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INTEGRATING USED FUEL DEGRADATION MODELS INTO GENERIC PERFORMANCE ASSESSMENT

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Used Fuel Disposition

Presentation Outline

- Used Fuel Disposition Campaign Background
- Used Fuel Degradation & Radionuclide Mobilization (UFD&RM) Overview
- Integration Summary
- Generic Performance Assessment Models (GPAM) Concepts
- Engineered Barrier System (EBS) Interface
- UFD&RM Model Structure
- Accomplishments and Summary

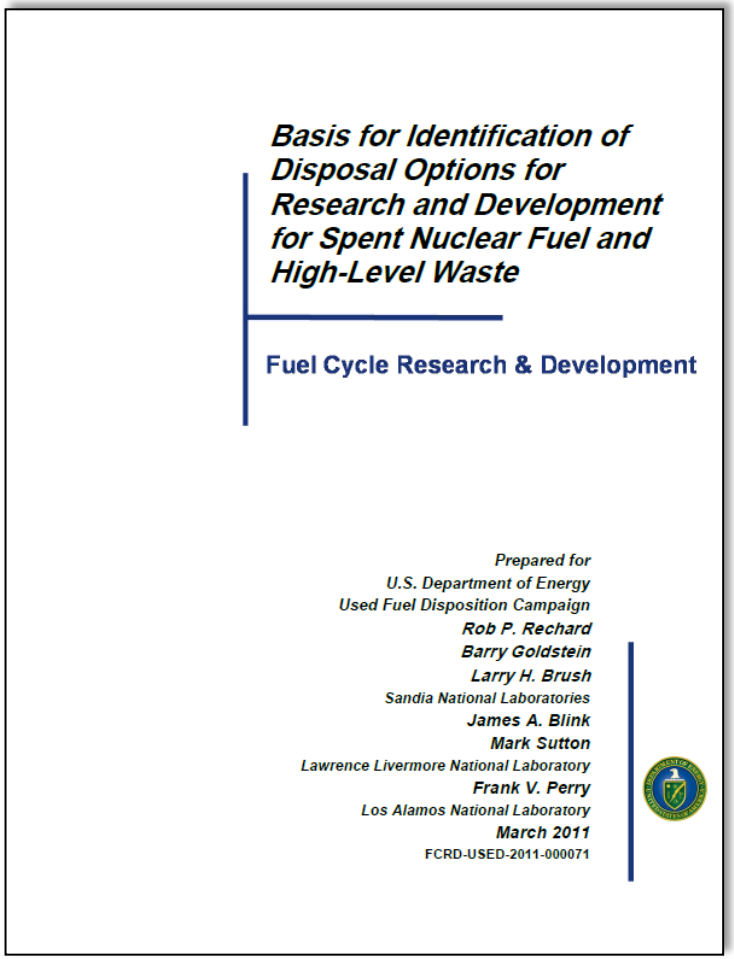
Used Fuel Disposition Campaign Background

DOE Office of Nuclear Energy Mission

"…advance nuclear power as a resource capable of meeting the Nation's energy, environmental, and national security needs…"

Used Fuel Disposition Campaign Mission

*"…identify alternatives and conduct scientific research and technology development to enable storage, transportation and **disposal** of used nuclear fuel and wastes generated…"*



Basis for Identification of Disposal Options for Research and Development for Spent Nuclear Fuel and High-Level Waste

Fuel Cycle Research & Development

Prepared for
U.S. Department of Energy
Used Fuel Disposition Campaign
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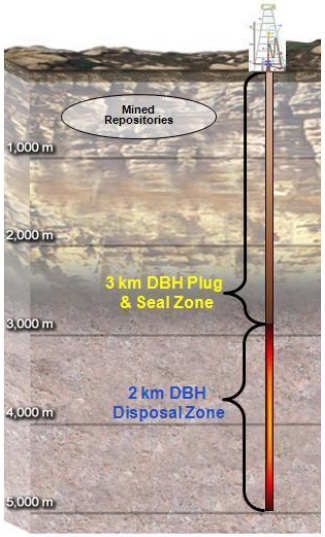
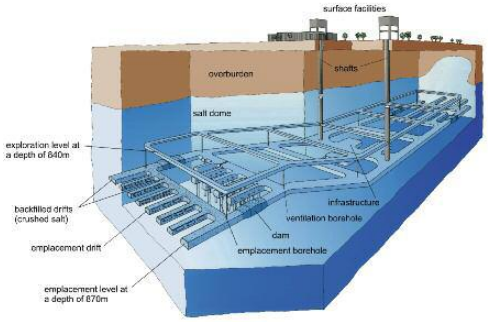
Los Alamos National Laboratory
March 2011
FCRD-USED-2011-000071

