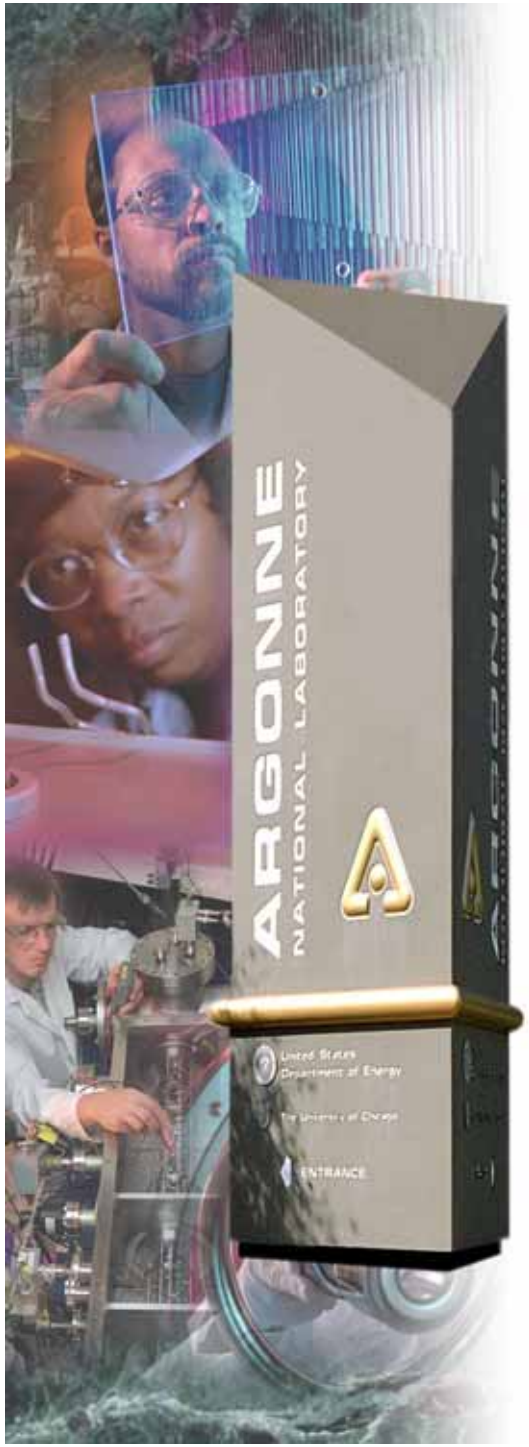

Radioactive Waste Management

Robert Finch

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Argonne National Laboratory

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Enabling our Energy Security through the Nuclear Fuel Cycle
July 20-22, 2004

Argonne National Laboratory

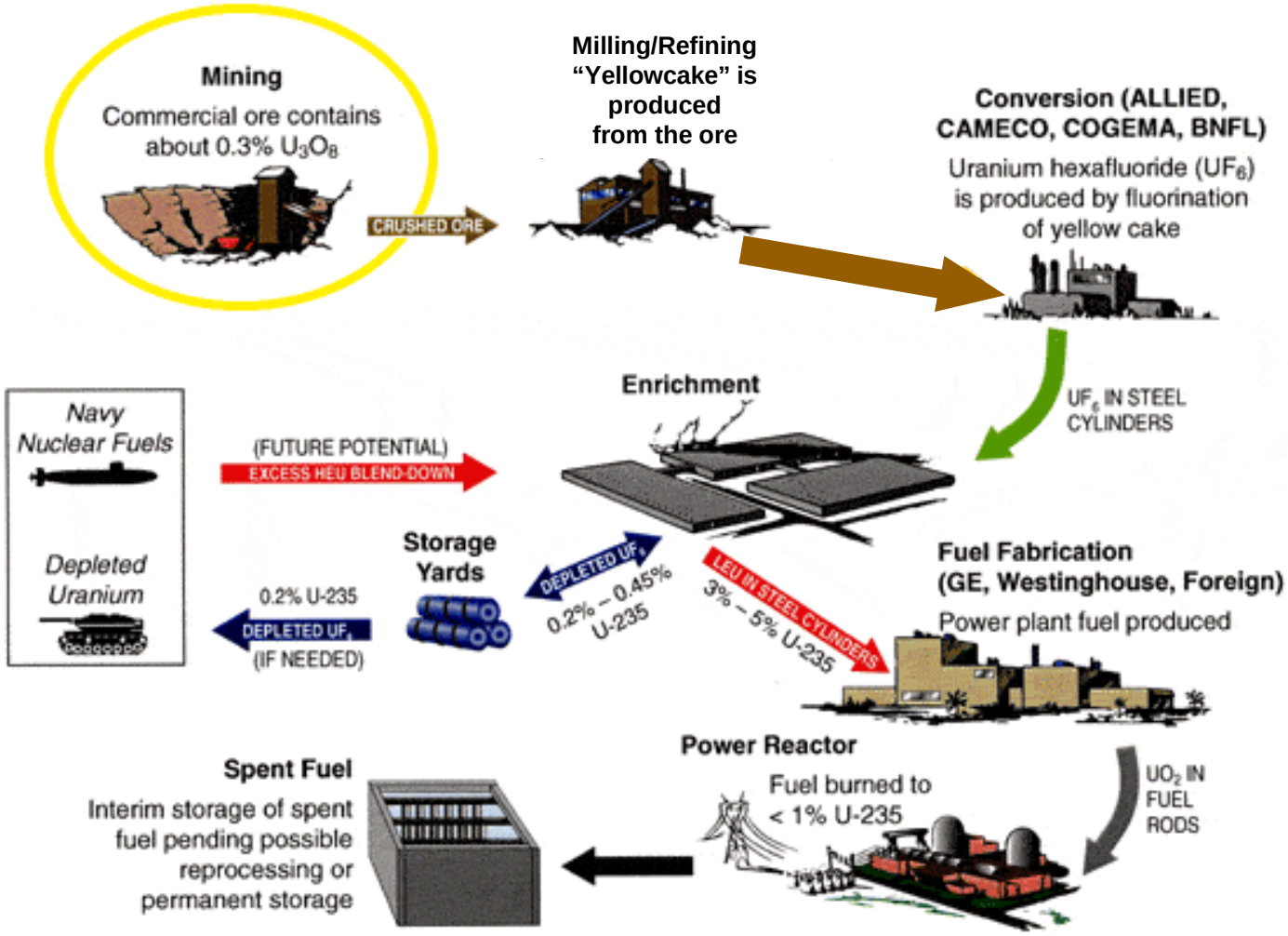
Office of Science
U.S. Department of Energy

A U.S. Department of Energy
Office of Science Laboratory
Operated by The University of Chicago

Classification of Radioactive Waste

- **High Level Waste**
  - Spent Nuclear Fuel
    - *Commercial (light-water reactors)*
    - *DOE owned (wide variety)*
  - Waste derived from spent fuel (reprocessing wastes)
- **Low Level Waste**
  - *Not* from spent fuel (e.g., medical, contaminated materials)
    - *Several subcategories (activity based)*
- **Transuranic (TRU) Waste**
  - Z > 92; t-1/2 > 20 years; > 100 nanocuries per gram
  - Excess Weapons Grade Plutonium
- **Uranium Mine & Mill Tailings**

Fuel Cycle

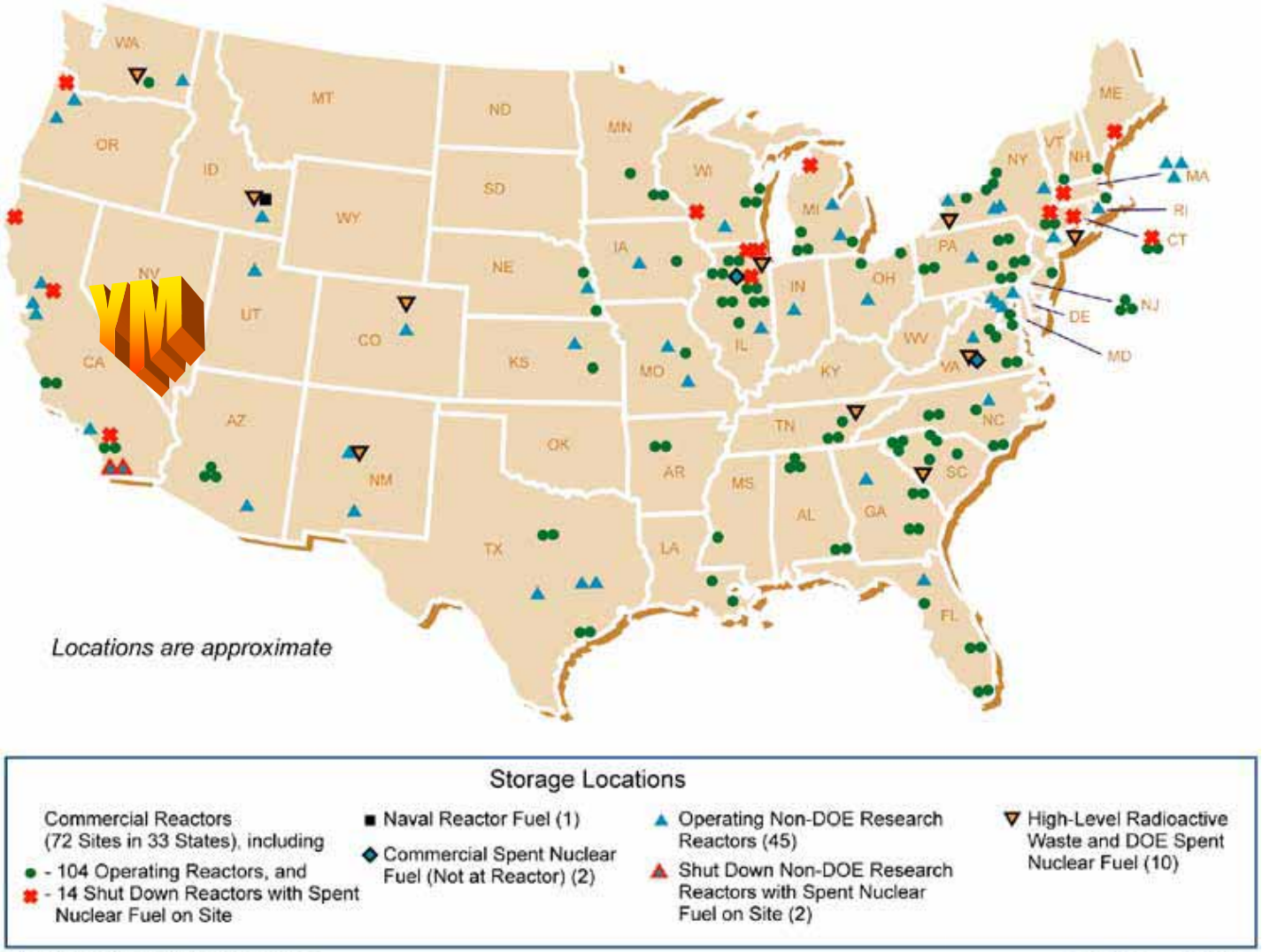

Mining
Commercial ore contains about 0.3% U_3O_8
CRUSHED ORE
Milling/Refining "Yellowcake" is produced from the ore
Conversion (ALLIED, CAMECO, COGEMA, BNFL)
Uranium hexafluoride (UF_6) is produced by fluorination of yellow cake
UF_6 IN STEEL CYLINDERS
Enrichment
Navy Nuclear Fuels
Depleted Uranium
(FUTURE POTENTIAL)
EXCESS HEU BLEND-DOWN
Storage Yards
DEPLETED UF_6
0.2% – 0.45% U-235
LEU IN STEEL CYLINDERS
3% – 5% U-235
0.2% U-235
DEPLETED UF_6
(IF NEEDED)
Fuel Fabrication (GE, Westinghouse, Foreign)
Power plant fuel produced
Power Reactor
Fuel burned to < 1% U-235
UO_2 IN FUEL RODS
Spent Fuel
Interim storage of spent fuel pending possible reprocessing or permanent storage

Effective Management of Radioactive Wastes

- **Public Acceptance & the Future of Nuclear Power**
  - Allay public fears that N-waste is an intractable problem
  - Sociopolitical & scientific issues for future repositories
- **National Security**
  - Safe & secure storage, transport and disposal
  - Reduce dependence on foreign energy sources
- **Environmental Impact**
  - Mitigation of Health Hazards
    - *Safe, permanent disposal (isolation from humans)*

Do benefits (N power) outweigh risks (of waste)?

