TA

SUBISSUE 3: SPENT FUEL DEGRADATION AND RADIONUCLIDE RELEASE

Presented by
Tae M. Ahn
(301) 415-5812 (tma@nrc.gov)
Division of Waste Management
U.S. Nuclear Regulatory Commission

Technical Contributors
T. Ahn, K. Chan (SwRI), R. Codell, J. Contardi, G. Cragnolino (CNWRA), C. Greene, O. Pensado (CNWRA), and N. Sridhar (CNWRA)

Appendix 7 Meeting: Container Life and Source Term
July 7-8, 1999, Lawrence Livermore National Laboratory, Livermore, CA

OUTLINE OF PRESENTATION

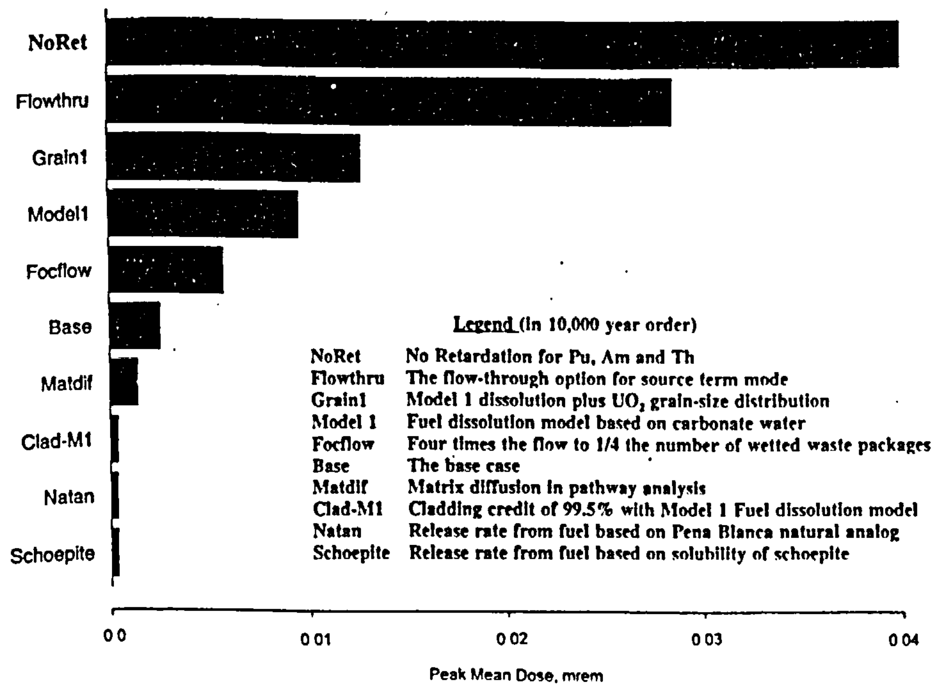
- NRC TPA3 SENSITIVITY ANALYSIS
- ACCEPTANCE CRITERIA
- RESOLVED TOPICS
- DETAILED TOPICS:
  - Modes of Water Contact
  - Dissolution Rate
  - Chemistry inside Waste Package
  - Update
  - Colloid
  - Dry Oxidation
  - Cladding
  - Release Scenarios
  - Disruptive Events
- CONCLUSIONS