http://energy.gov/ne/listings/uranium-fuel-disposition-rd-documents

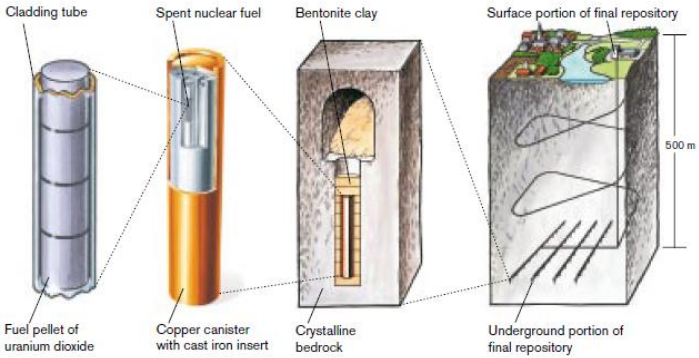
Used Fuel Disposition Campaign Background (Cont'd)

- **Mined repositories in granitic rocks, salt, and clay/shale rocks**

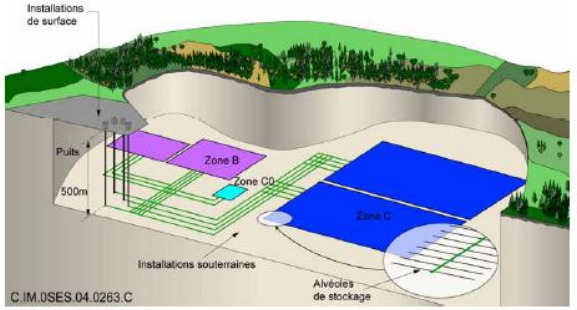




Example concept from Germany



Example concept from Sweden

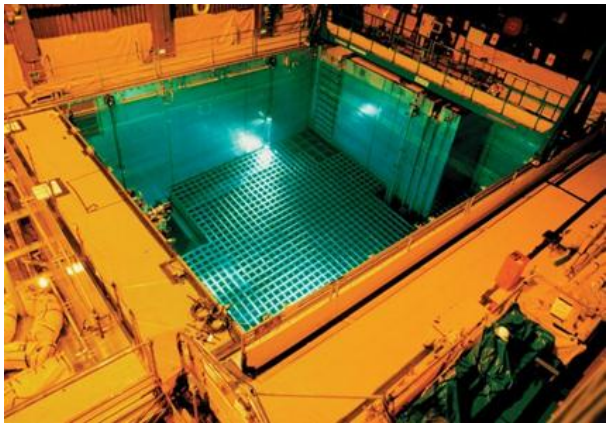


Example concept from France

- **Deep borehole disposal concepts**

Used Fuel Disposition Campaign Background (Cont'd)

