87 chapters
American Nuclear Society



Nuclear Science, Metallurgy & Materials

Chemistry & Separations

Applications

Nov 2019 Publication

Techniques

Future briefings will be provided on other materials.

Thank you!

Questions?

Email us: **NFO@lanl.gov**