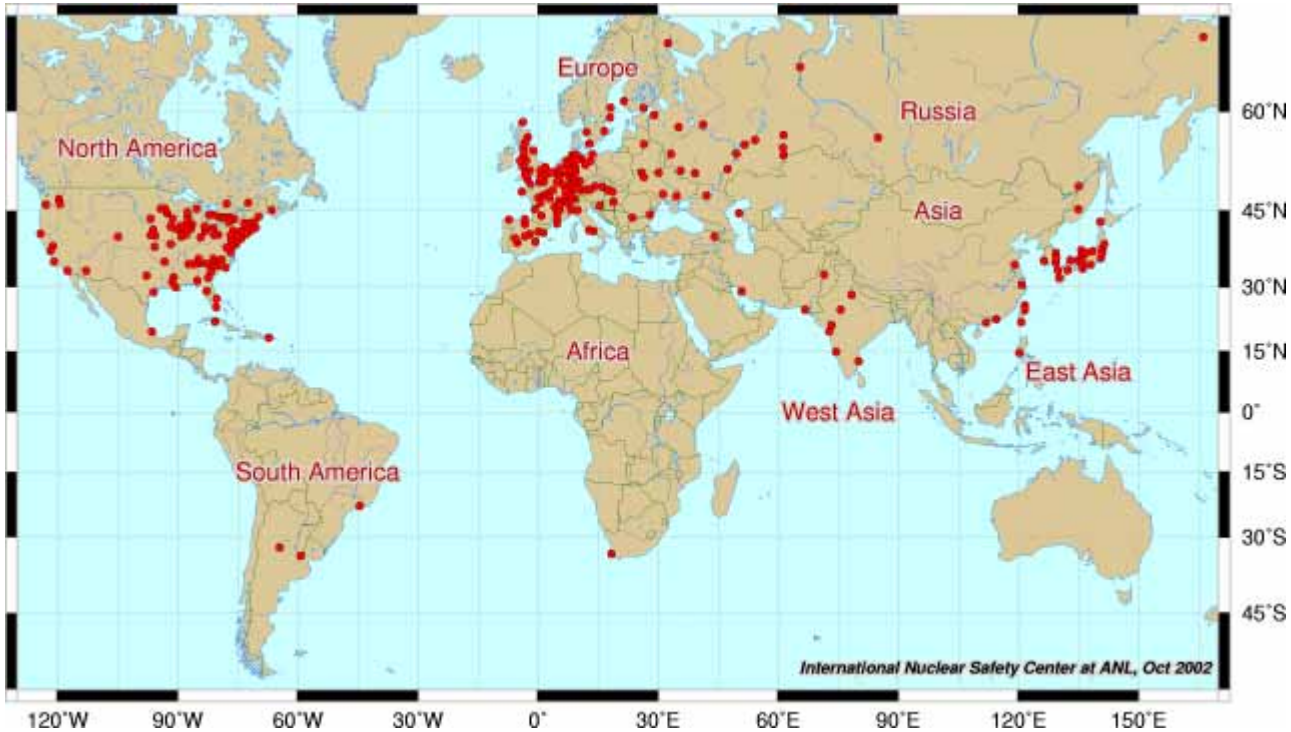
U.S. Reactors and Spent Fuel Storage



A Worldwide Concern

As of June 1998:

- 437 Nuclear Power Reactor Operating Worldwide
- 25 Nuclear Power Reactors Being Built
- 79 Nuclear Power Reactors Ordered or Planned



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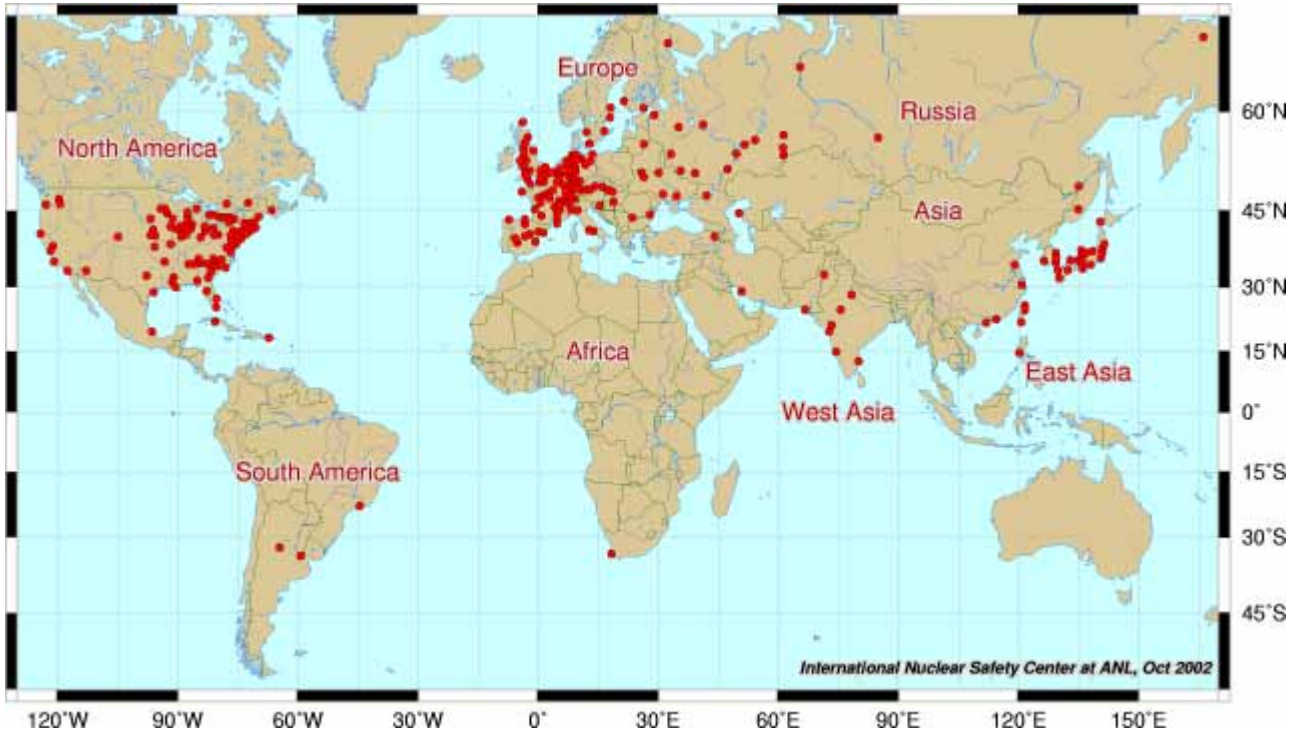
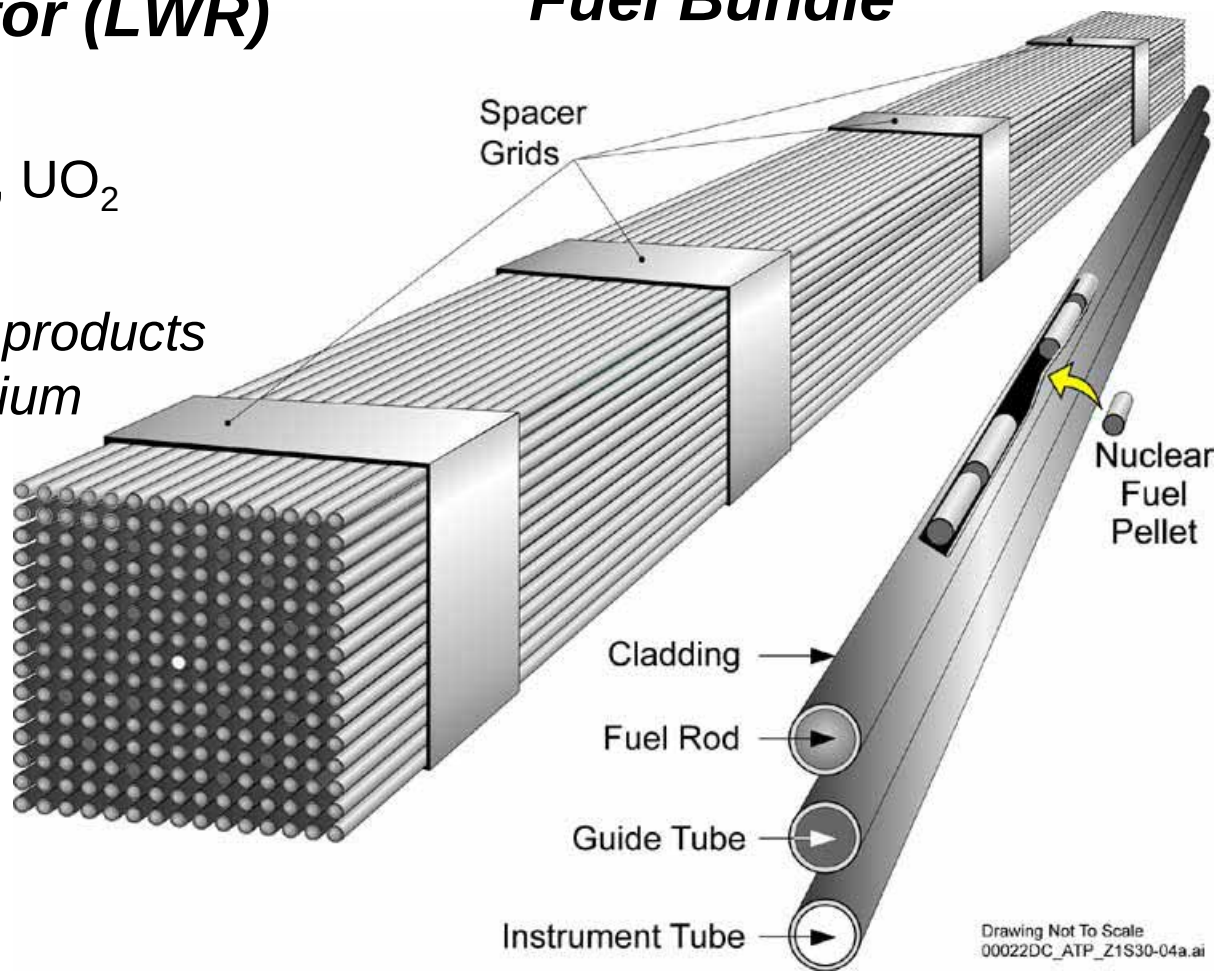


What is Spent Fuel?

Light Water Reactor (LWR)

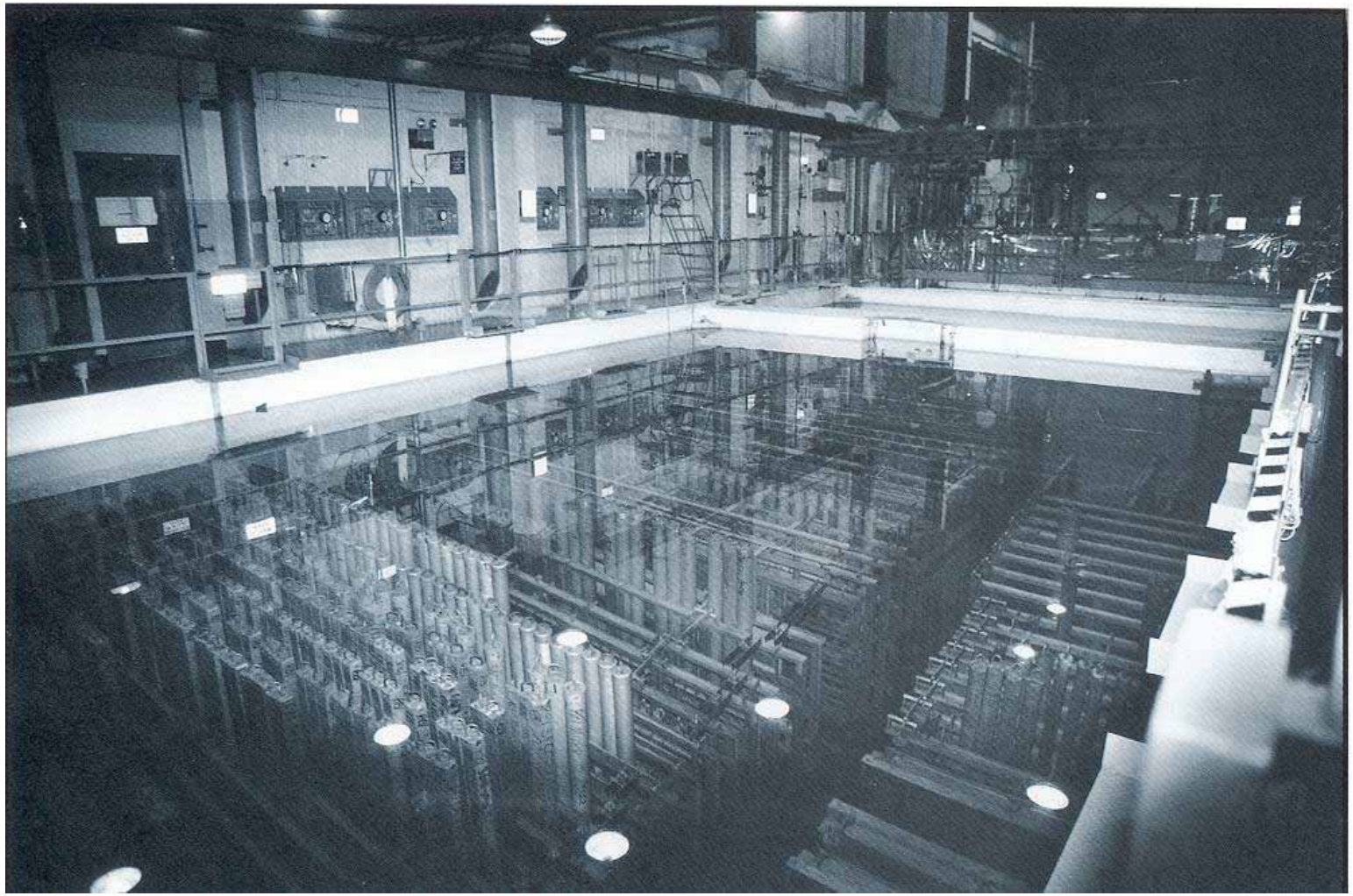
- **Fuel Pellets**
  - Uranium Dioxide, UO_2
    - *+ Minor solids*
    - *2 - 5% fission products and transuranium actinides*
- **Cladding**
  - Zirconium alloy
- **Spacer Grids**
  - Steel