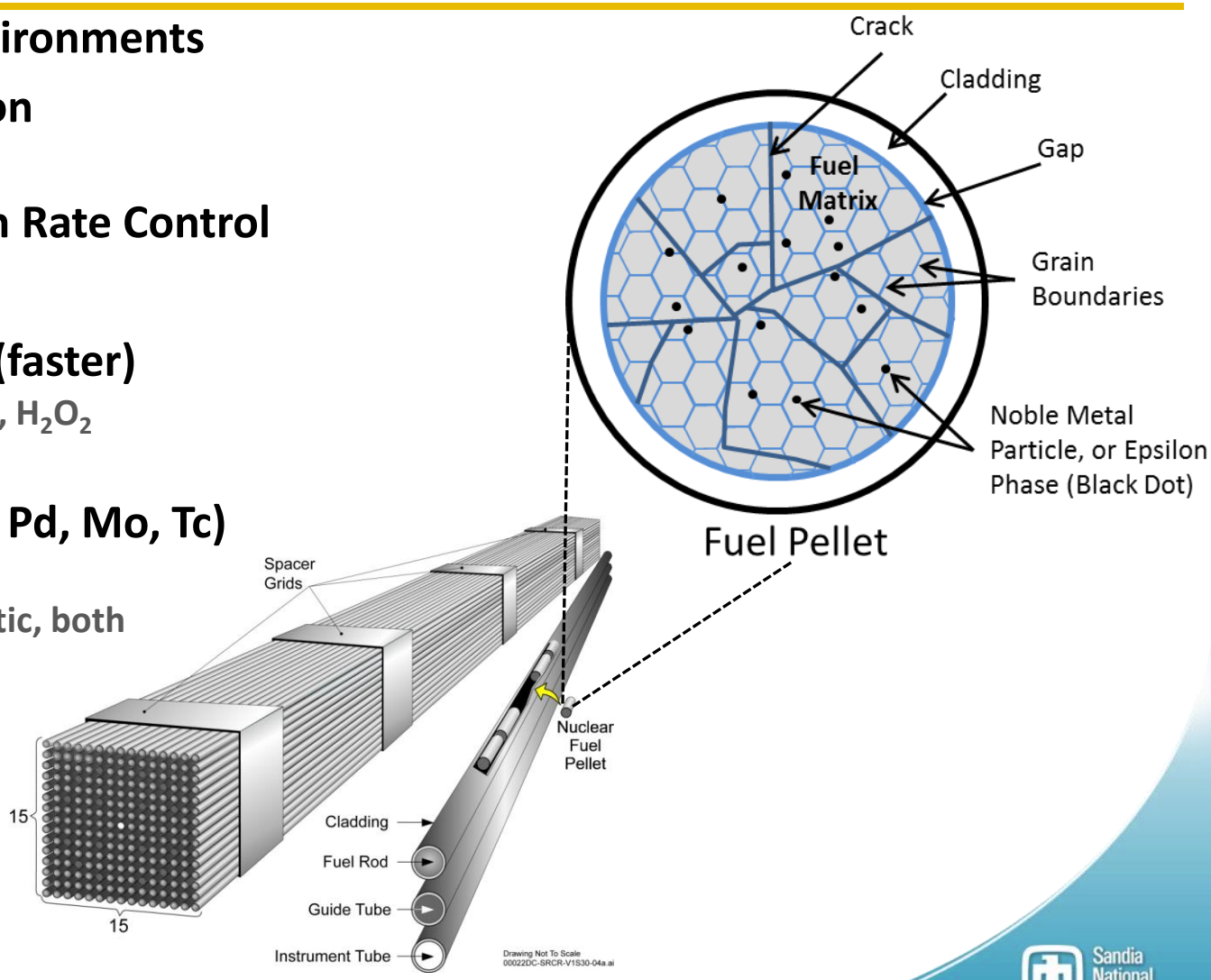
- **DOE's R&D Focus for Used Nuclear Fuel and High Level Waste (HLW) Disposal**
  - Provide sound technical bases
  - Multiple viable disposal options in the US
  - Increase confidence of generic disposal concepts
  - Develop the science and engineering tools
  - Support disposal concept implementation
- **Used Fuel Disposition Campaign Disposal Research and Development Roadmap**
  - Completed March 2011
  - Used to inform prioritization decisions for disposal research in FY12 and beyond



Used Fuel Disposition

Used Fuel Degradation & Radionuclide Mobilization (UFD&RM) Overview

- Reducing Disposal Environments
- Instant Release Fraction
  - At cladding failure
- UF Matrix Degradation Rate Control
- UO_2 Solubility (slow)
- Oxidative Dissolution (faster)
  - Radiolytic oxidants – e.g., H_2O_2
  - Reductants – e.g., H_2
- Epsilon Phase (Ru, Rh, Pd, Mo, Tc)
  - Protects Matrix
  - cathodic coupling, catalytic, both



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Used Fuel Degradation & Radionuclide Mobilization (UFD&RM) Overview (Cont'd)

- **Collaborative Effort – Argonne National Laboratory (ANL) and Pacific Northwest National Laboratory (PNNL)**
  - Carlos Jove-Colon, SNL Work Package Manager - Generic EBS Evaluation
  - William Spezialetti, Technical Lead US DOE (NE-53)
- **Investigate the Long-term Behavior of Used Fuel (UF) as a Waste Form**
  - Develop comprehensive understanding of current technical bases
  - Evaluate range of disposal environments
  - Identify long-term R&D opportunities
- **Integrate with Waste Form Campaign for other waste forms**
  - e.g., high-level waste (HLW) glass
- **International Activity – continued participation as**
  - Associated Group Participation in *FIRST* – nuclides EC Collaborative Project (Fast/Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel)
  - ROK – US Joint Fuel Cycle Study, Fuel Cycle Alternatives Working Group (Task 3)

Used Fuel Degradation & Radionuclide Mobilization (UFD&RM) Overview (Cont'd)