NRC TPA3 SENSITIVITY ANALYSIS

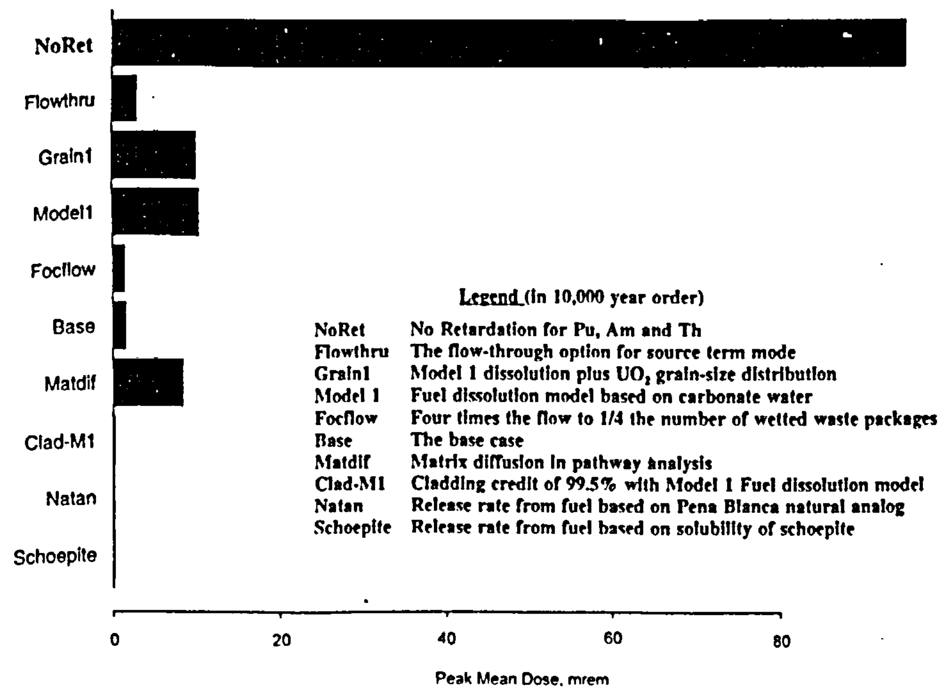
- **Sensitive Parameters: Surface Area/Grain Boundary Penetration, Retardation (Colloids?), and Cladding**