Fuel Bundle



Spent Fuel (de-clad)

Spent Fuel Storage



Some Radioactive Waste Management Options

- **Permanent Disposal**

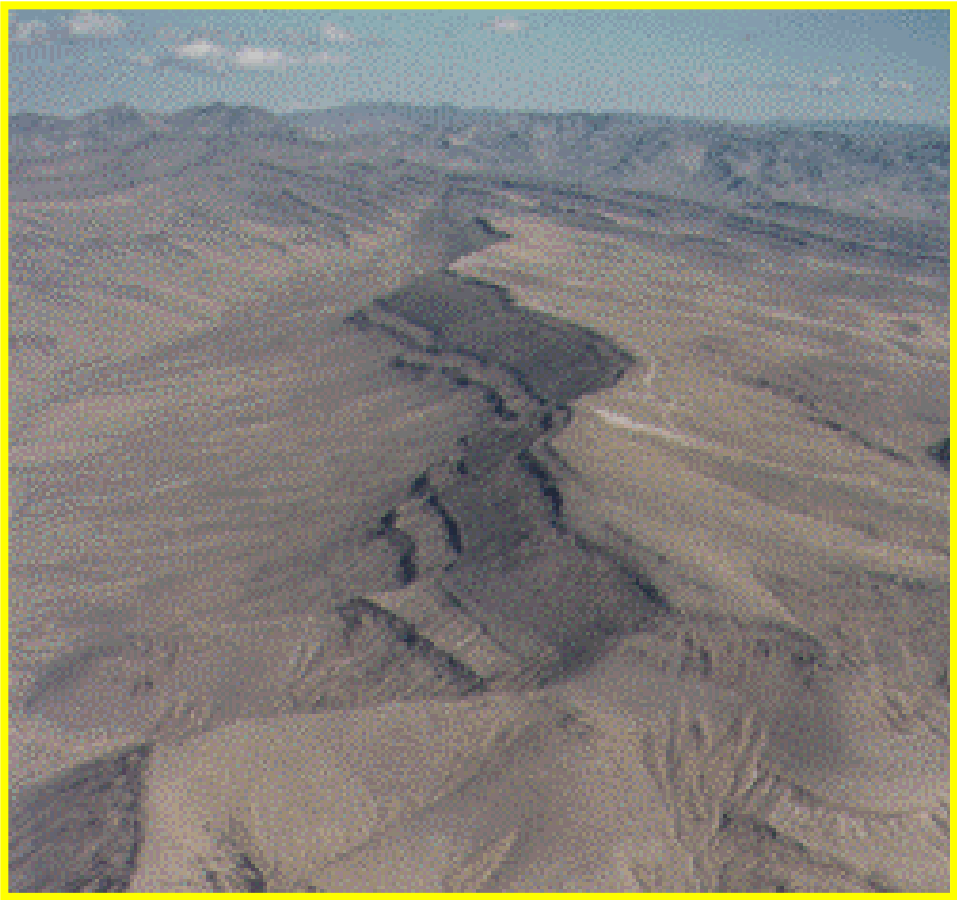
  (isolation from the human-accessible environment)

  - **<u>Geologic</u>**
    - *"Wet" vs. "Dry"*
  - Other
    - *Space, Sea bed, Ice caps, Deep bore holes, Remote islands*
- **Reprocessing**
  - *does not eliminate* need for permanent disposal
- **Transmutation**
  - ? Substantial development still needed …
- **Wait …**
  - Temporary monitored storage
    - *Single site or multiple sites?*

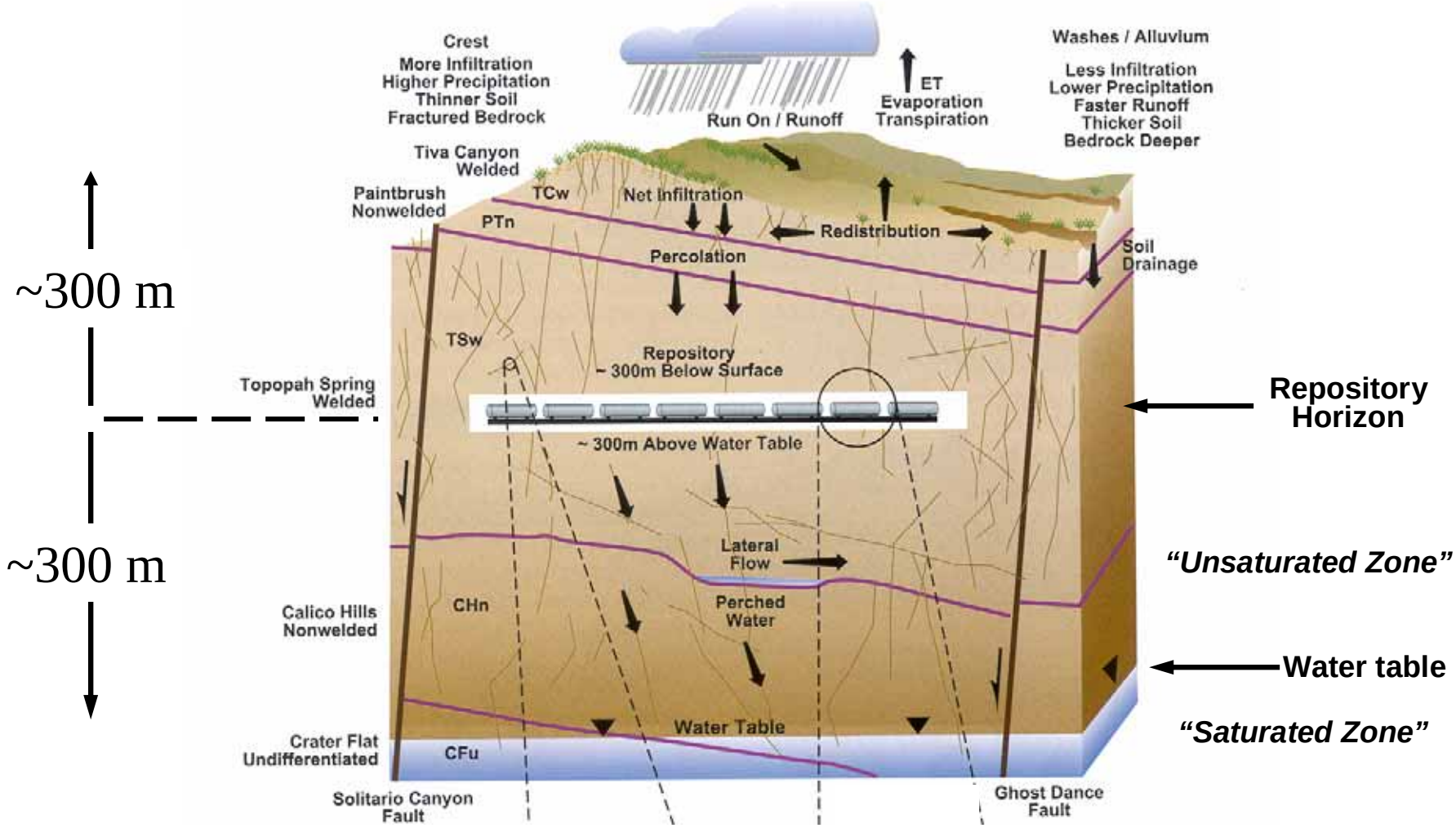
Geologic Disposal

Yucca Mountain, Nevada

- Under study since ~1983
- Since 1987: The *only* US site being considered as a potential repository for spent nuclear fuel