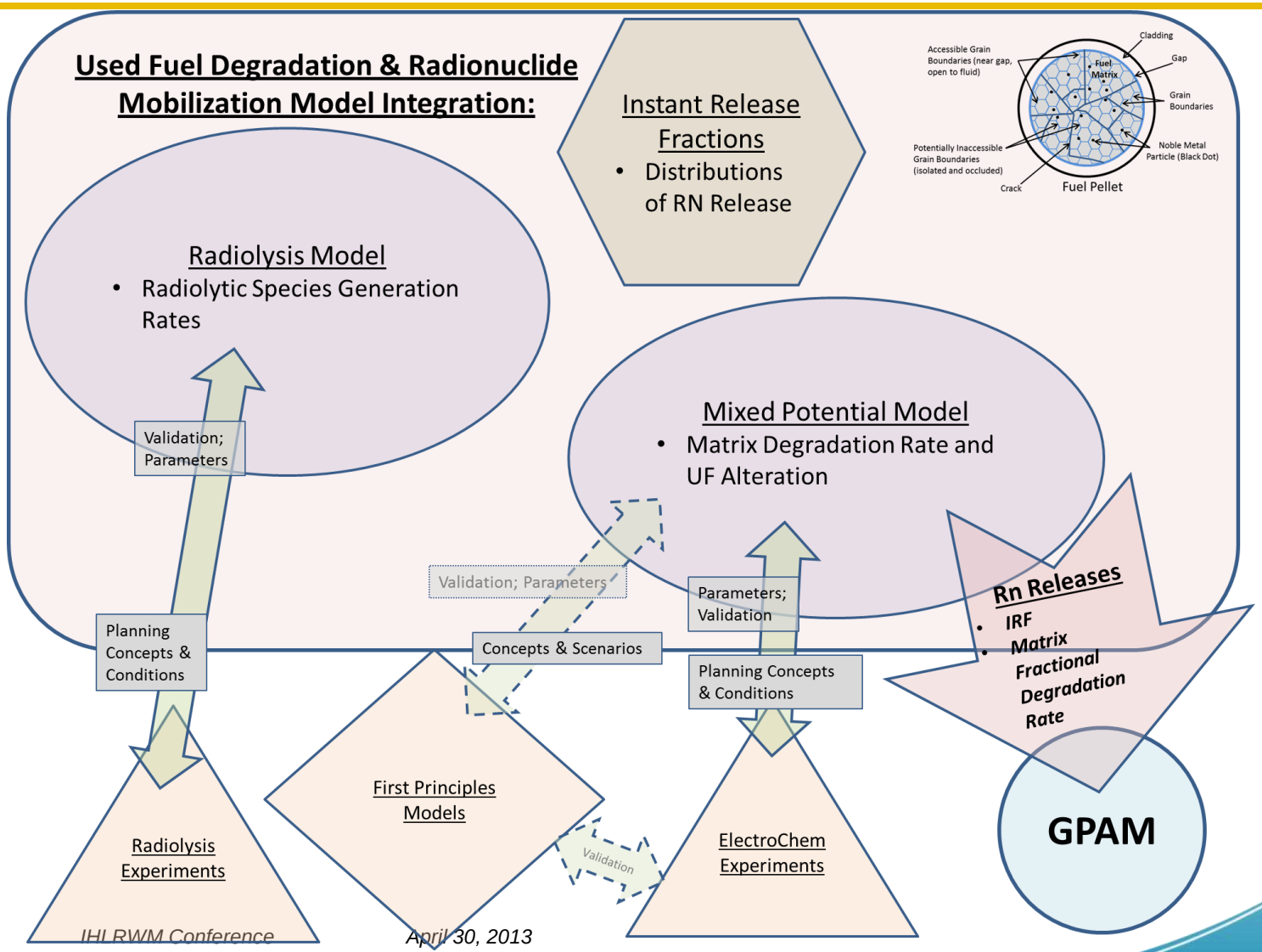
- **Process Modeling**
  - Radiolysis Model (RM) implementation/expansion (PNNL)
  - Mixed Potential Model (MPM) for matrix degradation implementation/expansion (ANL)
- **First principles molecular scale modeling**
  - UO_2, alteration products, and surface reactions (SNL)
  - Epsilon phase and surface reactions (PNNL)
- **Experimental Studies**
  - Electrochemical cell studies to quantify the epsilon phase effects (catalytic/cathodic) on UF matrix degradation (ANL)
  - Radiolytic species generation studies and UF degradation at future conditions (PNNL)
- **Integrating into Generic Performance Assessment Models (GPAM)**
  - Constrain Fast/Instant Release Fractions (IRF) at cladding breach (SNL)
  - Define idealized/flexible strategy for integration with/into GPAM (SNL, ANL, PNNL)
- ***Integration of EBS Models with Generic Disposal System Models*** **- Milestone Report FY12 (FCRD-UFD-2012-000277; Sept. 2012)**

Used Fuel Degradation & Radionuclide Mobilization (UFD&RM) Overview (Cont'd)