- **Figures**

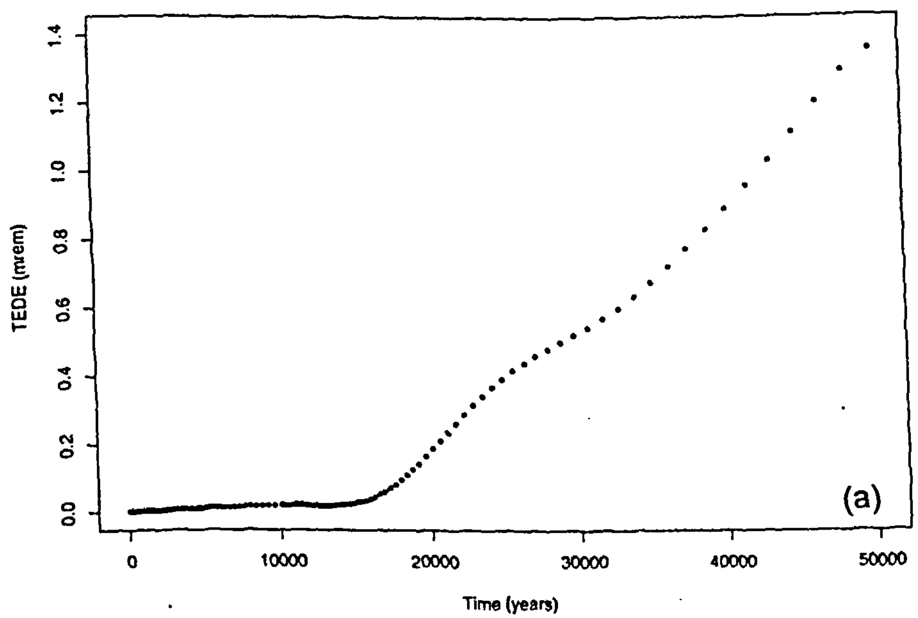
Peak Mean Dose for 10,000 Years

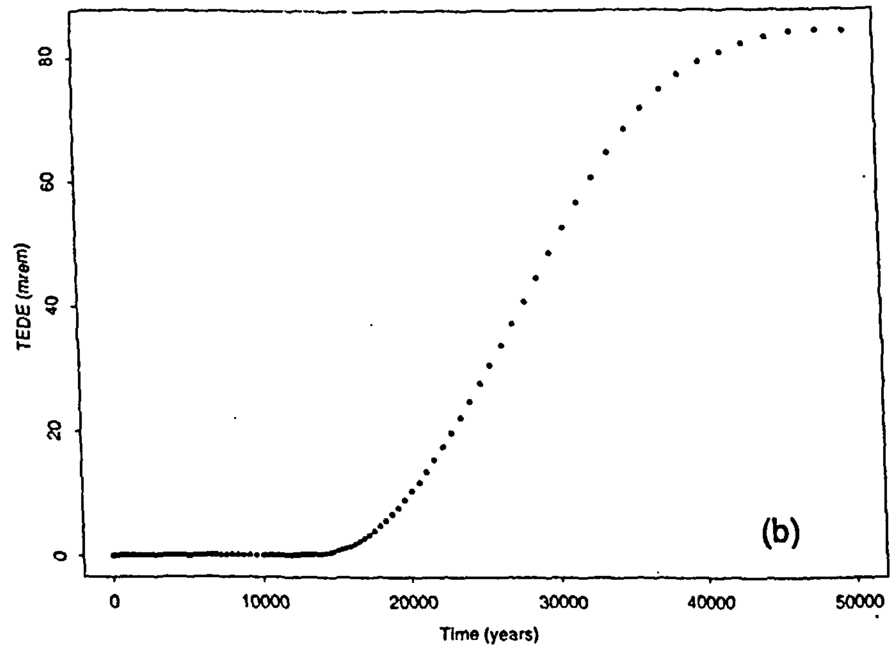


Peak Mean Dose for 50,000 Years



- No Retardation of Pu (Potential Colloid Contribution): TPA3.2 Outputs
(a) Nominal Case (McCartin, 1999) (b) No Retardation





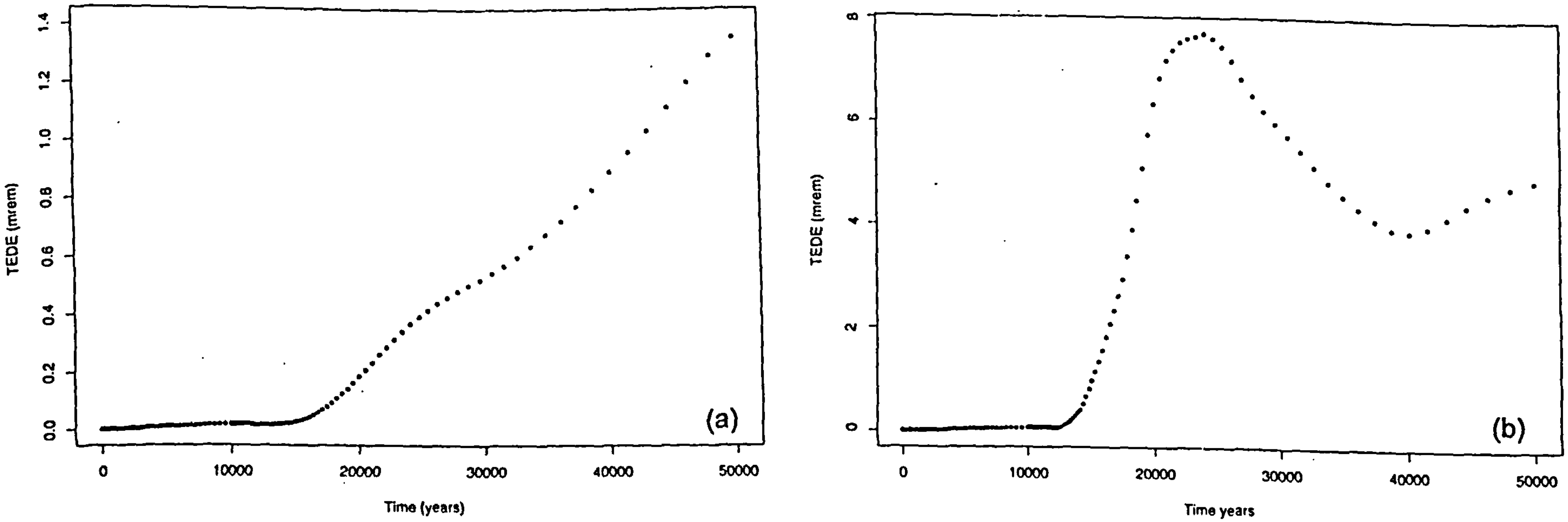


Figure. TPA3.2 Outputs (a) Nominal Case of particle model (McCartin, 1999)
(b) Grain Model (Contardi, 1999)

