U.S. DEPARTMENT OF ENERGY
OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT

NUCLEAR WASTE TECHNICAL REVIEW BOARD
FULL BOARD MEETING

SUBJECT: OVERVIEW OF SPENT FUEL MODELING CONCEPTS

PRESENTER: DR. RAY B. STOUT

PRESENTER'S TITLE AND ORGANIZATION: TECHNICAL AREA LEADER, WASTE FORM CHARACTERIZATION
LAWRENCE LIVERMORE NATIONAL LABORATORY (LLNL)
LIVERMORE, CALIFORNIA

PRESENTER'S TELEPHONE NUMBER: (510) 422-3965

PLAZA SUITE HOTEL • LAS VEGAS, NEVADA
OCTOBER 14 - 16, 1992

Spent Fuel Characterization Activities

Objective:

- To provide data, testing, and models for the physical properties, the degradation responses, and the radioactive release responses of spent-fuel waste forms for waste package and system performance assessments in the Yucca Mountain Project

Product:

- Preliminary Waste Form Characteristics Report [MO3]

Contents are the available

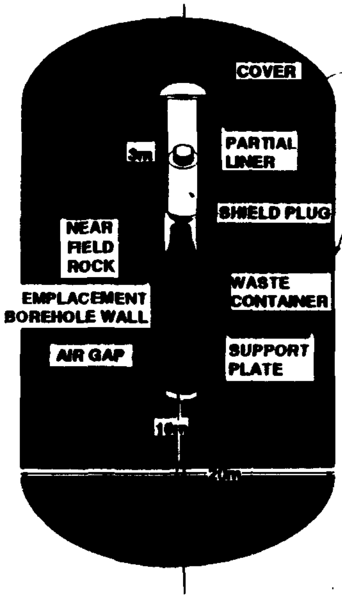
- Physical property data for existing and projected SFWP & DHLW inventories
- Radionuclide data for existing and projected SFWP & DHLW inventories
- Test data and models for potential release rates from SFWP & DHLW

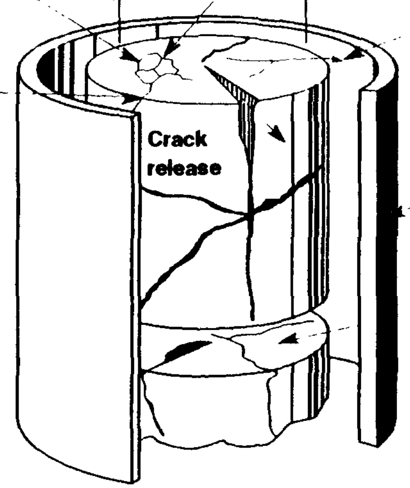
Outline

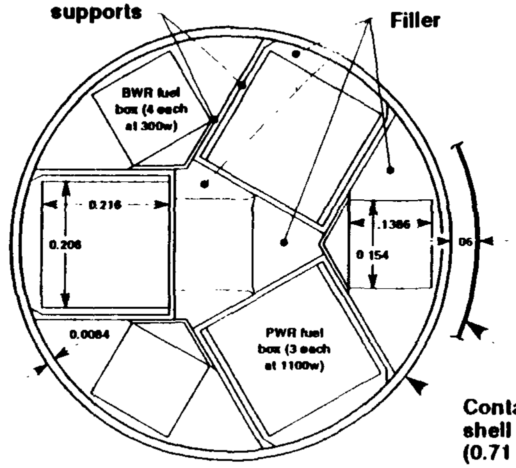
- Introduction: Spent fuel characteristics
- Release modes: gaseous and aqueous
- Models for test matrix design and for preliminary design: Performance Assessment (PA)
    - Gaseous
    - Cladding
    - Oxidation
    - Dissolution
    - Hardware
- Summary

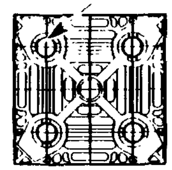
Spent Fuel Model Development: Problem of Length Scale

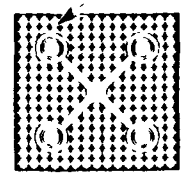
Illustration of conceptual waste package and portions of the EBS

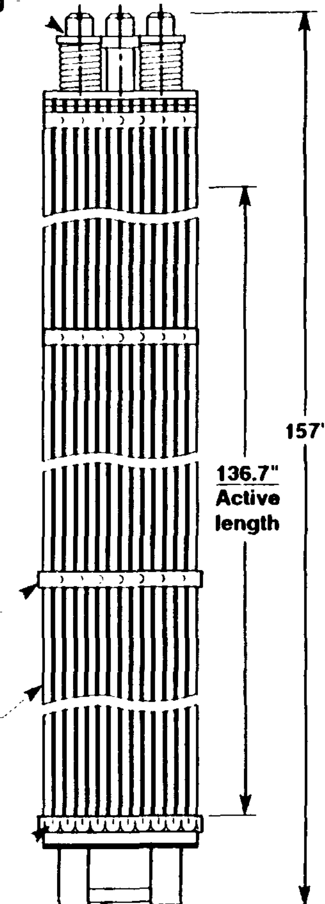












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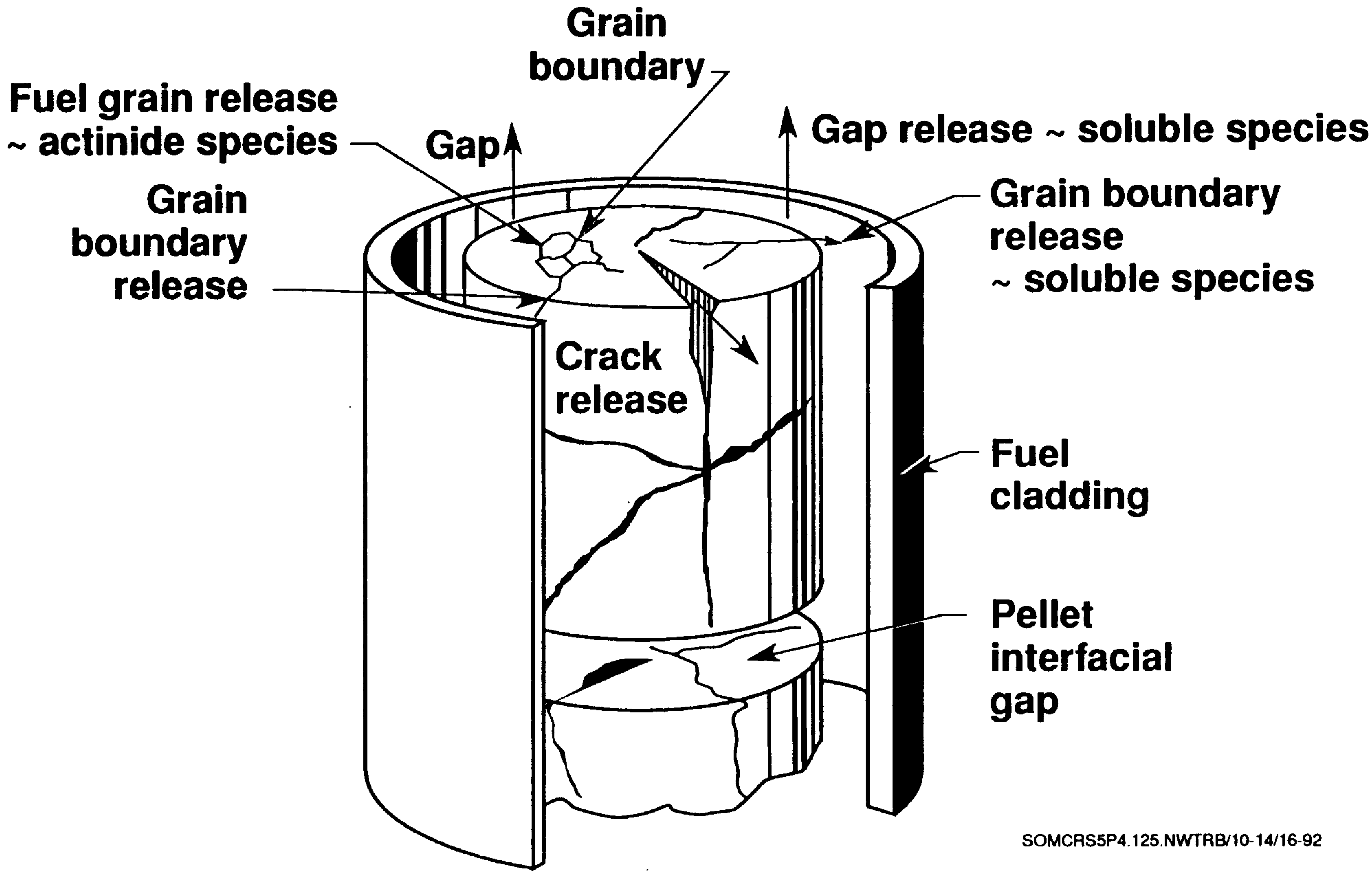
Spent Fuel Features Important to Oxidation and Release Responses