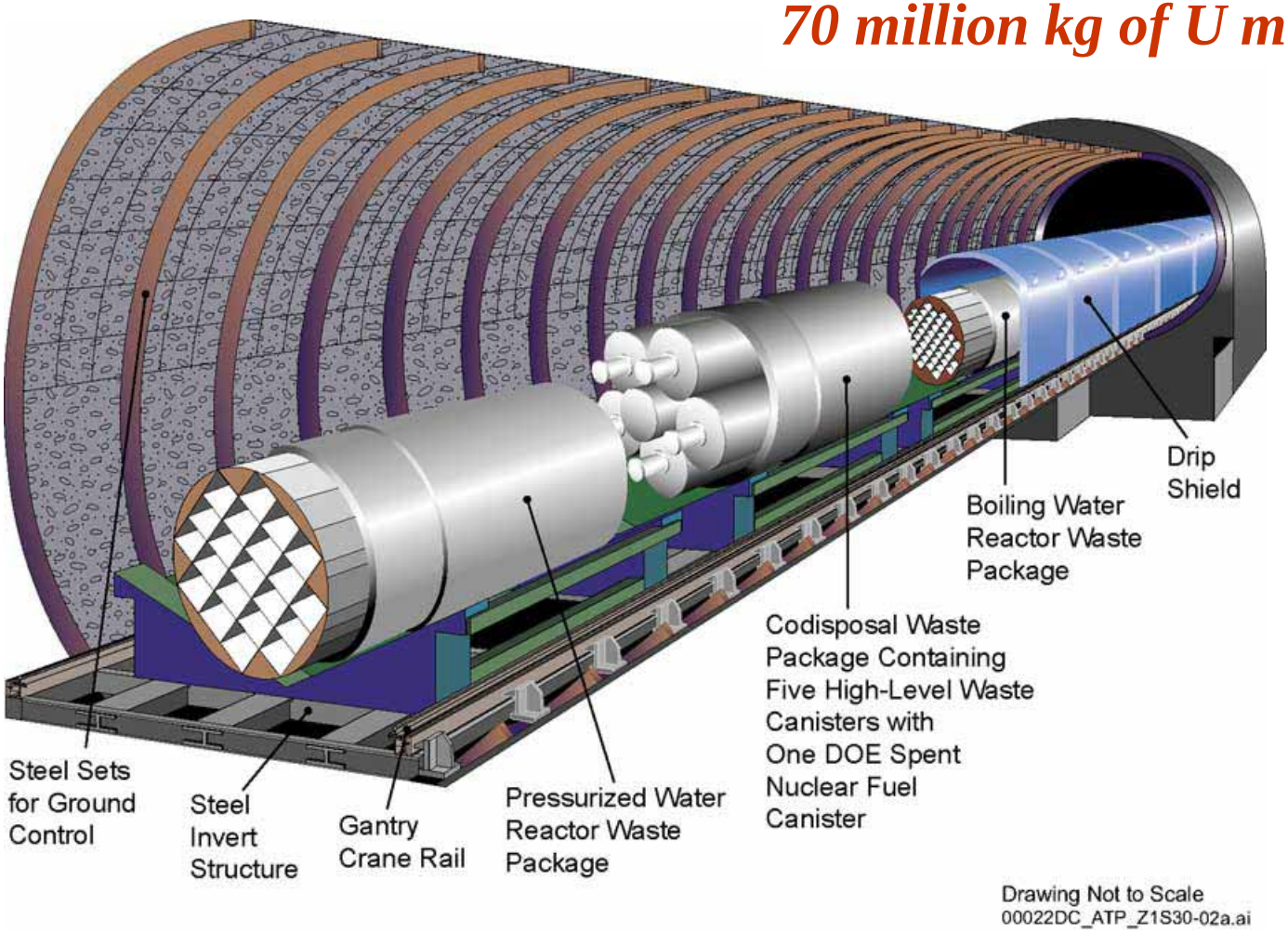


Geologic Barriers

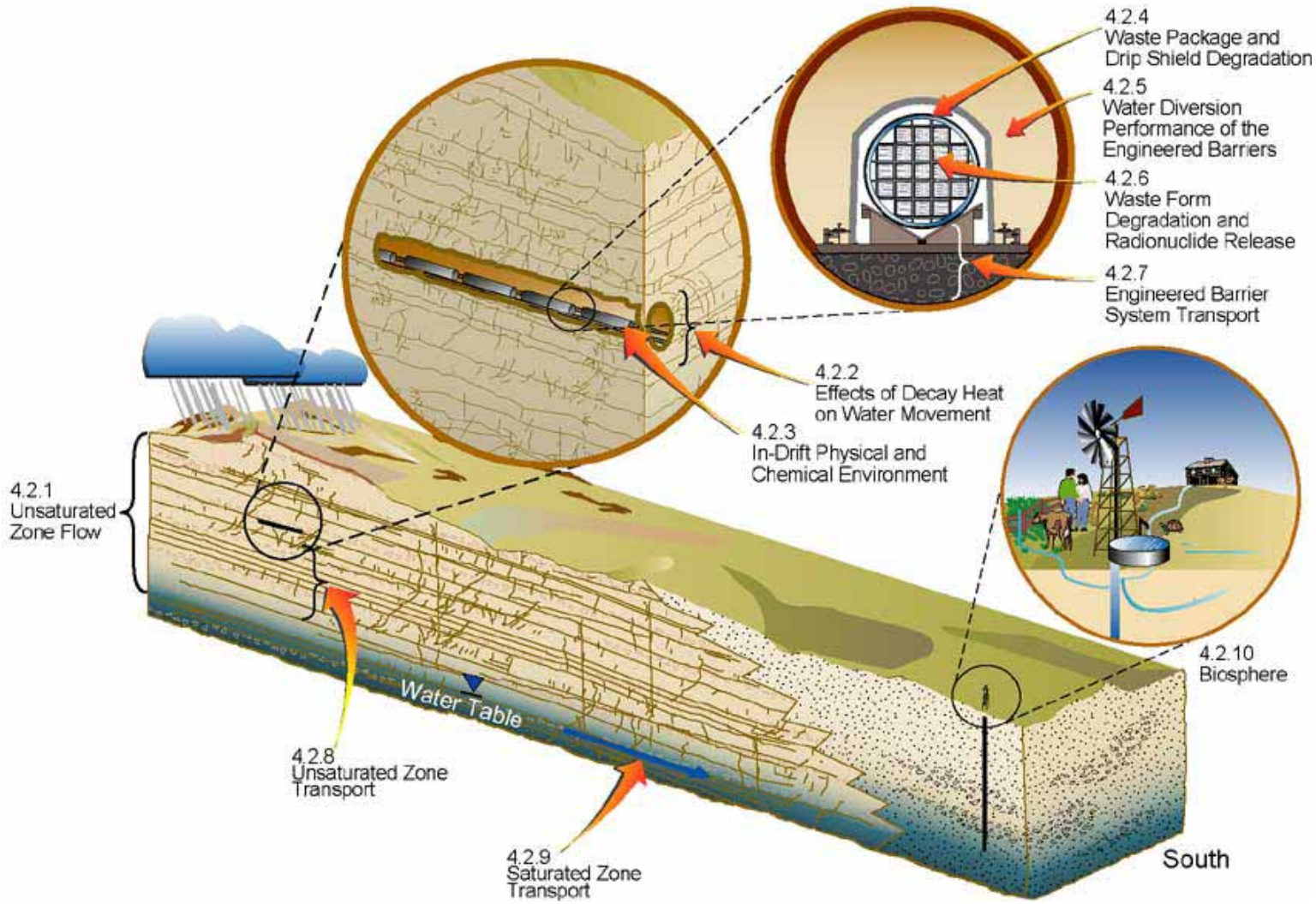


Engineered Barriers

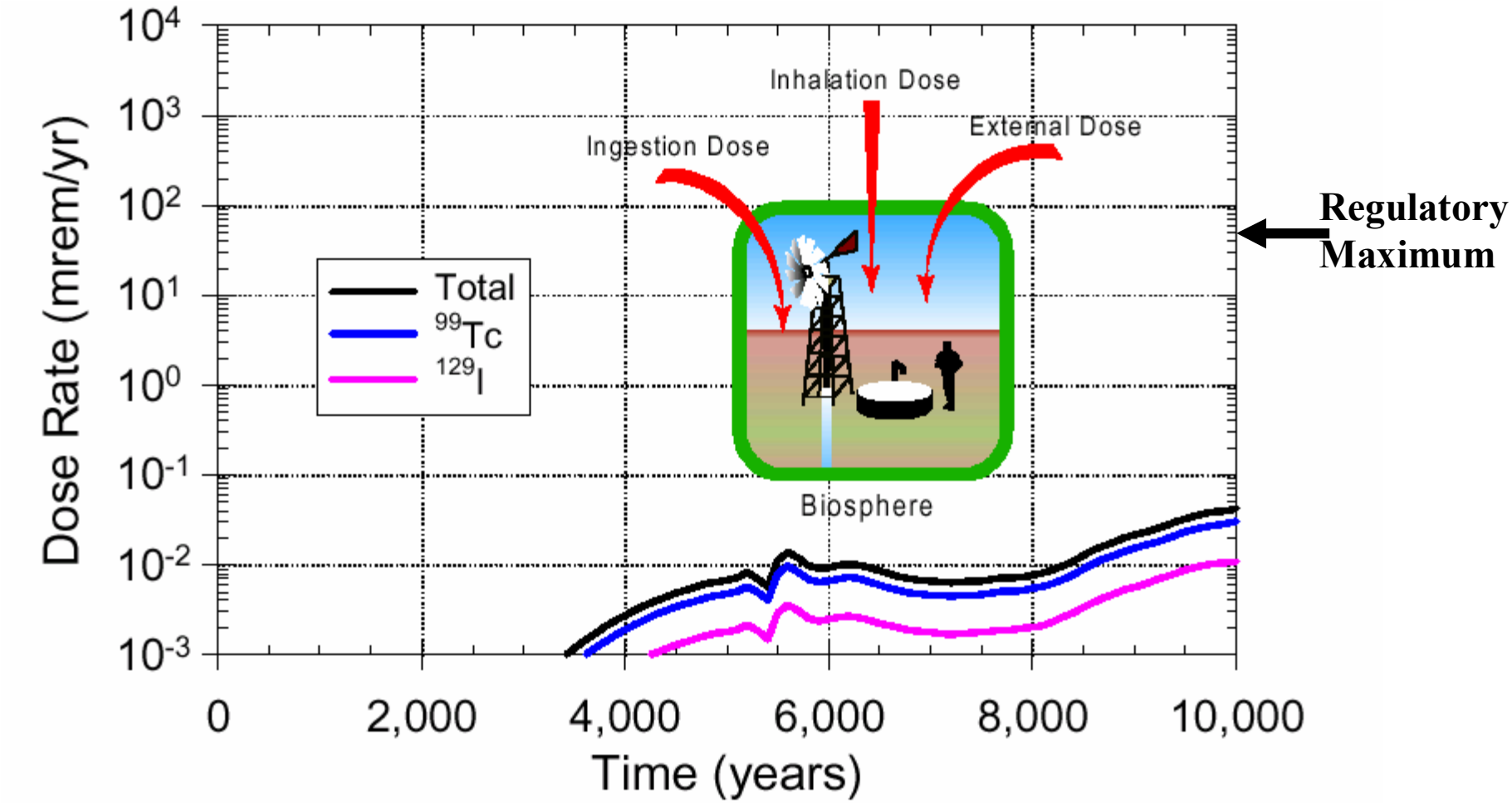
70 million kg of U metal



Assessing Long Term Repository Safety

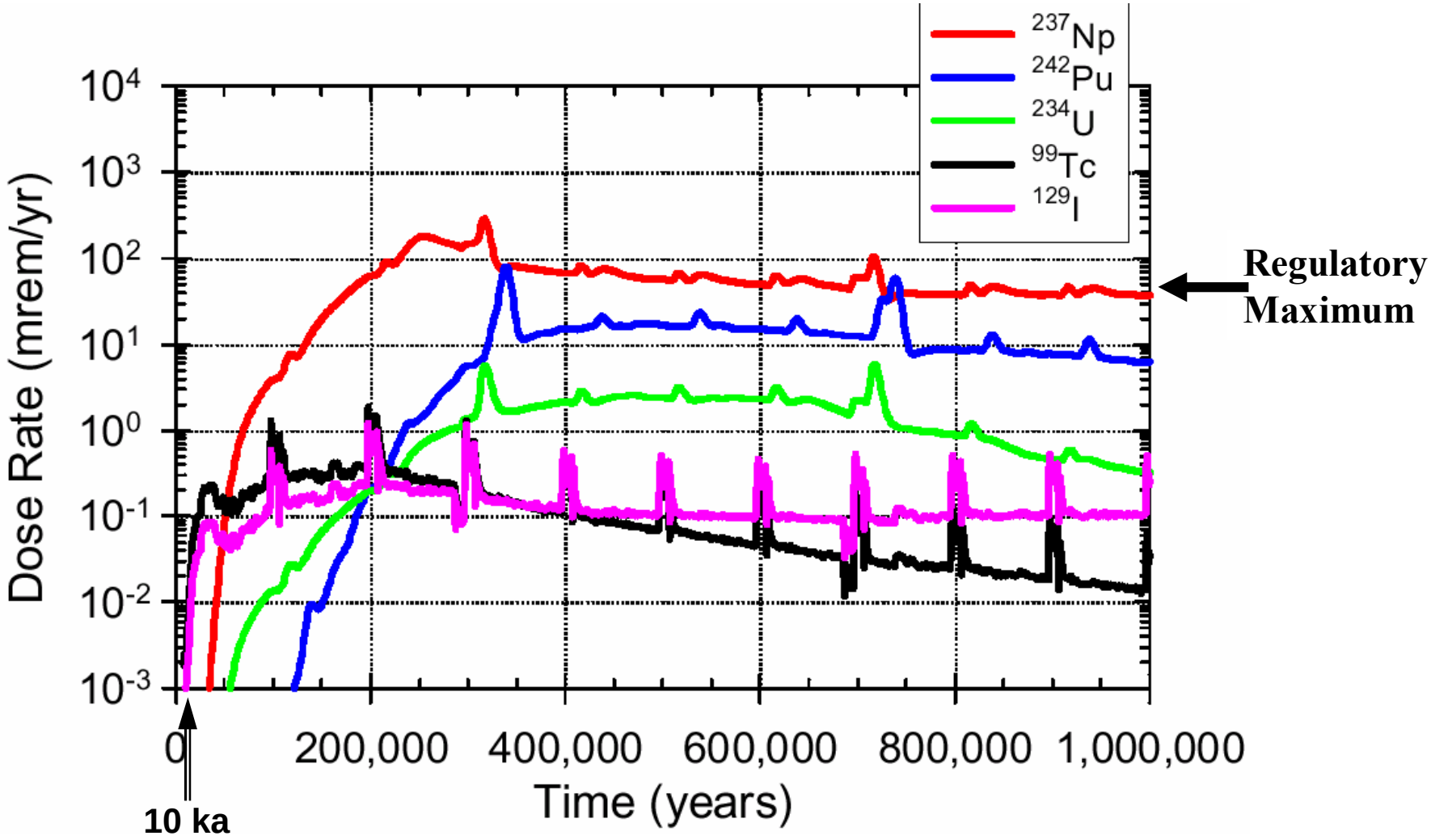


Assessing Safety: 10,000 year Dose Rate



US DOE TSPA-VA. December 1998. (www.ymp.gov)

Calculated Dose Rates to One Million Years



US DOE TSPA-VA. December 1998. (www.ymp.gov)

Performance Assessment Models *(Major Components)*

- **Unsaturated-zone Flow & Transport**
- **Thermal Hydrology**
- ➢ **Near-field Chemical Environment**
- ➢ **Waste-package Degradation**
- ➢ **Waste-form Alteration**
- **Saturated-zone Flow & Transport**
- **Biosphere**
- **Disruptive Events**
  - seismicity, igneous intrusion, criticality, human intrusion, etc.
- **Alternative Design Options**

What is Spent Fuel?

Light Water Reactor (LWR)