ACCEPTANCE CRITERIA

(1) DOE has considered all categories of SNF planned for disposal at the proposed YM repository.

(2) DOE has identified and considered likely processes for SNF degradation and the release of radionuclides from the EBS, as follows: dissolution of the irradiated UO_2 matrix, with the consequent formation of secondary minerals and colloids; prompt release of radionuclides; degradation in the dry air environment; degradation and failure of fuel cladding; preferential dissolution of intermetallics in DOE SNF; and release of radionuclides from the WP emplacement drifts.

(3) DOE has demonstrated that the numerical models used for SNF degradation and radionuclide release from the EBS are adequate representations, including consideration of uncertainties, of expected SNF performance and are not likely to overestimate the actual performance in the repository environment.

(4) DOE has considered the compatibility of SNF and the internal components of the WP, such as the basket materials in the evaluation of radionuclide releases. Specifically, the SNF should not compromise the performance of the WP.

(5) DOE has justified the use of SNF test results not specifically collected for the YM site for the environmental conditions expected to prevail after breaching of the containers at the YM site.

(6) DOE has conducted a consistent, sufficient, and suitable corrosion testing program at the time of the LA submittal. In addition, DOE has identified specific plans for further testing to reduce any significant area(s) of uncertainty as part of the performance confirmation program.

(7) DOE has established an adequate program of monitoring radionuclide release from the WP during the performance confirmation period, to assure that assumptions and calculations of SNF dissolution and radionuclide release SNF are appropriately substantiated.

RESOLVED TOPICS

- Type of Spent Nuclear Fuel
- Inventory and Distribution of Radionuclide
- Dry Oxidation
- Dissolution Process in General

DETAILED TOPICS

- Modes of Water Contact

(A) Types

- Bathtub (Immersion)

- Drip

- Vapor

(B) Conservatism of the scenario and simulation method (e.g., flow-through test) should be rationalized.

(C) Consistency among various test methods should be demonstrated.

(D) Various thermal loading and backfilling are not well considered.

- **Dissolution Rate**

- Grain boundary opening (Table, Gray, 1997)

Dissolution Rate ($mg \bullet m^{-2} \bullet d^{-1}$) and Estimated Grain Boundary Penetration of Unoxidized (UO_2) and Oxidized (U_4O_{9+x}) Spent Fuel

	Unoxidized			Oxidized		
Fuel	Grains	Particles	GBP[(a)]	Grains	Particles	GBP[(a)]
ATM-104[(b)]	3.4	33	4-6	3.5	166	~100
ATM-106[(b)]	1.5	25	6-9	8.2	241	12-18
ATM-105[(c)]	6.6	25	2-3	7.4	28	2-3

(a) Grain Boundary Penetration. Estimate of depth of water penetration into the grain boundaries (number of grain layers)

(b) 2×10^{-2} M total carbonate, pH =8, 25°C, atmospheric oxygen partial pressure

(c) 2×10^{-3} M total carbonate, pH =9, 50°C, atmospheric oxygen partial pressure

- Fuel type such as burnup, preoxidation (e.g., archived sample)