- Experimental Programs
- Interface/Coupling RM with MPM
- RM Expansion – Chemical System (e.g., Cl^-)
- MPM Enhancement - Explicit Epsilon Phase Representation
- Sensitivity Analyses RM and MPM
- First-principles Modeling
- UFD&RM Model(s) Implementation into GPAM
- Milestone Report - *Used Fuel Degradation: Experimental and Modeling Report* (October, 2013)

Used Fuel Disposition

UFD&RM Activities and Models Integration Summary



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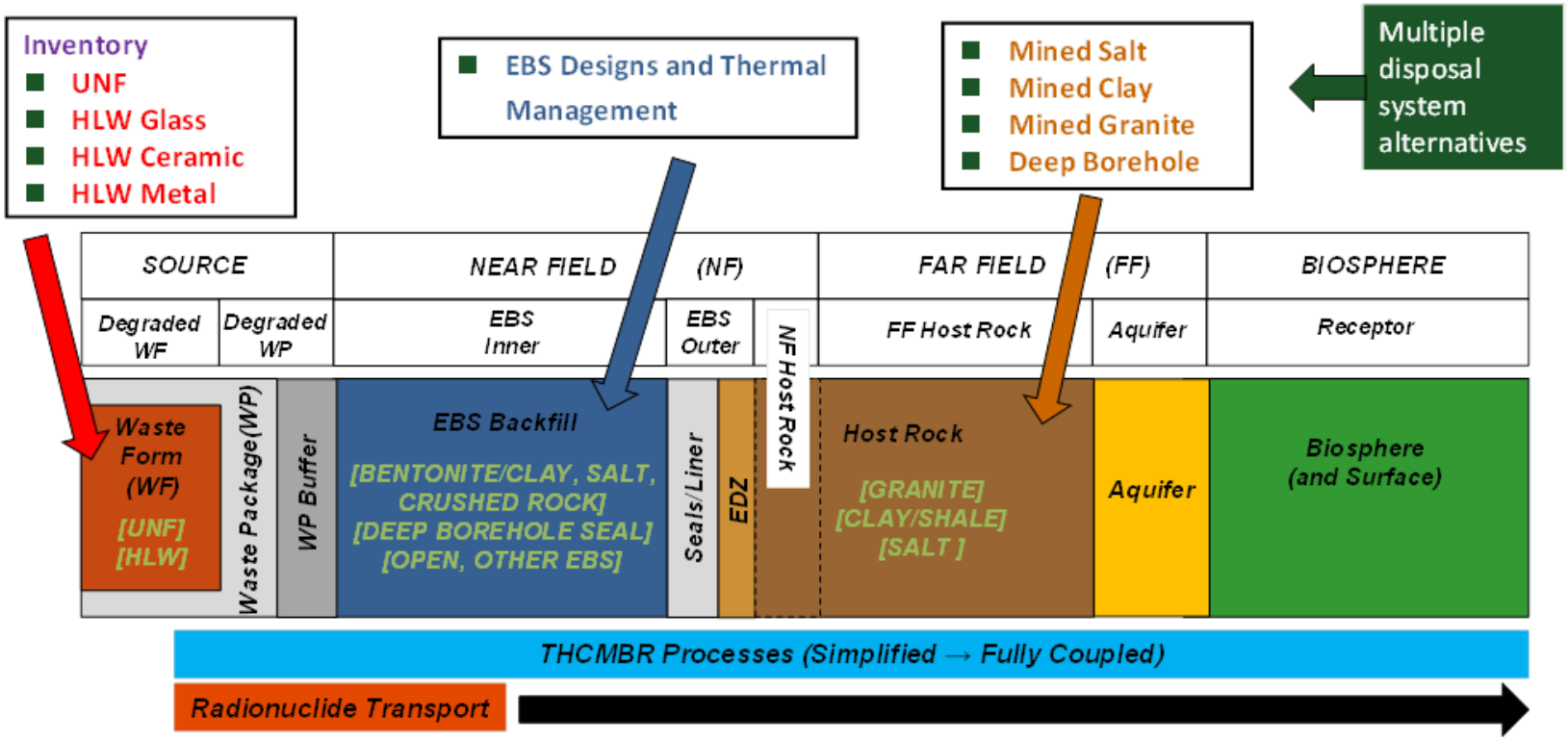
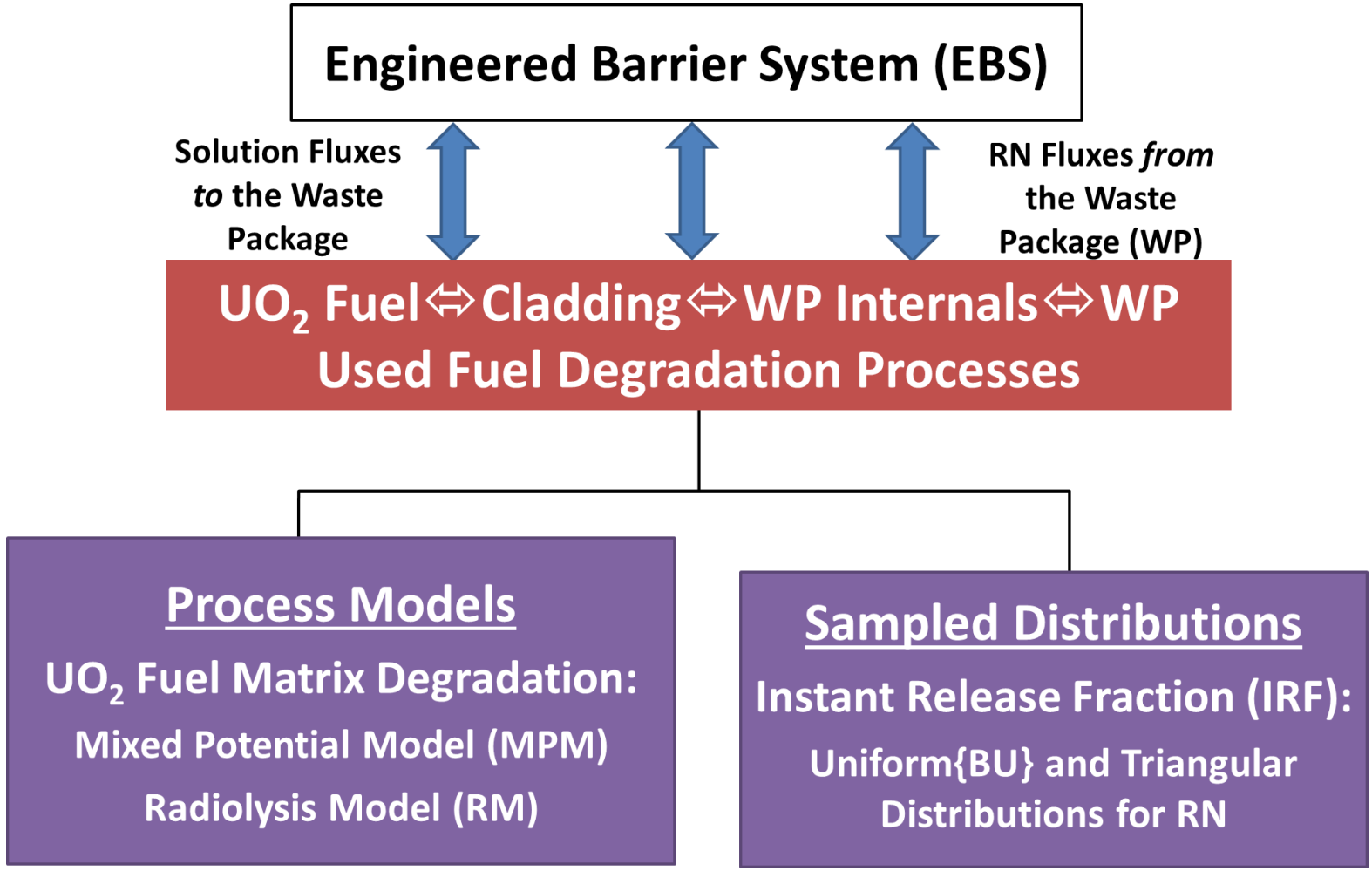
GPAM – Conceptual Framework