Illustration of the cladding gap and the fragment surfaces.
Fragment surface consists of grain boundary and grain areas.



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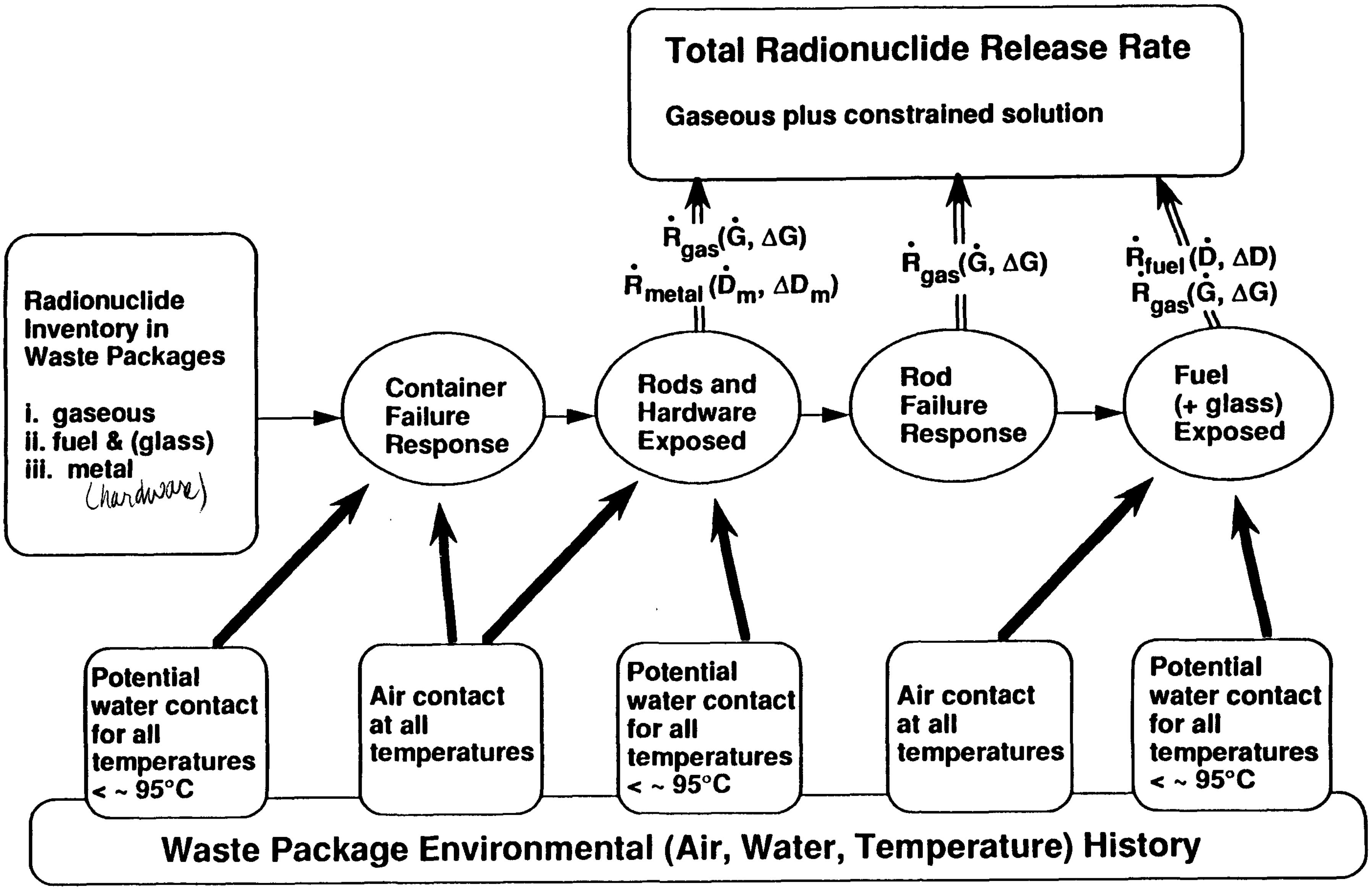
Source Term Release-Rate Diagram



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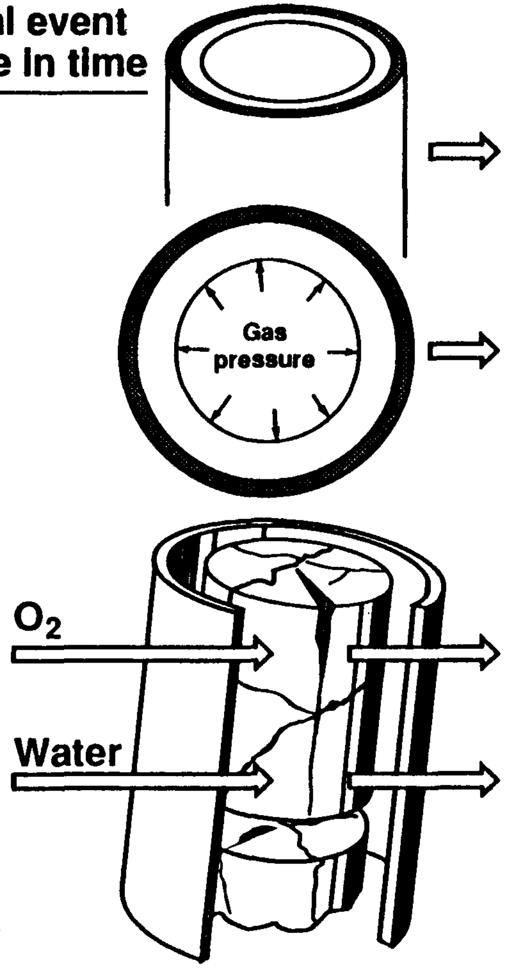
Spent Fuel Responses