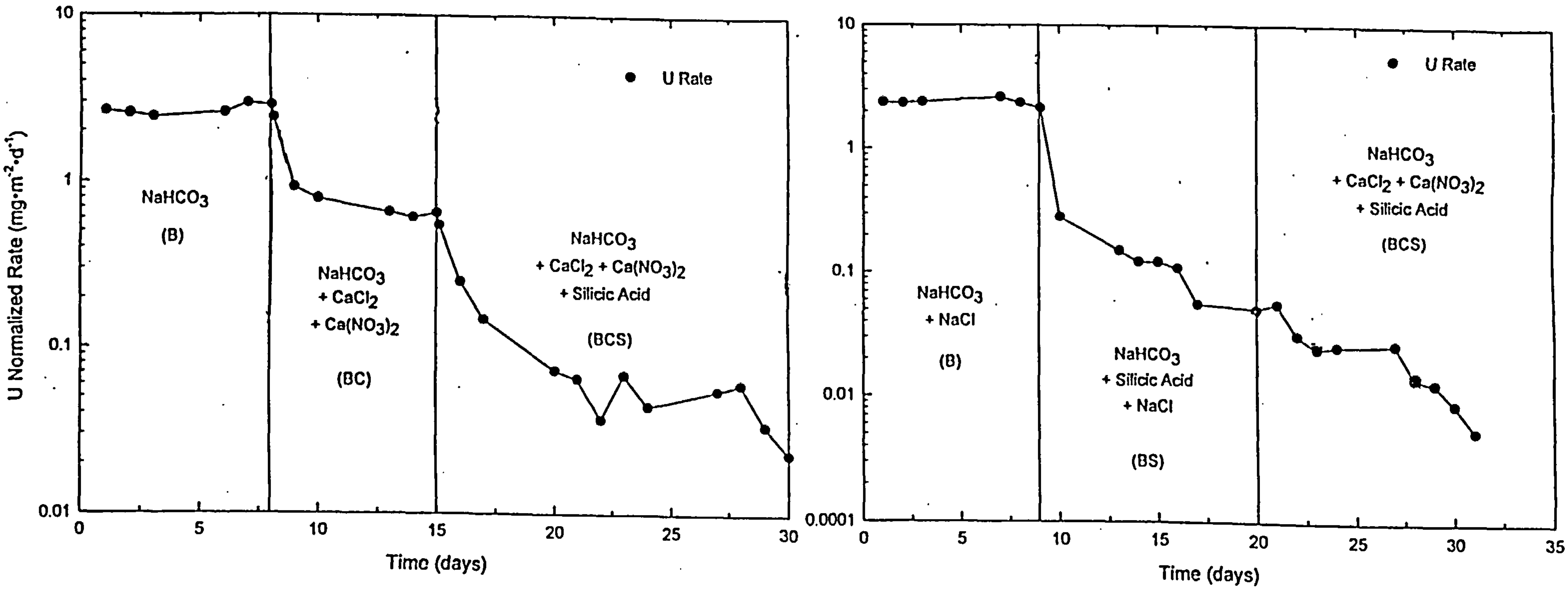


Figure. Effects of Solution Composition on Dissolution Rate, Flow-through Tests of 44 ~ 105 μm UO_2 at 25 °C (Gray and Wilson, 1995)