- GPAM concepts, including interfaces, features, and processes
- Range of processes and process model fidelities

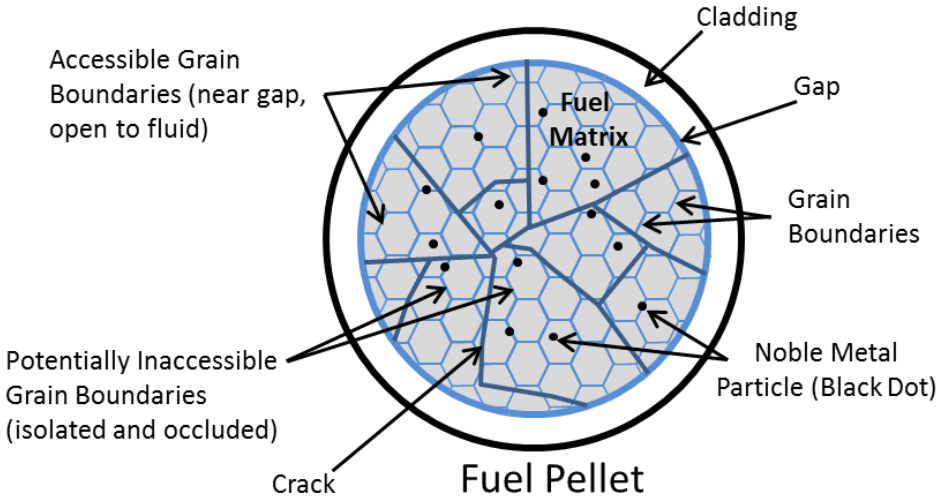


UFD&RM⇔EBS Interface



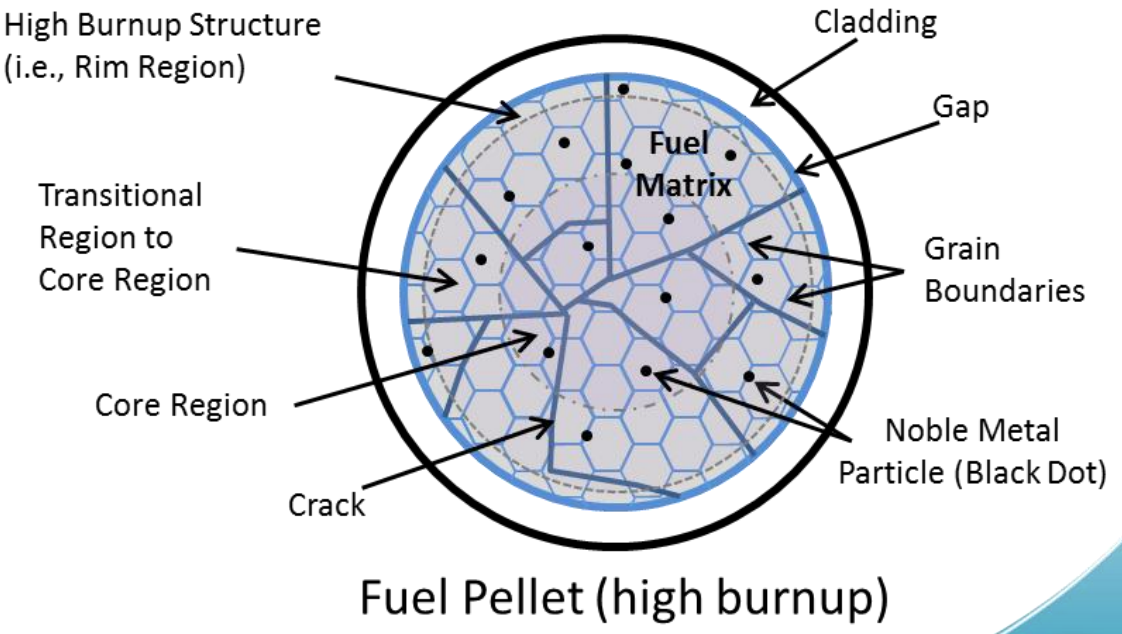
UFD&RM Model Structure and Concepts

- **Used Fuel Degradation and Radionuclide Mobilization Model Concepts**
  - **The *instant release fraction* (IRF) comprised of fission products (including fission gases) located in**
    - *The rod plenum regions (e.g., Kr and Xe)*
    - *The fuel gap (between pellet and cladding*
    - *The accessible grain boundaries/pellet fractures*
  - **The *matrix* inventory that includes the UF matrix itself and radionuclides located in**
    - *The inaccessible grain boundaries/pellet fractures*
    - *Solid solutions (e.g., Pu, Np) within the matrix*
    - *The epsilon phase (noble metal particles)*