Potential event sequence in time

t
time



Gaseous release response from cladding	— Carbon [14] release	— $\dot{G}$ and ΔG	
Cladding degradation response	— Rod failure time response	— $\dot{R}_f$	
UO_2 oxidation response	— Phase change kinetics	— P_{NM}, eg	$P_{12} \sim UO_2$ $P_{49} \sim U_4O_9$ $P_{38} \sim U_3O_8$
	— Oxidation response	— $\dot{O}$	
UO_2 dissolution response	— Dissolution response	— D (gm/m^2/day) and ΔD	
Geochemistry solution response	— Solubility limits and Colloidal response	— S_L and C_R	

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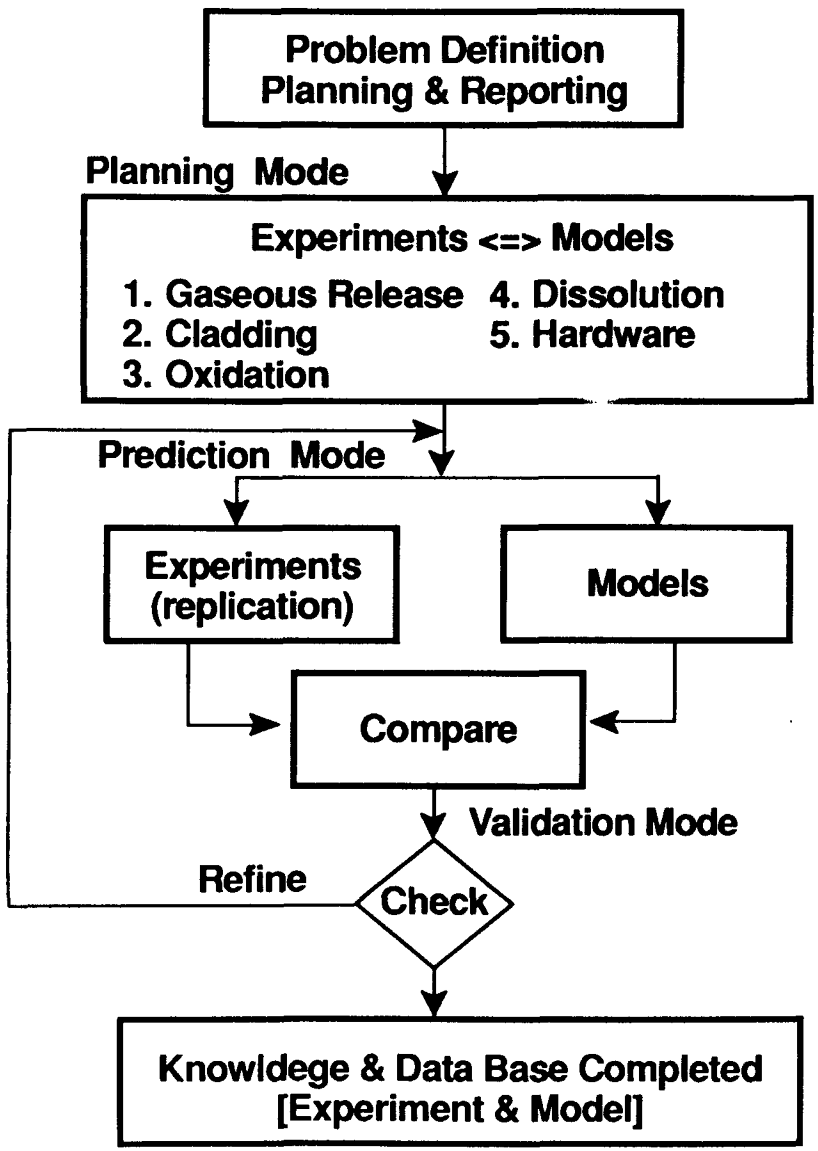
Spent Fuel: Knowledge & Data Base Activities



Main topics - Spent Fuel

1. Gaseous Release Response
2. Cladding Failure Response
3. Oxidation Response
4. Dissolution Response
5. Hardware Release

Pre-advanced Conceptual Design & Assessments: Preliminary Waste Form Characteristics Report v. 1.0

Advanced Conceptual Design & Assessments: Waste Form Characteristics Report v. n.0

License Application Design: Waste Form Characteristics Report

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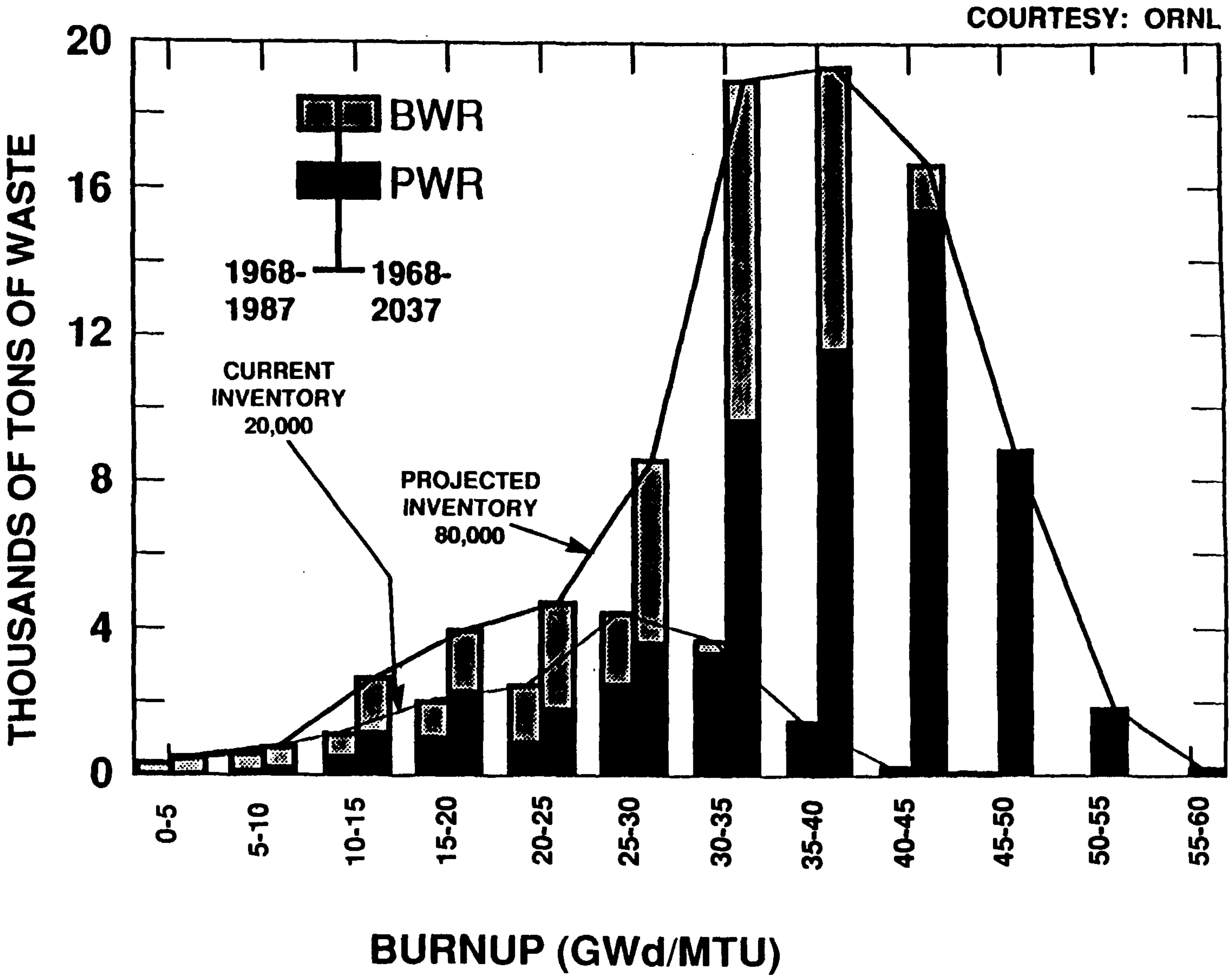
Spent Fuel Inventory: History and Projection