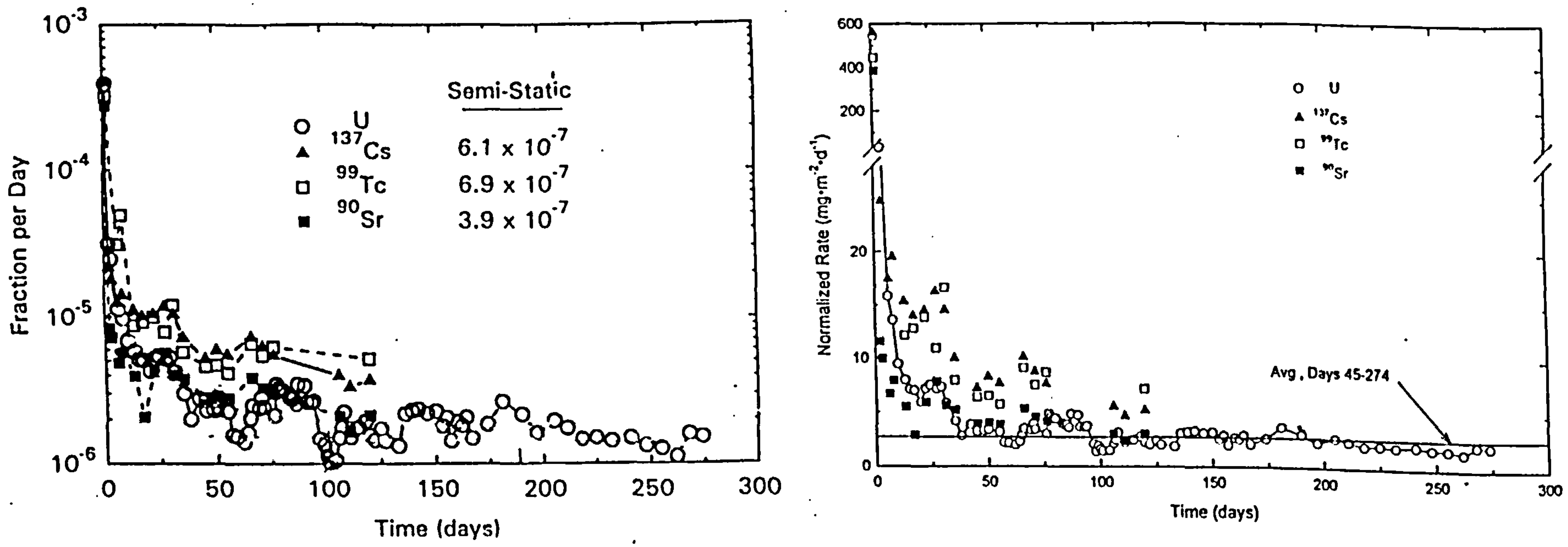


Figure. Spent-Fuel Dissolution Rates of Archived Particles, Flow-through Tests at 25 °C (Gray and Wilson, 1995; Gray, 1992; Wilson, 1990)

- Contribution of grain boundary inventory: semistatic tests and drip tests (Finn et al., 1996)

Species	High Drip Rate		Low Drip Rate		Vapor		Semistatic [9]	
	ATM-103	ATM-106	ATM-103	ATM-106	ATM-103	ATM-106	TP	ATM-101
^{238}U	8E-5	2E-4	2E-6	1E-5	4E-8	4E-7	1E-4	1E-4
^{239}Pu	8E-6	1E-4	1E-5	2E-5	1E-7	3E-7	1E-4	1E-4
^{237}Np	1E-3	1E-4	4E-5	6E-5	7E-7	6E-7	1E-4	1E-4
^{241}Am	3E-3	2E-4	4E-4	1E-4	3E-6	7E-7	1E-4	2E-4
^{244}Cm	7E-3	3E-4	2E-3	4E-4	2E-5	3E-6	1E-4	2E-4
^{137}Cs	2E-3	3E-3	1E-5	1E-6	7E-7	4E-6	5E-3	1E-2

[a]The error bars for ^{137}Cs are ±0.5%. The error bars for the actinides are ±50%. Results do not account for material incorporated into alteration products or sorbed on the fuel.

[b]These are for fractions for the first 1.6 years of reaction.

[c]Three cycles (460 d) at 85°C for Turkey Point (TP) fuel, 27 (MW•d)/kg U and a fission gas release of 0.3%; and two cycles (360 d) for ATM-101 fuel, 30 (MW•d)/kg U and a fission gas release of 0.2%.

- Effects of Ca and Si, secondary minerals, and partial cladding protection

(1) Activation Energies are from immersion tests (Wilson, 1990)

(2) Three groups of dissolution rate

- J-13 well water, synthetic groundwater, granitic groundwater, tap water, and distilled water: (2.4×10^{-4} ~5.4) mg m^{-2} d^{-1} at room temperature (RT)
- chloride solution: (5×10^{-3} ~5.7) mg m^{-2} d^{-1} at RT
- carbonate solution: (0.23 ~ 3.3) at RT

(3) Tests of particles may increase the dissolution rate by as high as a factor 10 compared with grain tests, but the difference depends on (a) details of sample types such as size or oxidation state, (b) spent-fuel types such as fresh, archived, or different burnup, and (c) contribution of grain boundary inventory.