- **Fuel Pellets**
  - Uranium Dioxide, UO_2
    - *+ Minor solids*
    - *2 - 5% fission products and transuranium actinides*
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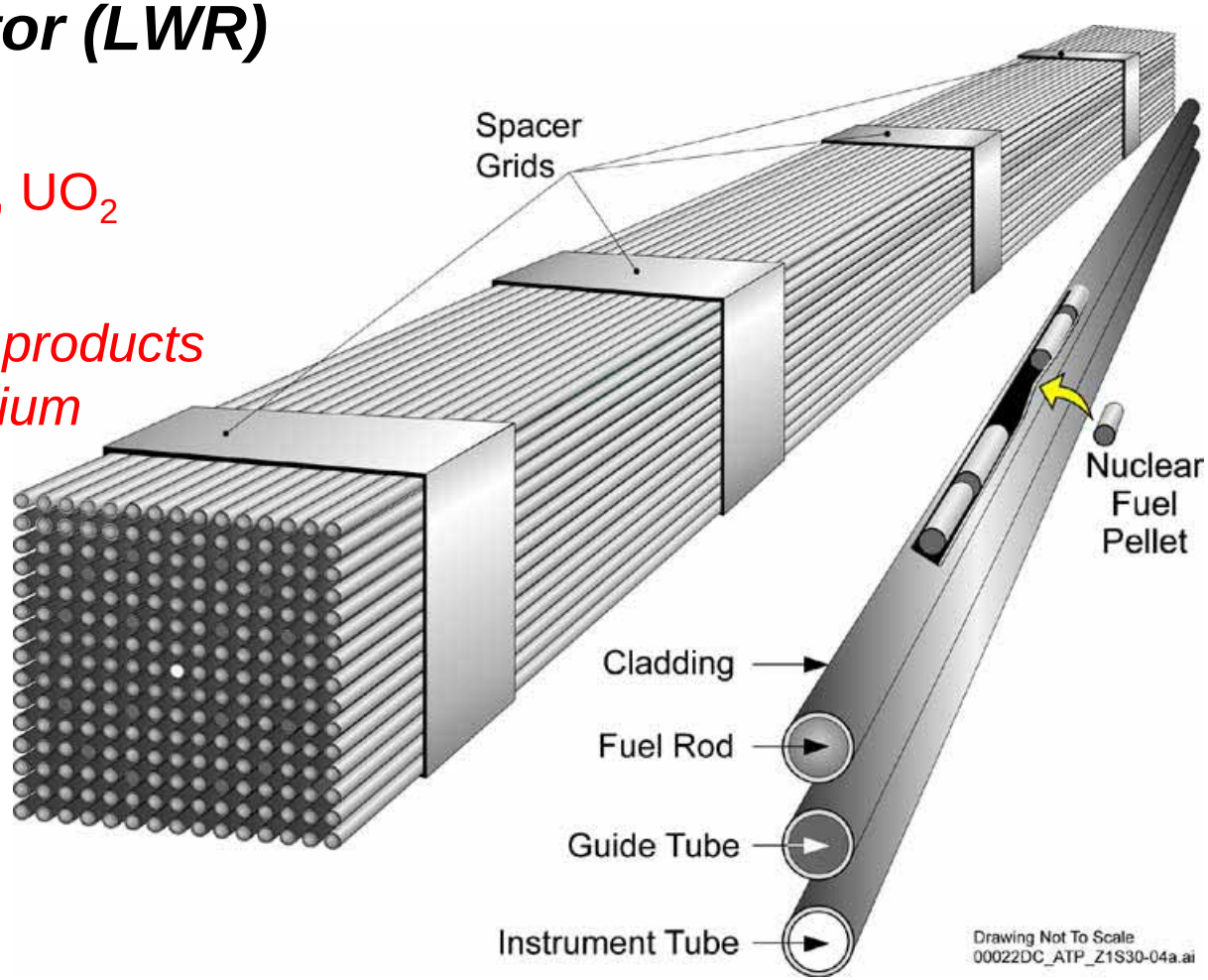
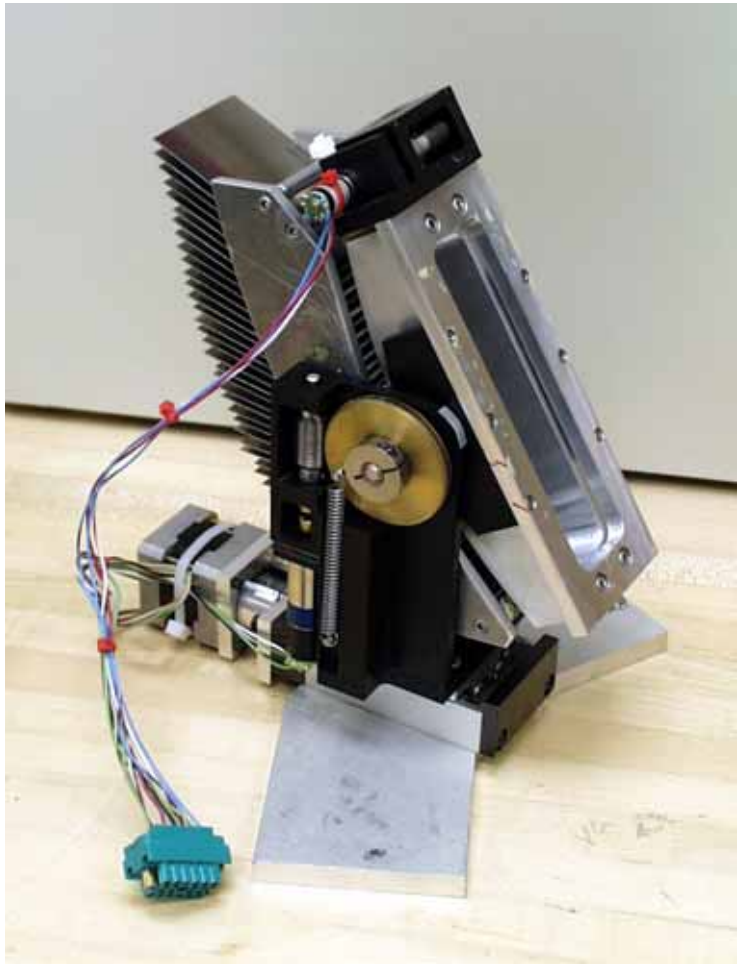


Advanced Photon Source

Bent Laue Analyzer



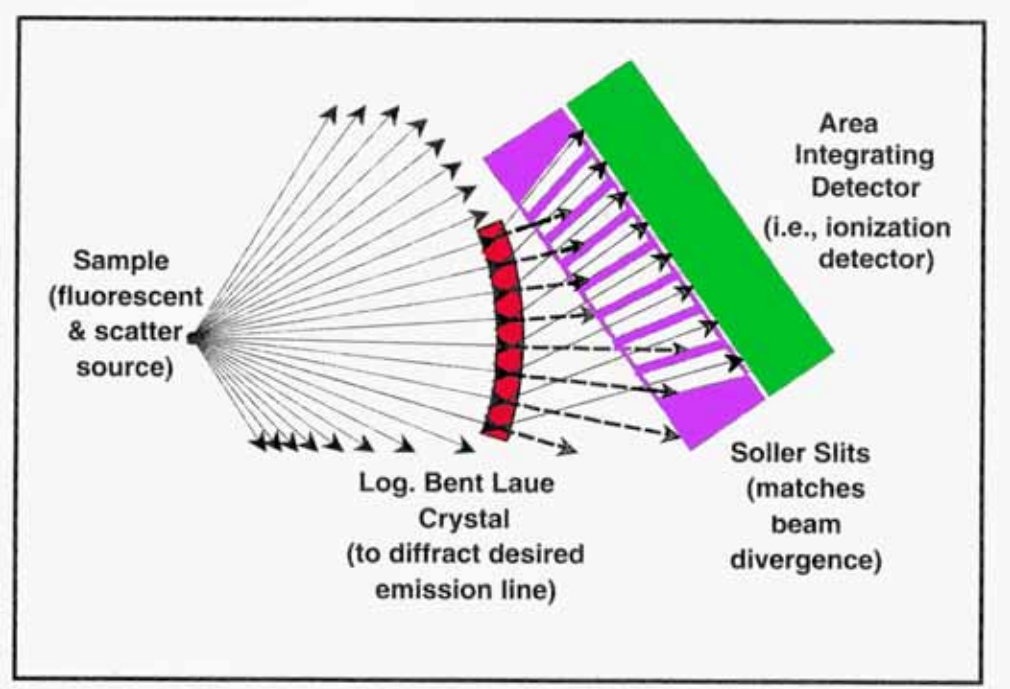
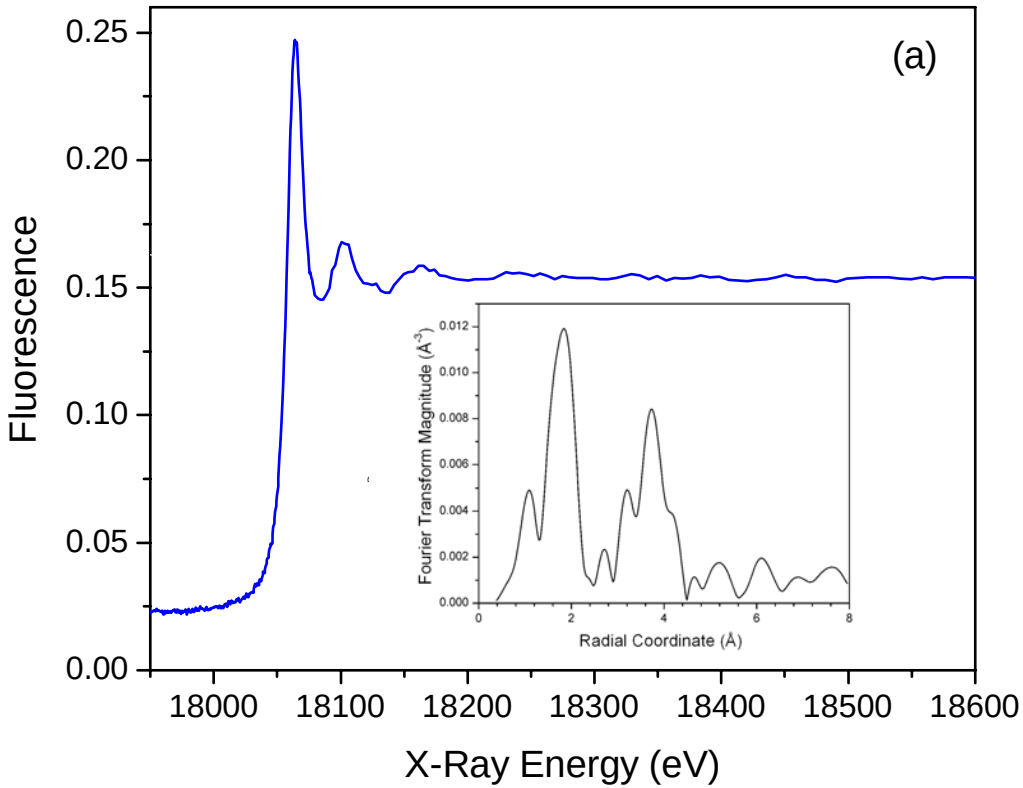


FIG. 1. A sketch of the "logarithmic bent Laue analyzer" concept.

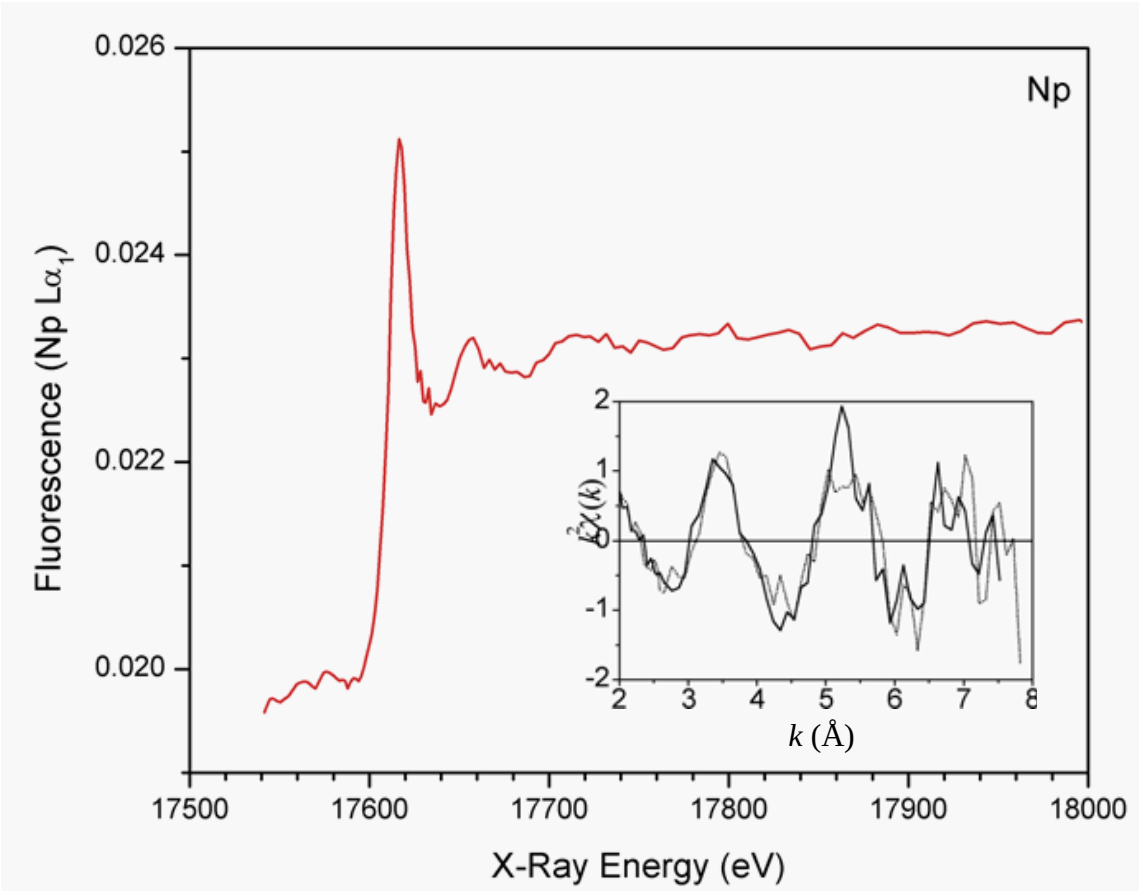
From *APS Forefront*, Argonne National Laboratory, October 1, 2002

Plutonium in Unaltered Spent Fuel



- 8300 ppm Pu
- Pu(IV) oxide in UO_2
  - $R_{Np\text{-}O}$ = 2.34(3) Å
  - $R_{Np\text{-}An}$ = 3.85(4) Å