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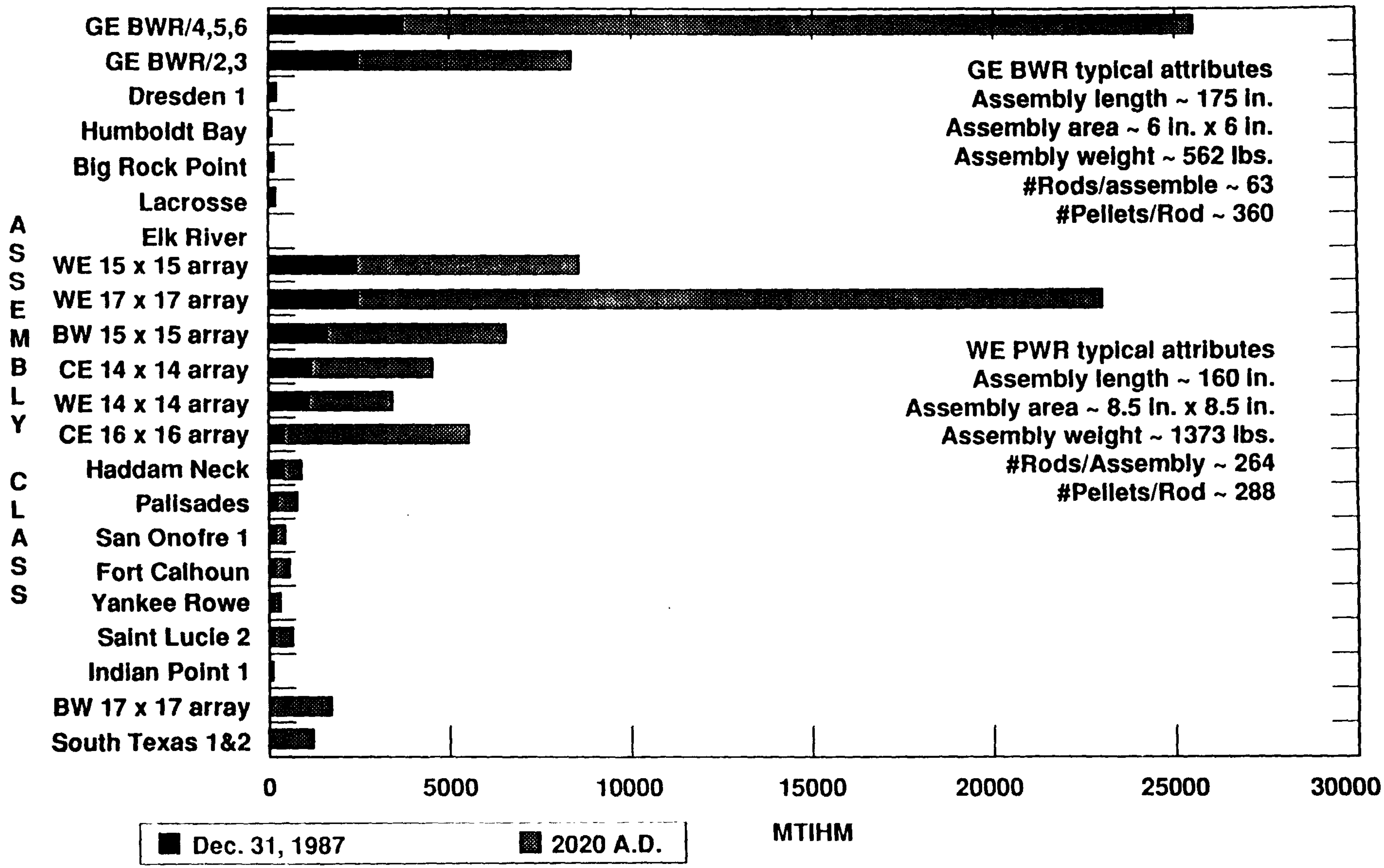
Assembly Class Quantities and Typical Dimensions



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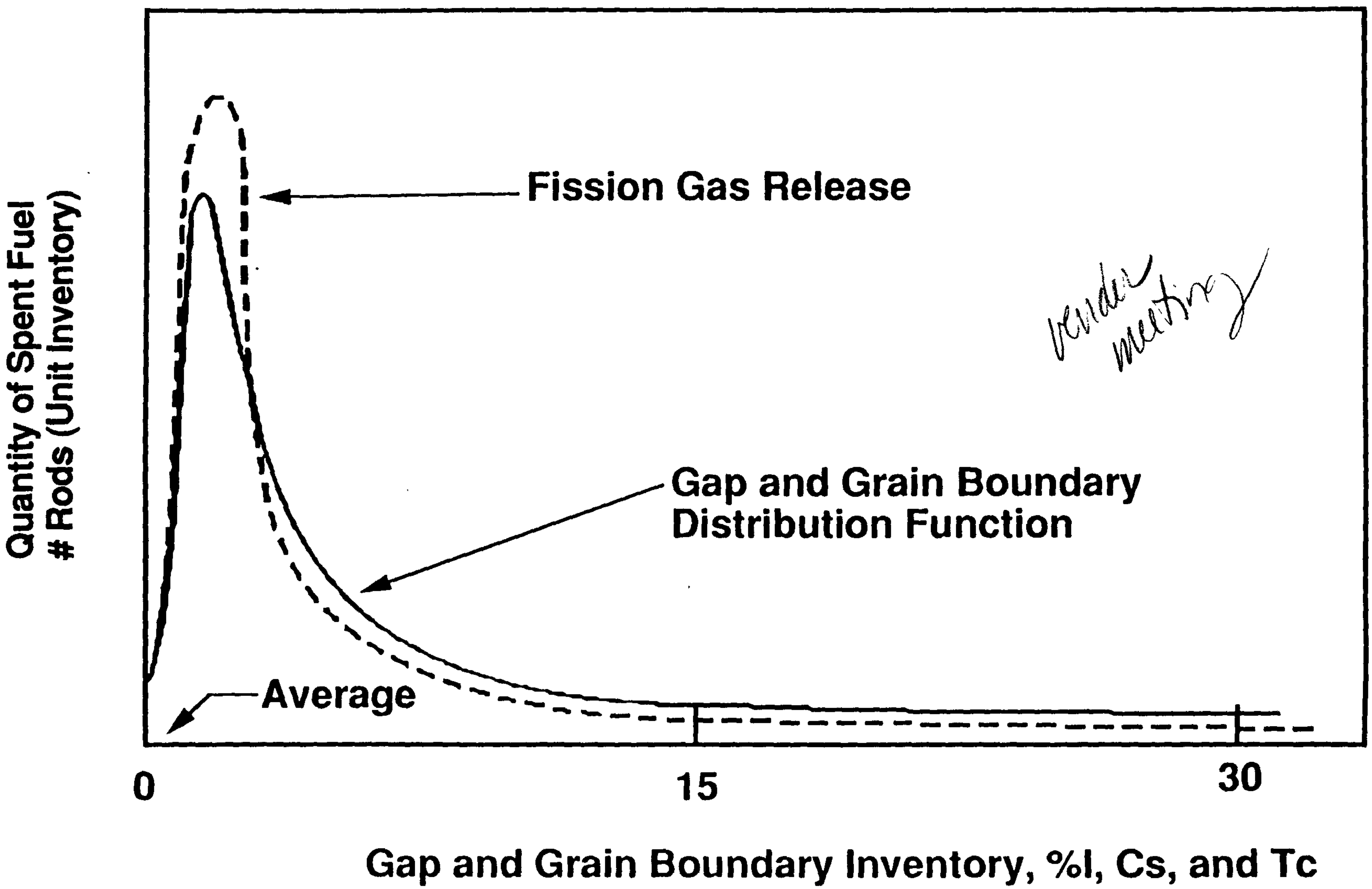
Illustrative Rod Population Distribution of Gap and Grain Boundary Inventory