- The use of accelerated tests in pure carbonate solutions introduced additional uncertainties from cladding protection and low juvenile failure of container. From the system's engineering view, this is not desirable.

- **Chemistry inside Waste Package**

  - High Concentration of Chloride and Metal Chloride Complexes

    - Sustaining localized corrosion requires presence of metal-chloride complexes at a concentration of about 15 percent of saturation. Hydrolysis of metal cations leads to extremely acidic pH values.
    - Dilution of this solution leads to cessation of pit growth.
    - WP internal environment is packed and hence has many crevices where high concentration electrolytes will prevail.

  - Oxidizing Conditions

    - Alpha radiolysis will create highly oxidizing conditions close to the surface of spent fuel
    - The packed regions near the fuel may have other oxidized species such as Fe^{+3}

  - Currently there is insufficient understanding of the range of environments that may be present inside the WP: localized concentration and bulk dilution. Modeling and experimental tools exist to investigate the extent of variation of internal environment so that SF dissolution calculations may be more realistic

- Update:

  grain boundary inventory, important radionuclides in PA, Np solubility

- Colloids:

  The values for the parameters are either unknown or contained large amounts of uncertainties. Why not assume the conservative case of total irreversibility?

- Dry Oxidation:

  Radionuclide release from the juvenile container failure is an important contributor to dose. In using realistic dissolution model, dry oxidation in defective waste package will alter the dissolution rate of spent fuel by opening up grain boundaries.

- Cladding

  - Damage or failure during storage or transportation is not well known. For example, the temperature may go up near 500 C during the transportation

  - Damage during the reactor operation is not well known. Interagency research is initiated for spent fuel stored at INEEL.

  - Localized corrosion data

  - Hydride embrittlement, creep, stress corrosion cracking, and clad- splitting by matrix oxidation need to be reevaluated.

- Release Scenarios

- The role of perforation is not well understood: controlled release from limited diffusion path (Table, Ahn et al., 1998) or accelerated release by the formation of severe chemistry

Reduction of Radionuclide Release from Pinhole (~0.02 cm Diameter) or from Slit - **(~0.015 Diameter x ~2.54 cm Length) Defected Cladding in J-13 Well Water. Ratios are Radionuclide Releases from defected-cladded Spent-Fuel to that from Bare Spent-Fuel Matrices. Data from Wilson (1990)**

Radionuclide	Reduction in Release
Np-237	~ 1/70
Tc-99	~ 1/140
I-129	~ $1/(7 \times 10^5)$
Sr-90	~ 1/65
Pu-(239+240)	~ $1/(7 \times 10^4)$
Am-241	~ $1/(3 \times 10^3)$

Data were selected for Cycle 2 and Cycle 3. The Cycle 1 data was the first semi-static immersion test and included the release of spent-fuel grains and the gap inventory. Cycle 2 tests were retests of first-cycled samples that had been placed in new leachant after removing the altered layer. In general, tests after Cycle 2 showed little gap inventory if it was not of a large amount initially. Detection limits of radionuclides were used in the calculation when hole-defected or slit-defected samples did not release detectable radionuclides.

- **Disruptive Events:**

  - U_3O_8 formation will facilitate the direct release under volcanic conditions.

  - Disruptive events including seismicity, faulting and igneous activities may lead to the about 0.1 % container (conservatively cladding too) failure. This is the same order of magnitude of juvenile failure rate of container

SUMMARY

(1) The dose is sensitive to surface area, dissolution model, Pu retardation (colloids), and cladding protection.

(2) Acceptance criteria were presented.

(3) Detailed topics discussed include mode of water contact, dissolution rate models, chemistry inside waste package, grain boundary inventory, colloids, dry oxidation, cladding, release scenarios, and disruptive events.