Neptunium in Unaltered Spent Fuel



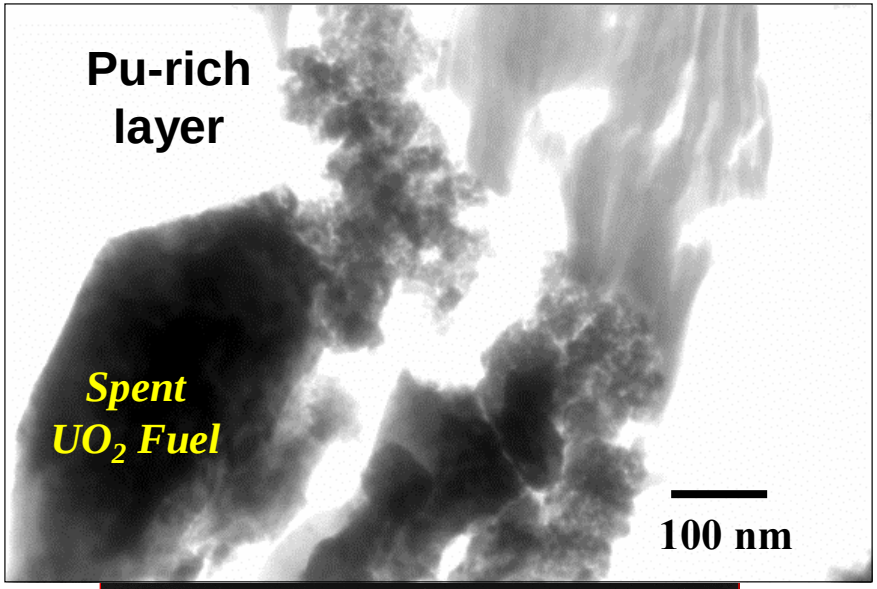
- 600 ppm Np
- Np(IV) oxide in UO_2
  - $R_{Np\text{-}O}$ = 2.34(3) Å
  - $R_{Np\text{-}An}$ = 3.85(4) Å

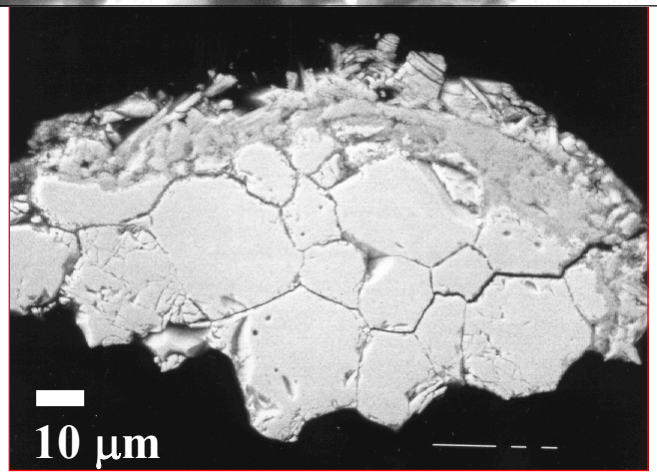
Fortner, Finch, Kropf, and Cunnane, submitted to Nucl. Tech.

Waste Form Corrosion

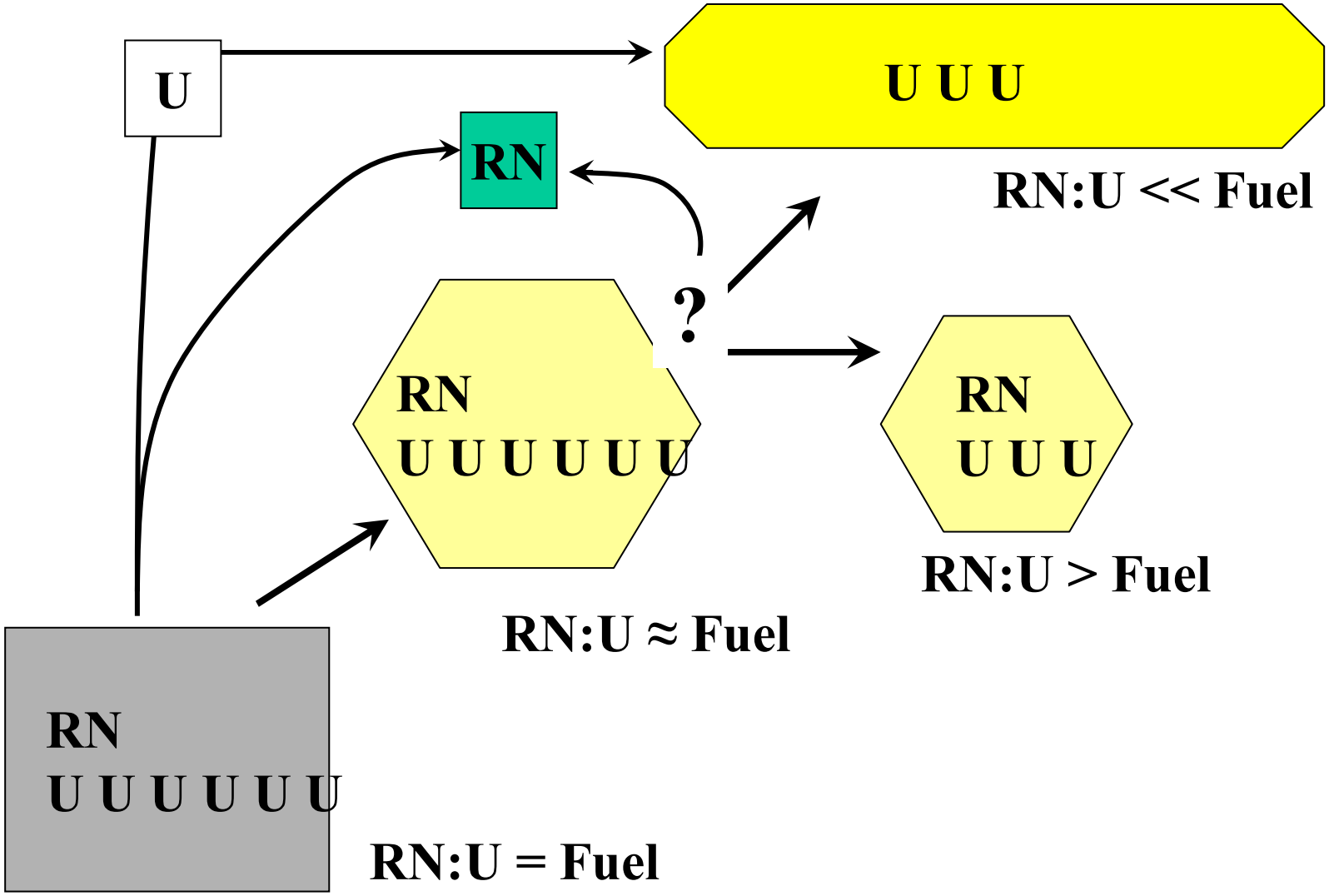
- **What is the long-term behavior of waste forms?**
  - Dissolution & corrosion → Radionuclide (RN) release
    - *Rates of dissolution*
- **What roles for corrosion products?**
  - Solid fission-product and actinide hosts → RN retention
  - Colloids → Potential RN migration
  - Sorption to steel corrosion products
- **Water chemistry**
  - Deliquescence
  - Thermal evolution of groundwater
  - Radiolysis
    - *Decomposition of H_2O by radiation*

Spent Fuel Corrosion

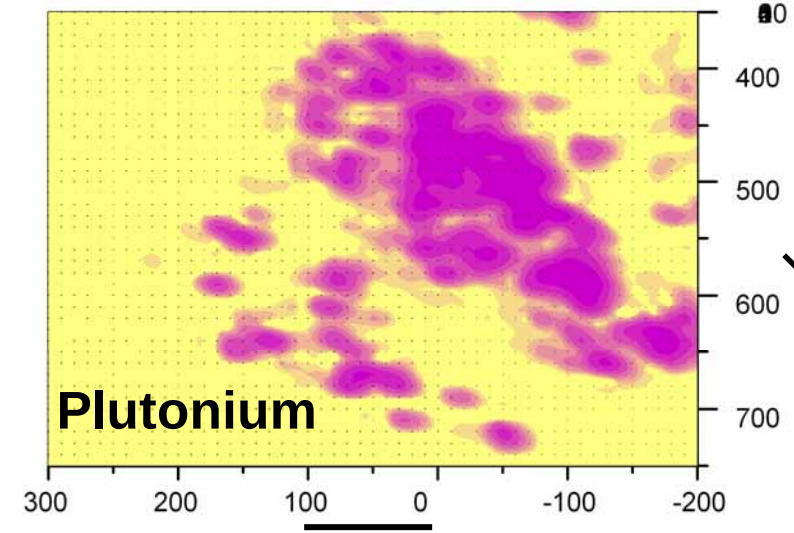




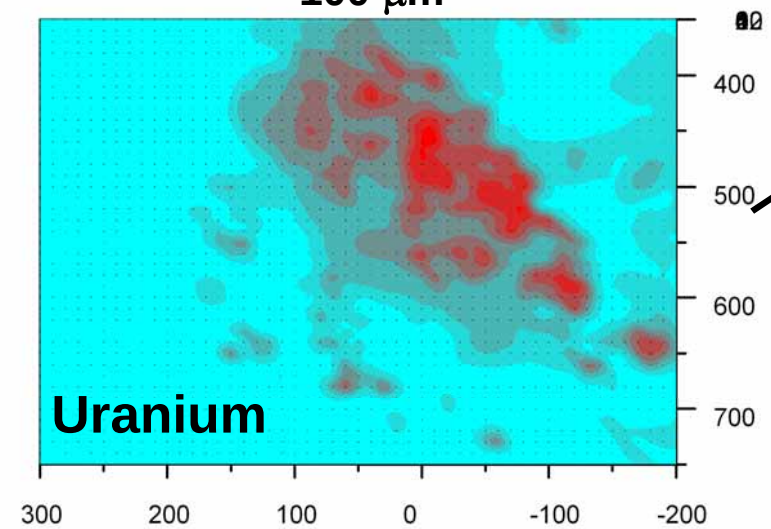
Long Term Alteration of Radionuclide Solids



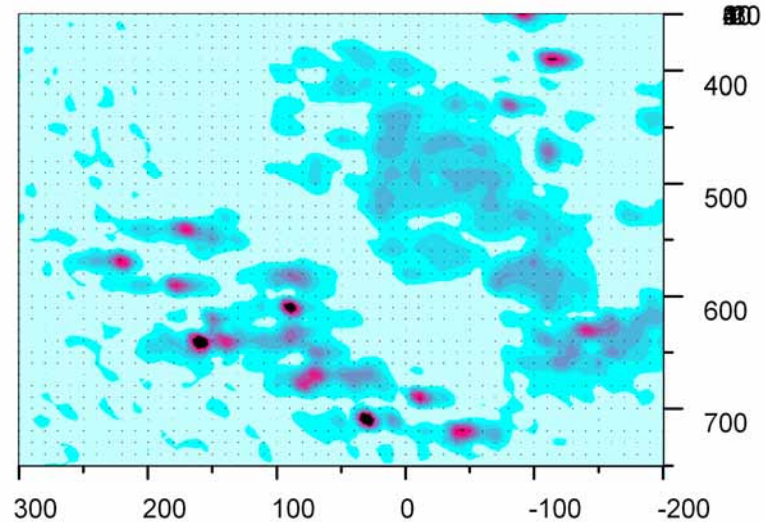
Plutonium & Uranium (Corroded Spent Fuel)



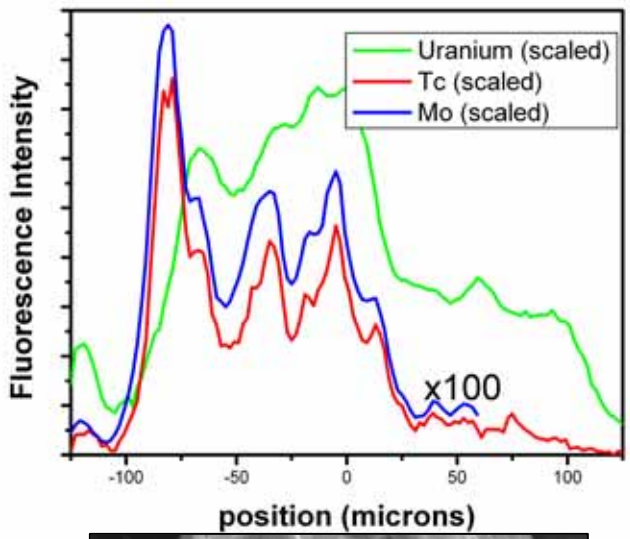
100 μm

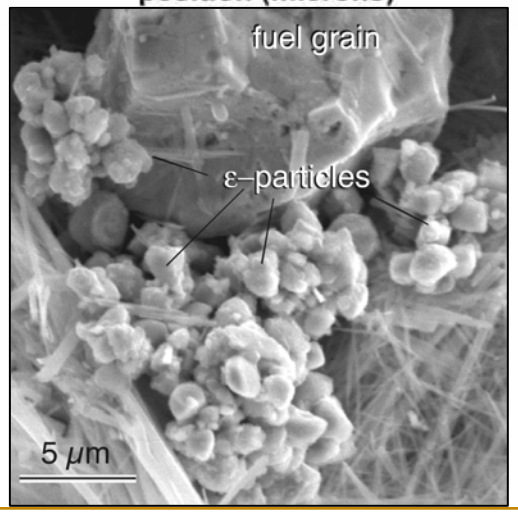


Ratio (Pu/U)