vendor meeting

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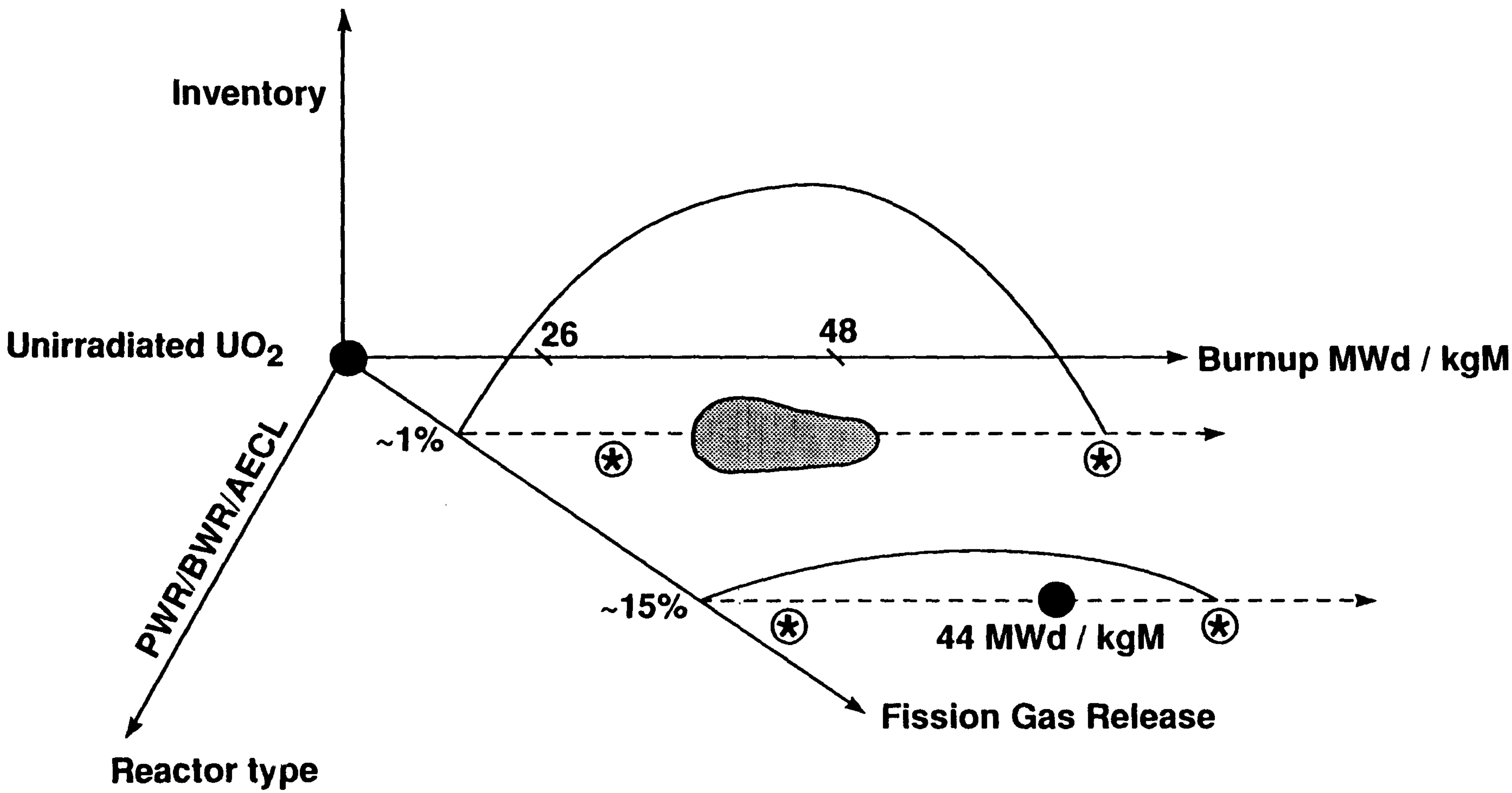
Spent Fuel Inventory Attributes for Test Matrix (Illustrative)



⊛ Future SF testing inventory:
low and high burnup with low and high FGR.

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Gaseous Release

$$\dot{R}\text{elease}_{(gas)} = C \otimes R_o \otimes \dot{G}_{(rod)} + C \otimes R \otimes \dot{G}_{(fuel)} + \dot{C} \otimes R_o \otimes \Delta G_{(rod)} + C \otimes \dot{R} \otimes \Delta G_{(fuel)}$$