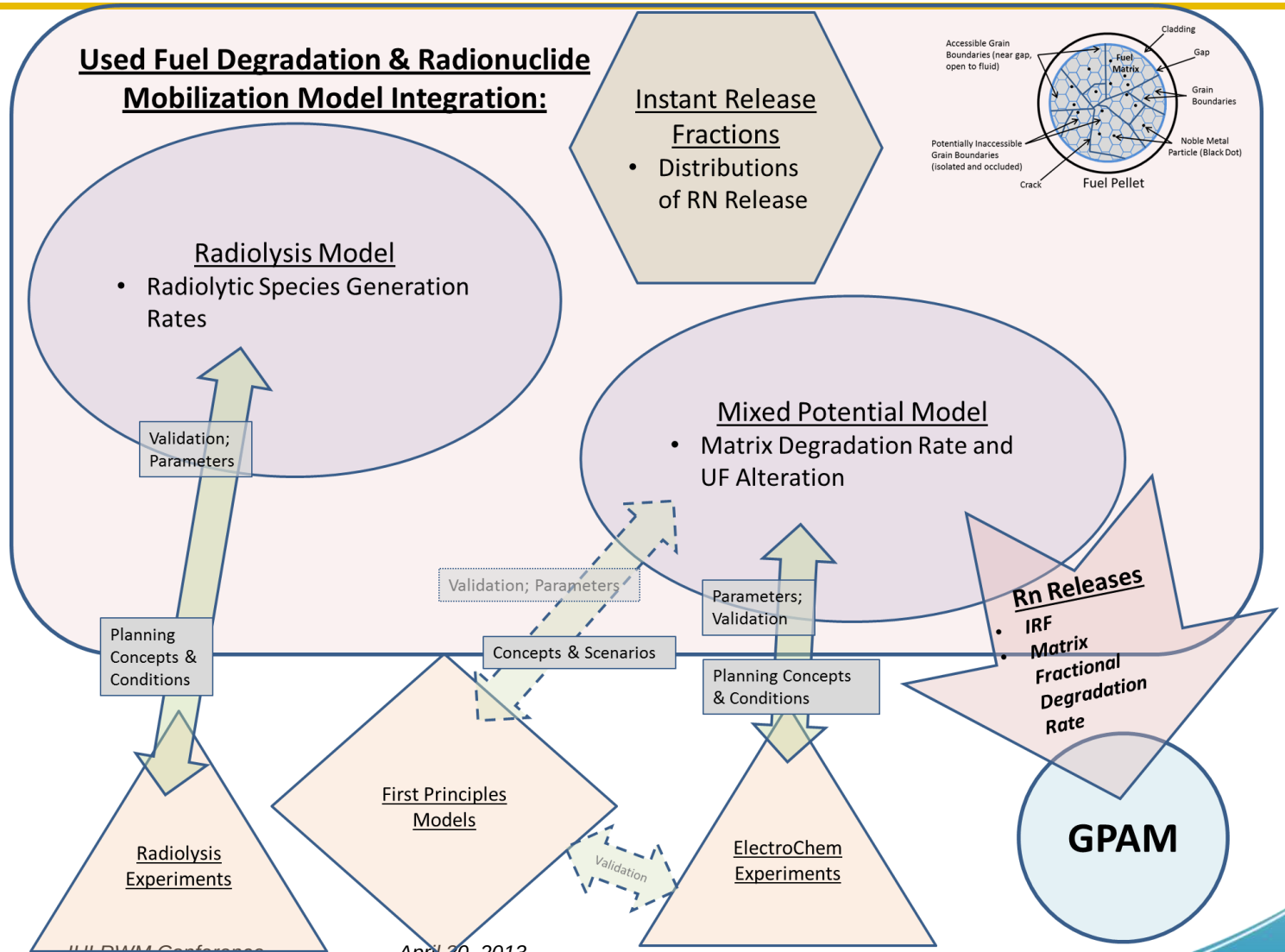
UFD&RM Model Structure and Concepts (cont'd)

- **Future Efforts**
  - Couple physical/chemical degradation of cladding and mechanical evolution of degrading fuel
  - Address specifics of high burnup fuels (>45 MWd/kgU)
  - Mixed oxide used fuels



Used Fuel Disposition

UFD&RM Activities and Models Integration Summary



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UFD&RM Activities Accomplishments/Summary

- **Delineated Constraints for the IRF for UF in Two Sets of Distributions**
  - Triangular distributions (BU at or below 50 MWd/KgU)
  - Uniform distributions (BU up to 75 MWd/KgU)
- **Radiolysis Model**
  - Comprehensive set of radiolysis reactions for H_2O
  - CO_2 speciation
  - Good agreement with other studies
  - Alpha radiolysis for H_2O_2 generation in aqueous solution
- **Mixed Potential Model (Ver. 1)**
  - Verification/Validation with the Canadian MPM
  - Electrochemical and thermodynamic processes (fuel – fluid interface)
  - Hydrogen oxidation reactions
  - Epsilon phase
  - Electrochemical experiments with both UO_2 and "epsilon phase" electrodes

UFD&RM Activities Accomplishments/Summary (cont'd)

- **Computational First-principles Studies of**
  - **UO_2 bulk and UO_2 surface chemistries**
    *(Weck et al., 2012, Dalt. Trans. **41**(32))*
  - **Uranyl peroxide hydrates studtite and metastudtite**
    (Weck et al., 2013, Dalt. Trans. DOI: 10.1039/C3DT32536A)
    - *Good agreement with experimental X-ray diffraction results*
- **Idealized/flexible strategy for GPAM Integration**
  - **Multiple coupling approaches**
  - **Delineate details to couple to other EBS models**
  - **Define GPAM implementation for generic evaluations of the safety case.**