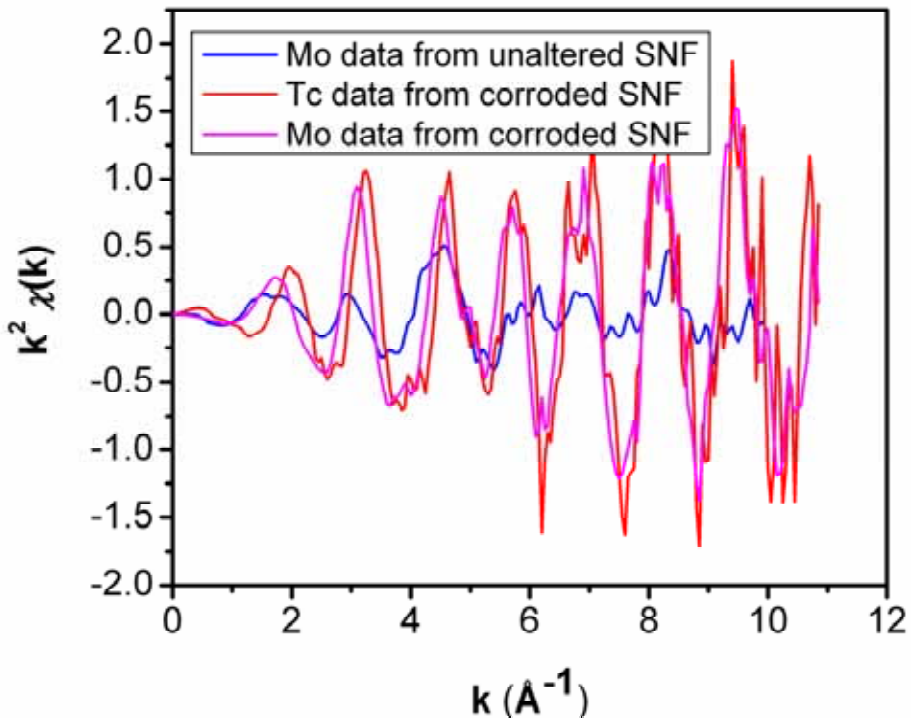


X-Ray Fluorescence Line Scans: Technetium



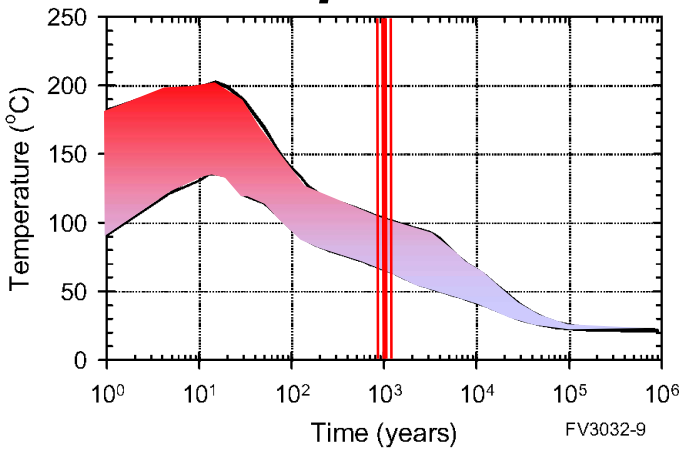


- **Epsilon Particles**
- **XAFS shows they are metallic**

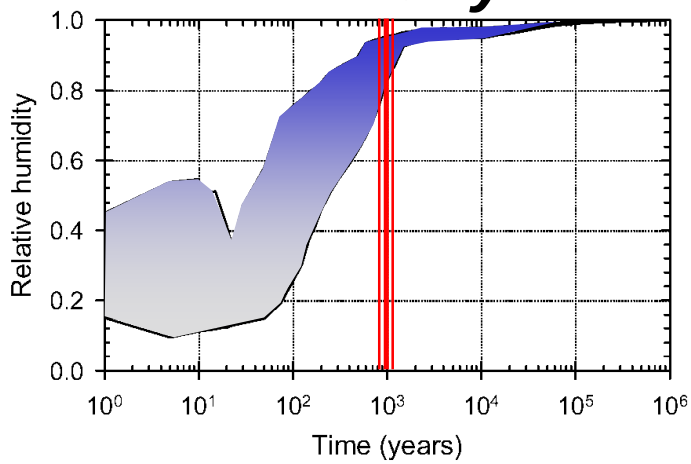


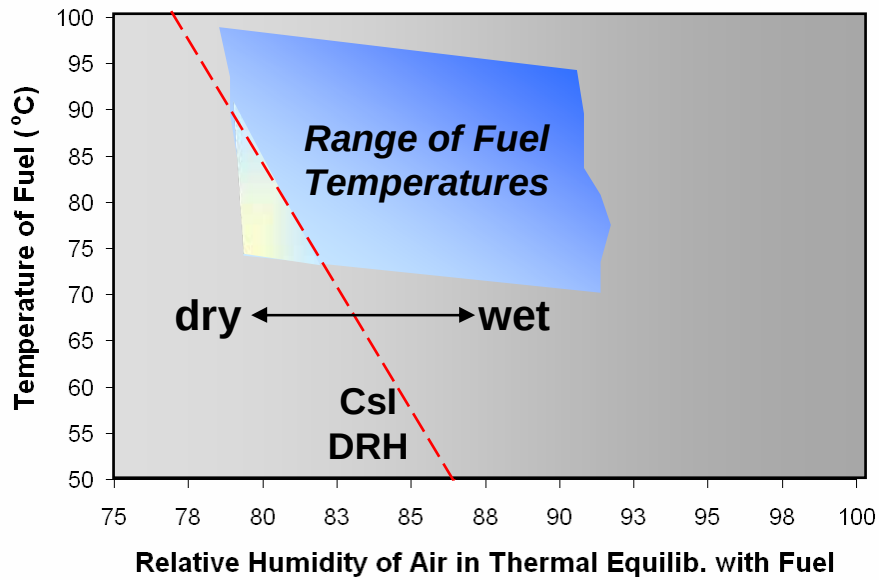
Repository Temperature & Humidity

Temperature



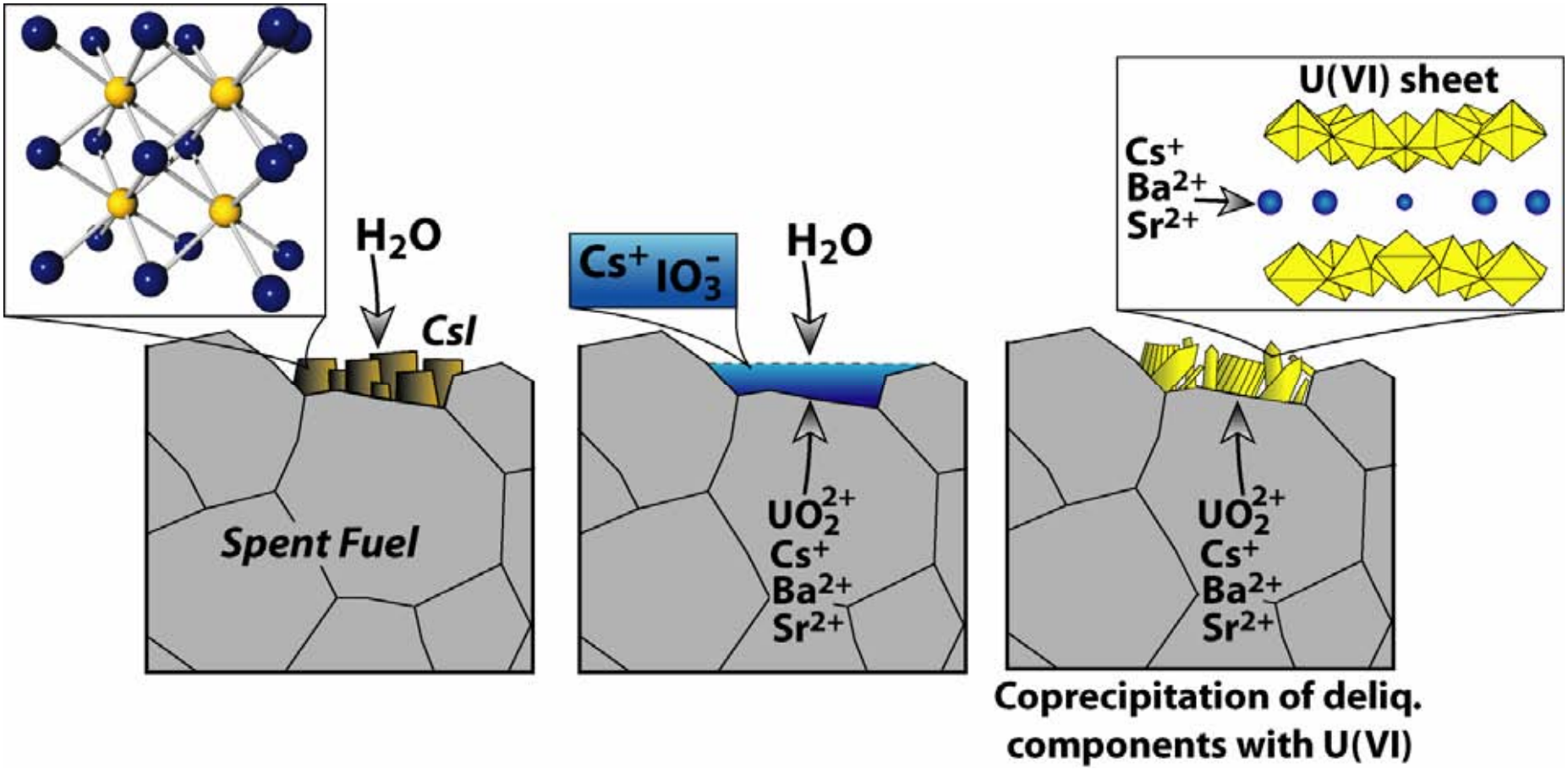
Humidity





From TSPA-VA December 1998

Deliquescence & Corrosion Products



Incorporation of certain elements may shut off deliquescence

Colloids

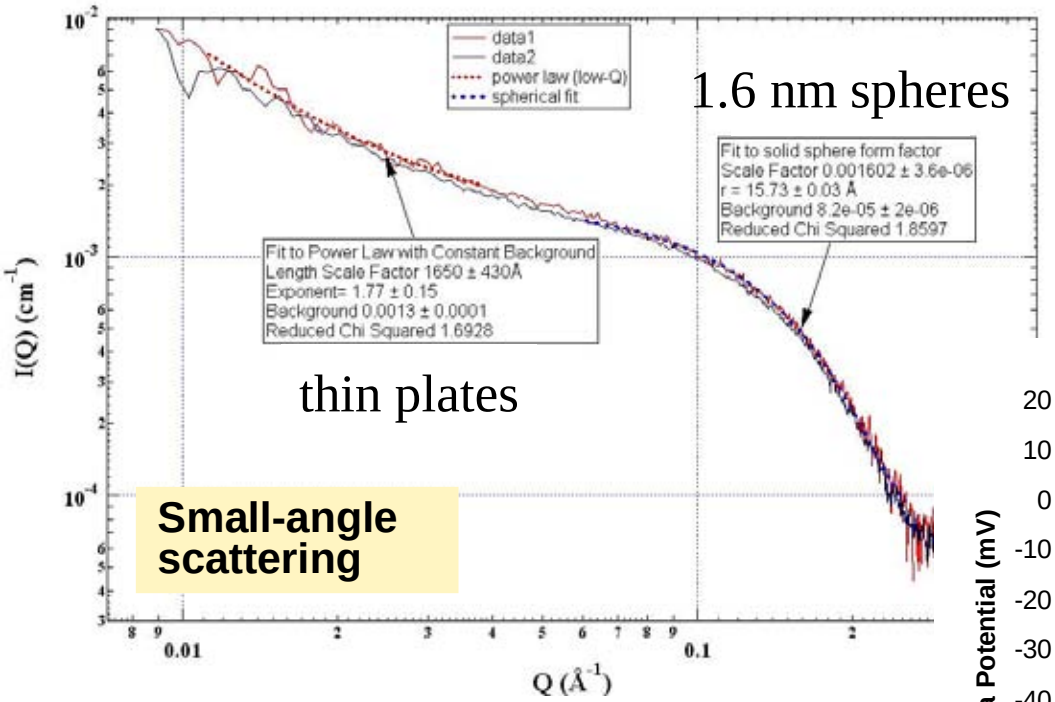
Small: up to ~1000 nm

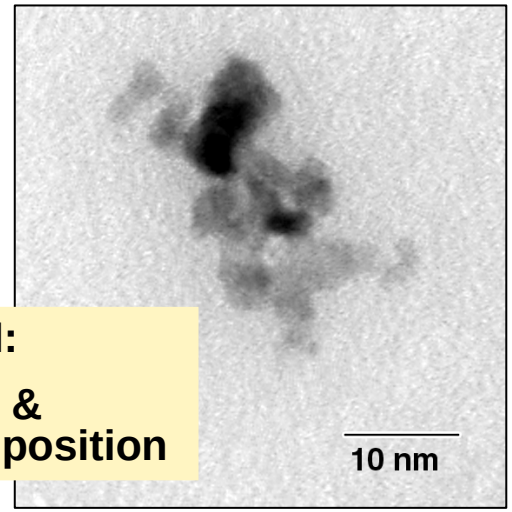
Large specific surface area

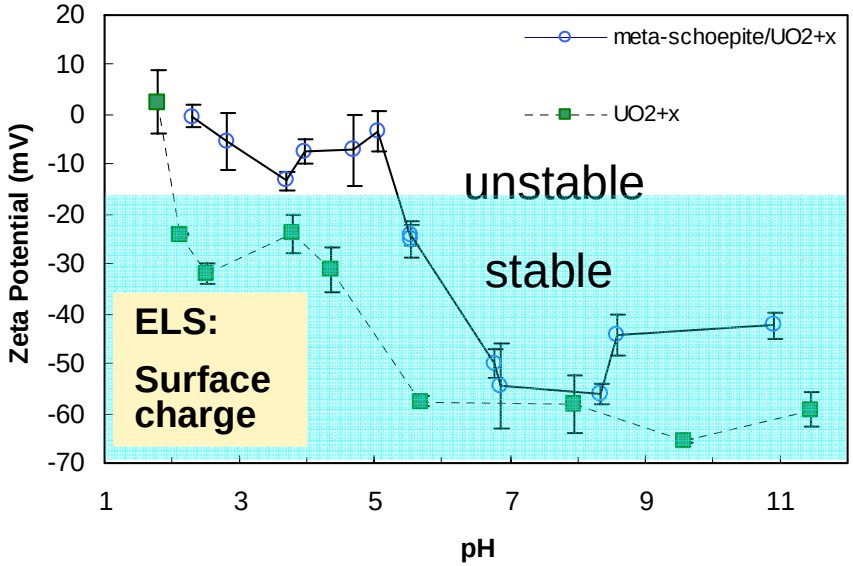
Three types of radionuclide-bearing colloids