Waste Form Problems ... provide response functions (models) for

R, $\dot{R}$, — rod failure response

$\dot{G}_{(rod)}$ and $\dot{G}_{(fuel)}$ . . . gaseous release rate

$\Delta G_{(rod)}$ and $\Delta G_{(fuel)}$ . . . rapid release increment

<u>Primary Concern</u> – Carbon-14

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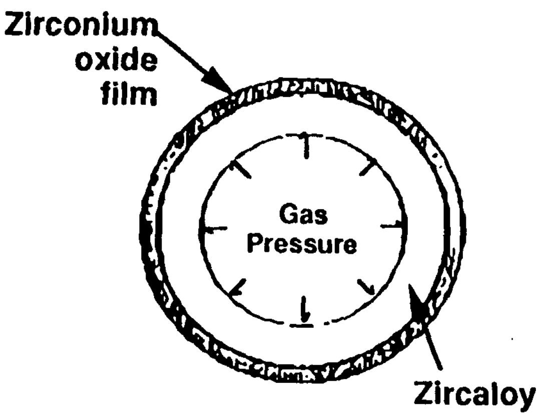
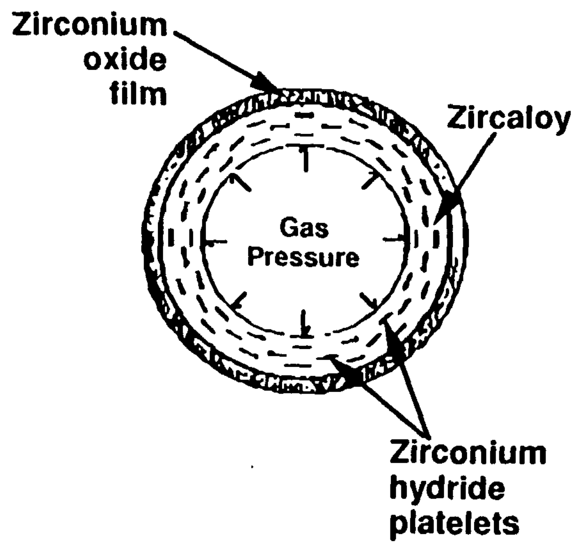
Zircaloy Cladding Degradation

Oxide film failure – stress corrosion cracking inhibitor

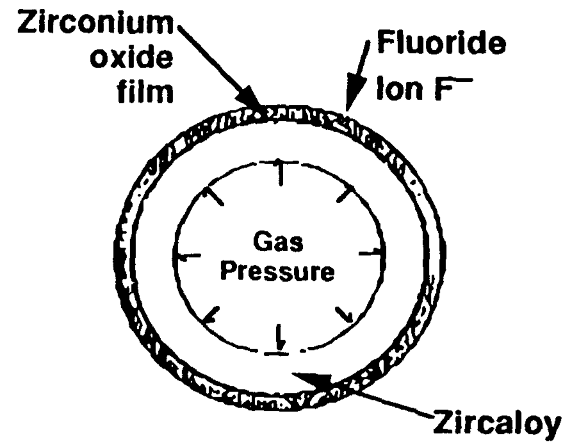


Thin-walled tube
Radius/thickness ~8

Hydride platelet precipitation

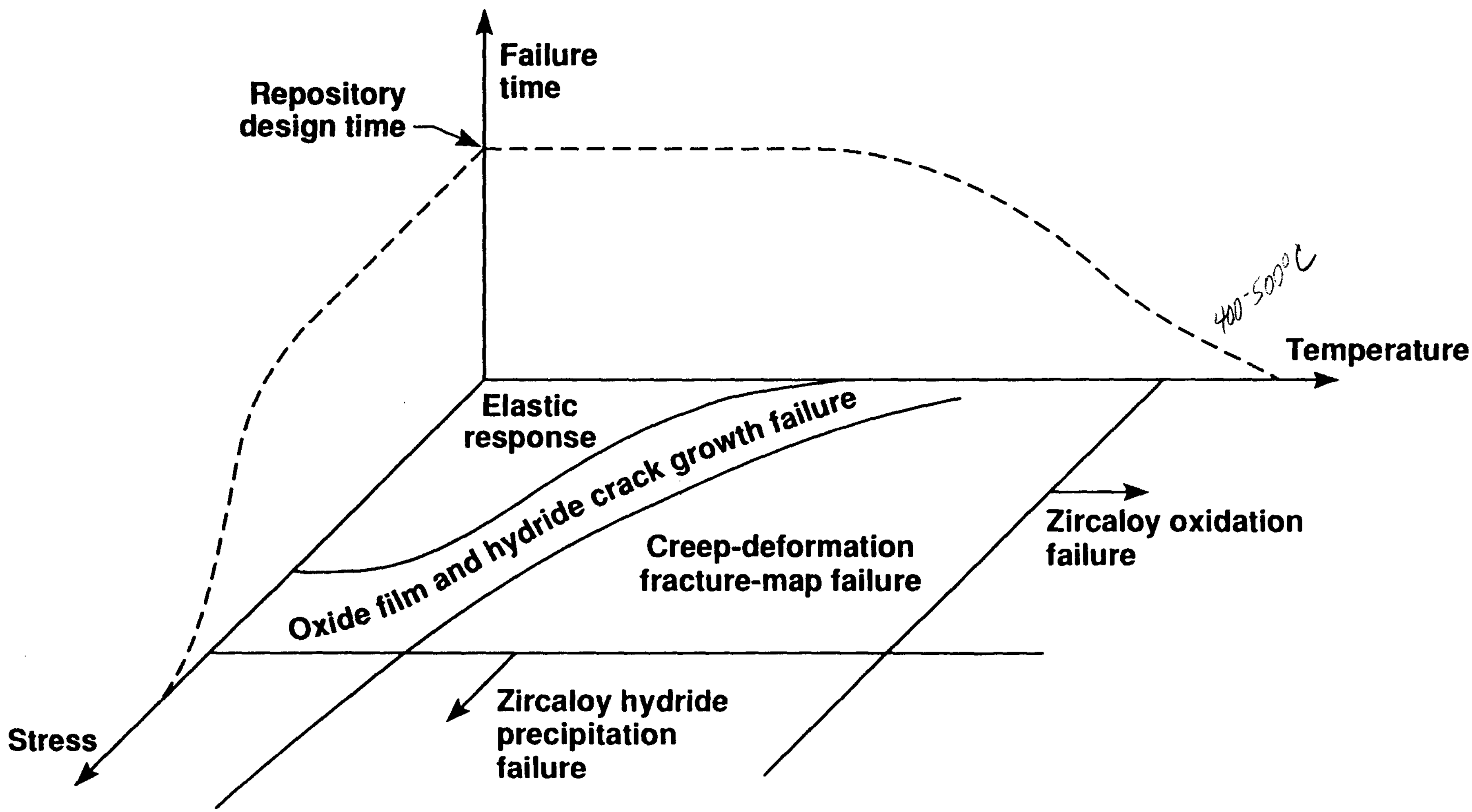


Zircaloy-fluoride corrosion response



Zircaloy Cladding Failure Modes Illustrated

Time to failure function will depend on stress-temperature-time histories and on initial hydrogen, oxide film, and stress conditions