1. Existing groundwater colloids
   - *May sorb radioelements from solution*
2. "True" colloids
   - *Precipitated from saturated solution*
3. "Primary" colloids
   - *Detached from altered waste forms*

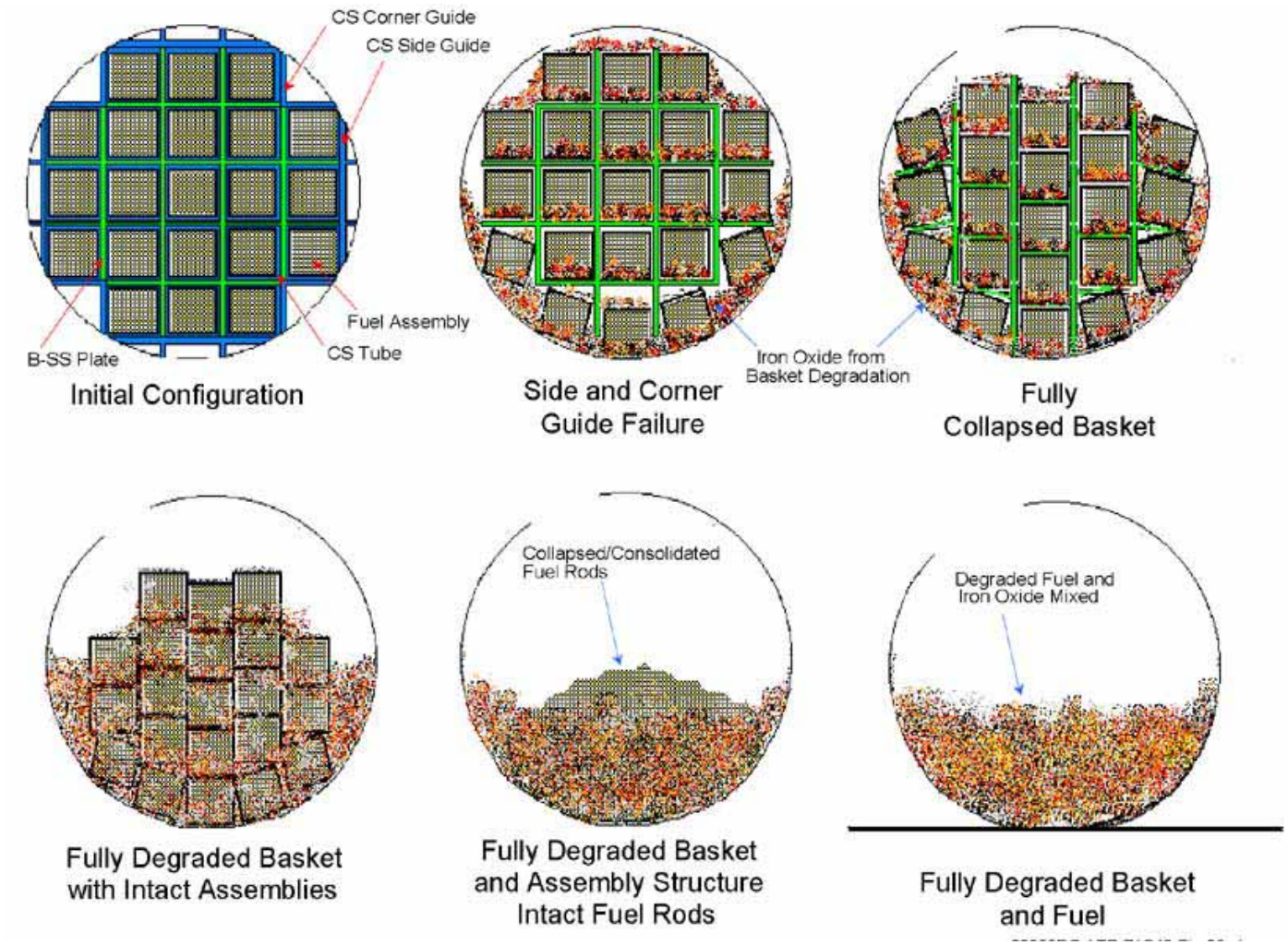
Developing Mechanistic Colloid Models







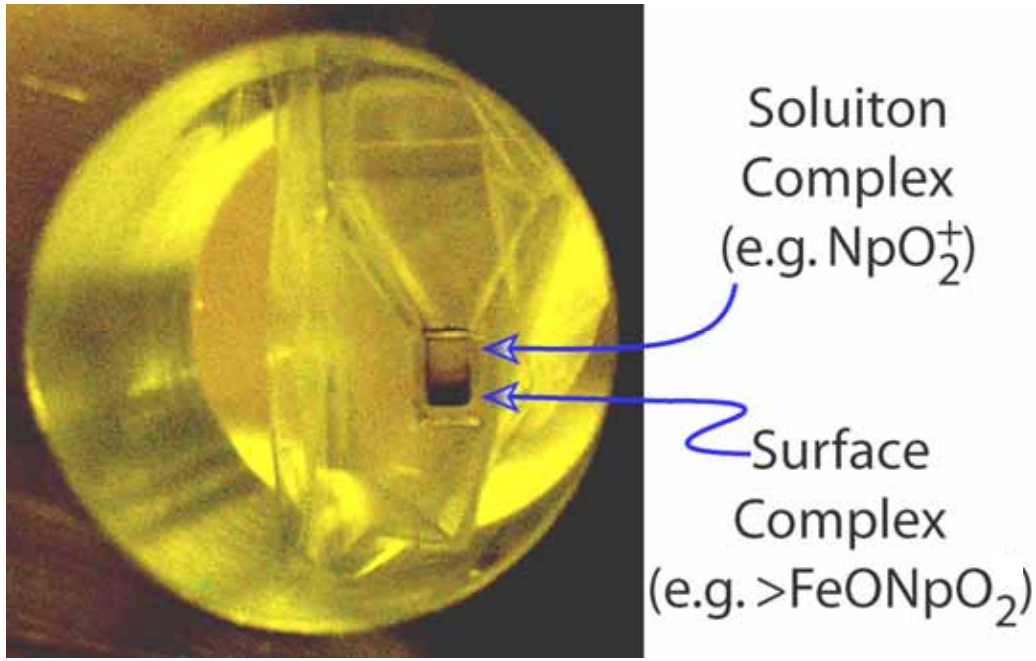
Degradation of the Waste Package



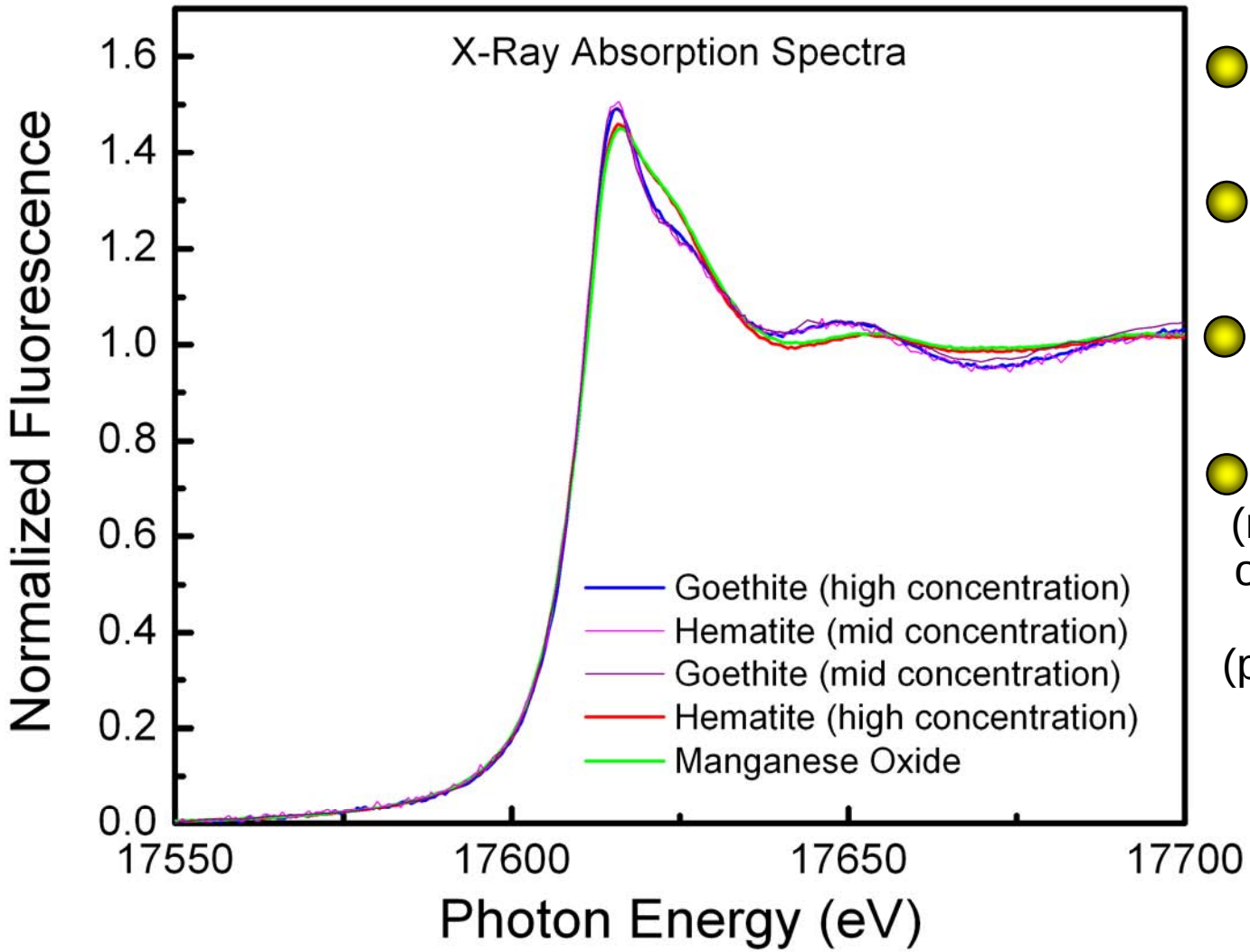
X-ray Adsorption Spectroscopic analyses of neptunium at mineral interfaces: surface complexation

New sample holder design fabricated for these experiments

Hematite equilibrated with 10^{-5} Molar Np(V) solution



Neptunium Sorption on Fe & Mn Oxides



- Sorbent / Solution: ~1 g/L
- High concentration: 10^{-4} Molar Np
- Mid concentration: 10^{-5} Molar Np
- Goethite & Hematite (mid con) are Np(V). In other samples Np may be mixed valence (prelim. interpretations).

Some Additional Waste Form Corrosion Issues

- **Processes affecting dissolution of spent fuel [rates]**
  - Evolution of effective surface area
  - Surface complexation
  - Roles of minor components (trace elements/minor solids)
- **Very Long Term Processes**
  - Radionuclide solubilities (Henry's Law behavior)
  - Sorption/desorption & Ostwald ripening
- **Radiation effects**
  - Water radiolysis
  - Radiation damage
  - Evolution of fuel properties
- **Future geologic repositories?**
  - Siting & design
    - *Applying what we've learned …*

Acknowledgements

ANL-CMT

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Other colleagues

- Rodney Ewing (UM), David Wronkiewicz (UMR), Peter Burns (NDU), John Hanchar (GWU)

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