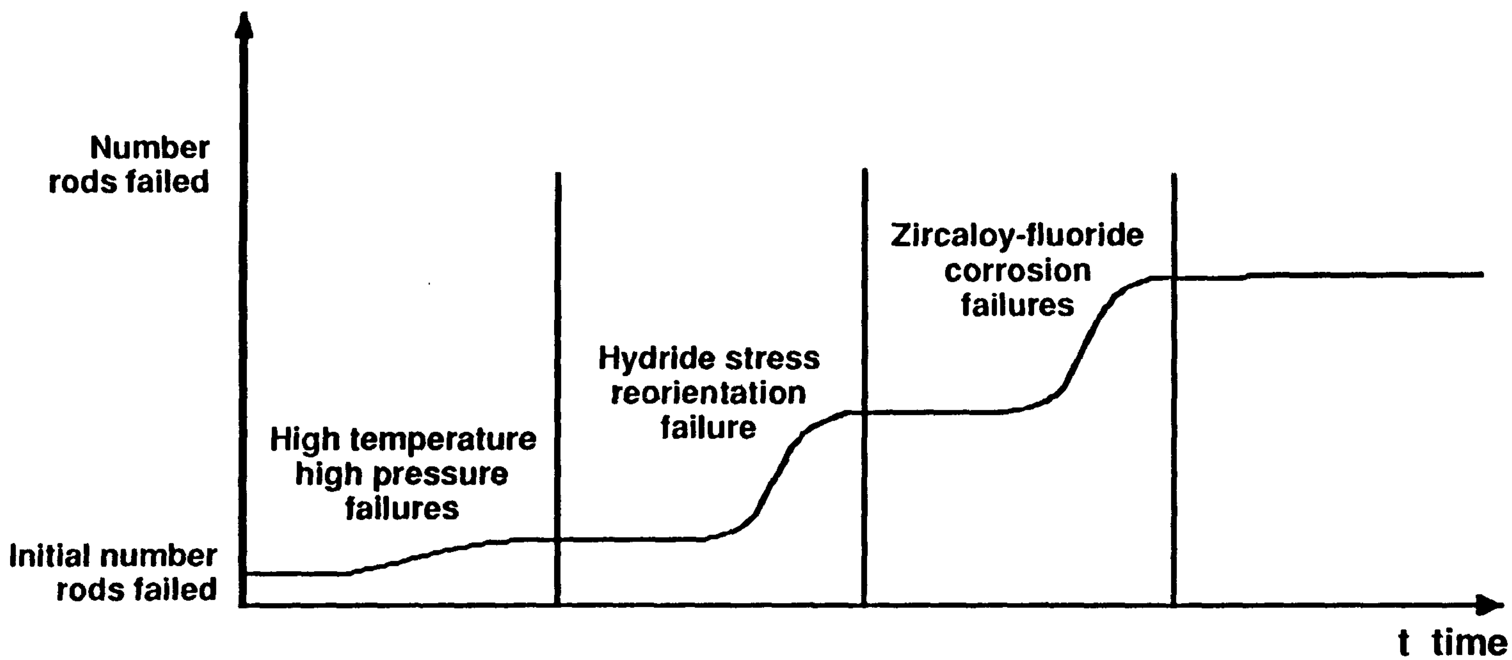
Pressurized Tube Testing System Developed

Developed apparatus for Zircaloy cladding failure testing:

1. High-temperature oxidation, time testing
2. High-temperature and high-stress deformation, time testing
3. Oxide film through-crack, strain-time testing
4. Hydride precipitation-reorientation, temperature-time testing
5. Oxide film crack propagation due to hydrides, stress-time testing

Zircaloy Cladding Degradation: Expected Total Modeling Response

Schematic of cumulative response curve for probable number of failed spent fuel rods at a time t



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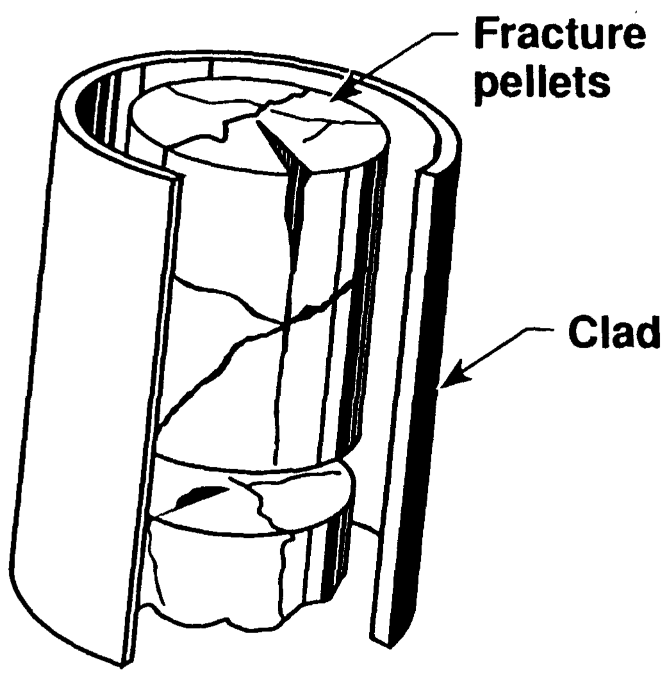
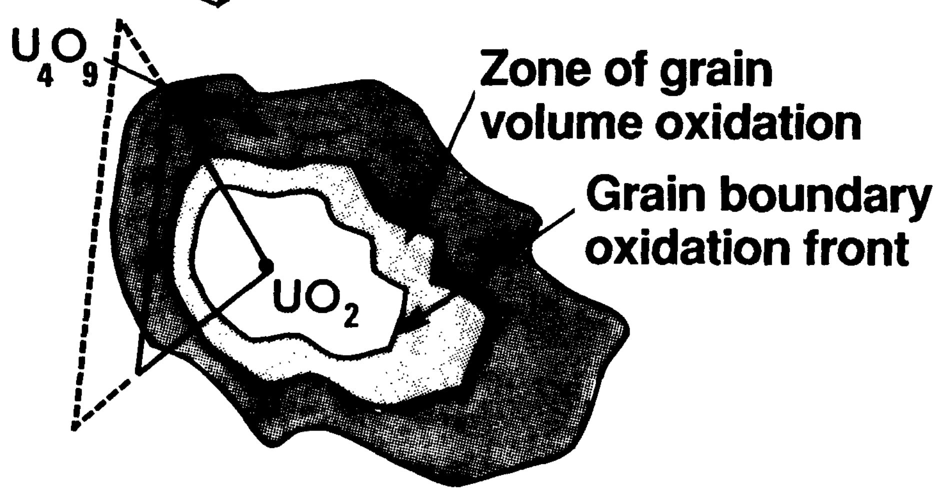
Oxidation Response





Fragment cross-section

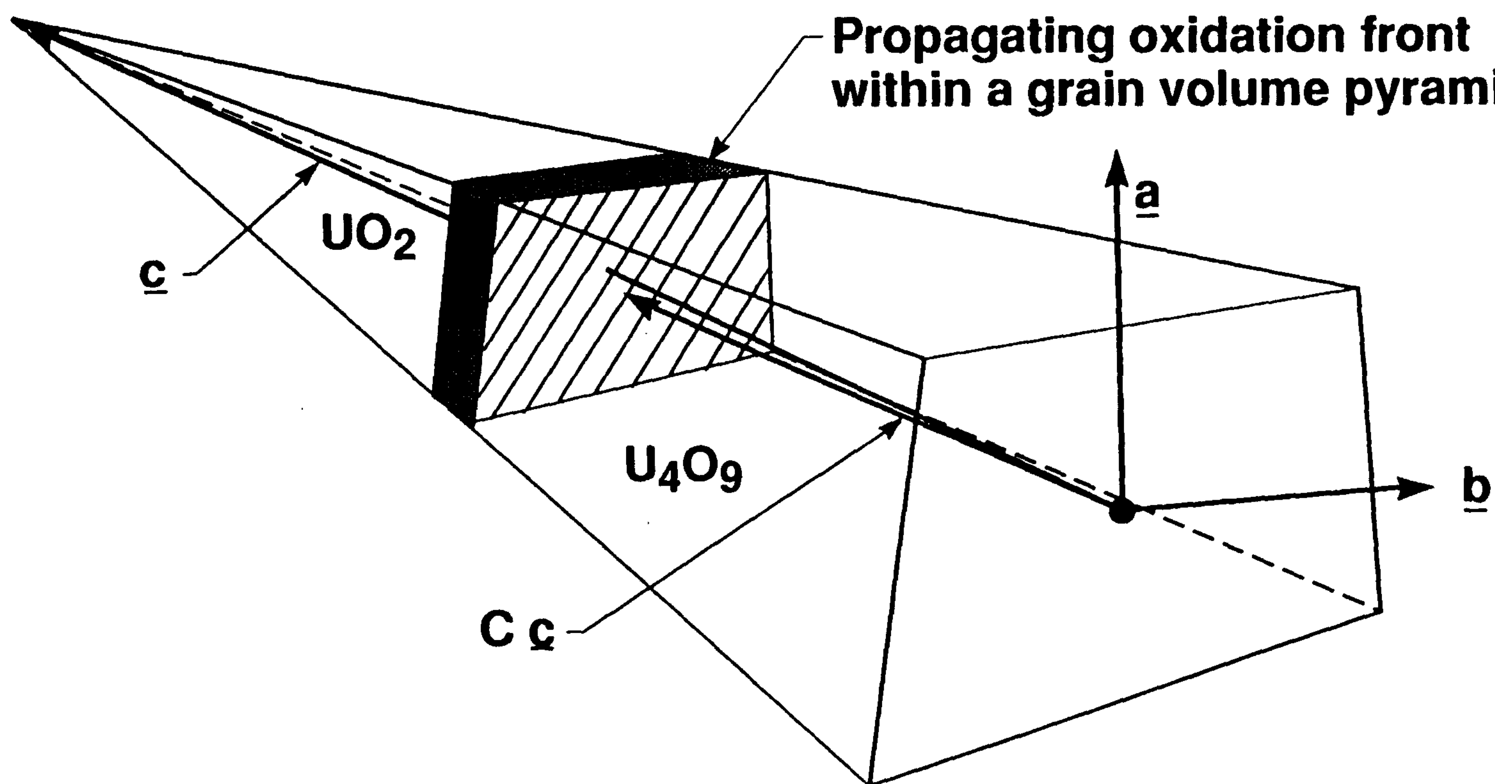
Fuel pellets, nominally 0.5cm to 0.6cm radius and ~ 2cm length, fracture into fragments due to thermal strains during first full power cycle

Tests on spent fuel fragments have shown a grain boundary oxidation front moving into fragments, followed by a spatial zone where oxidation of individual grain volumes occur

Oxidation kinetics depend statistically on fragment/grain sizes and shapes in a test sample; any fragment/grain can be subdivided into different sized pyramidal volume subsets to obtain a statistical distribution function

Grain Volume Oxidation Front

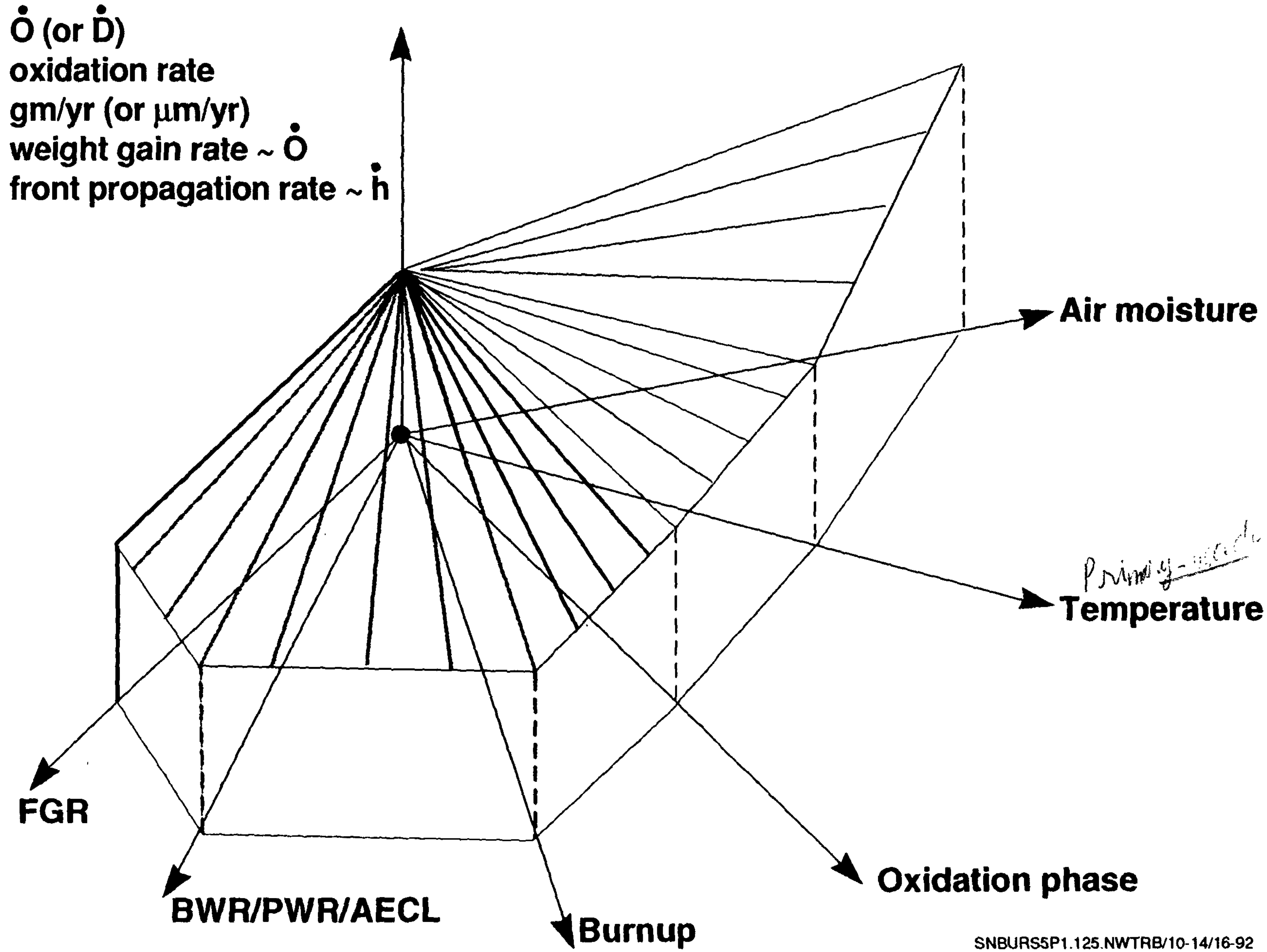
Pyramidal volume in an oxidizing grain volume and its associated physical attributes.



Oxygen weight gain rate

$$\dot{O}(t) = N e_{ijk}\, a_j\, b_k\, c_i\, (1-C)^2\, \dot{C}$$

Spent Fuel Oxidation Rate Response Surface



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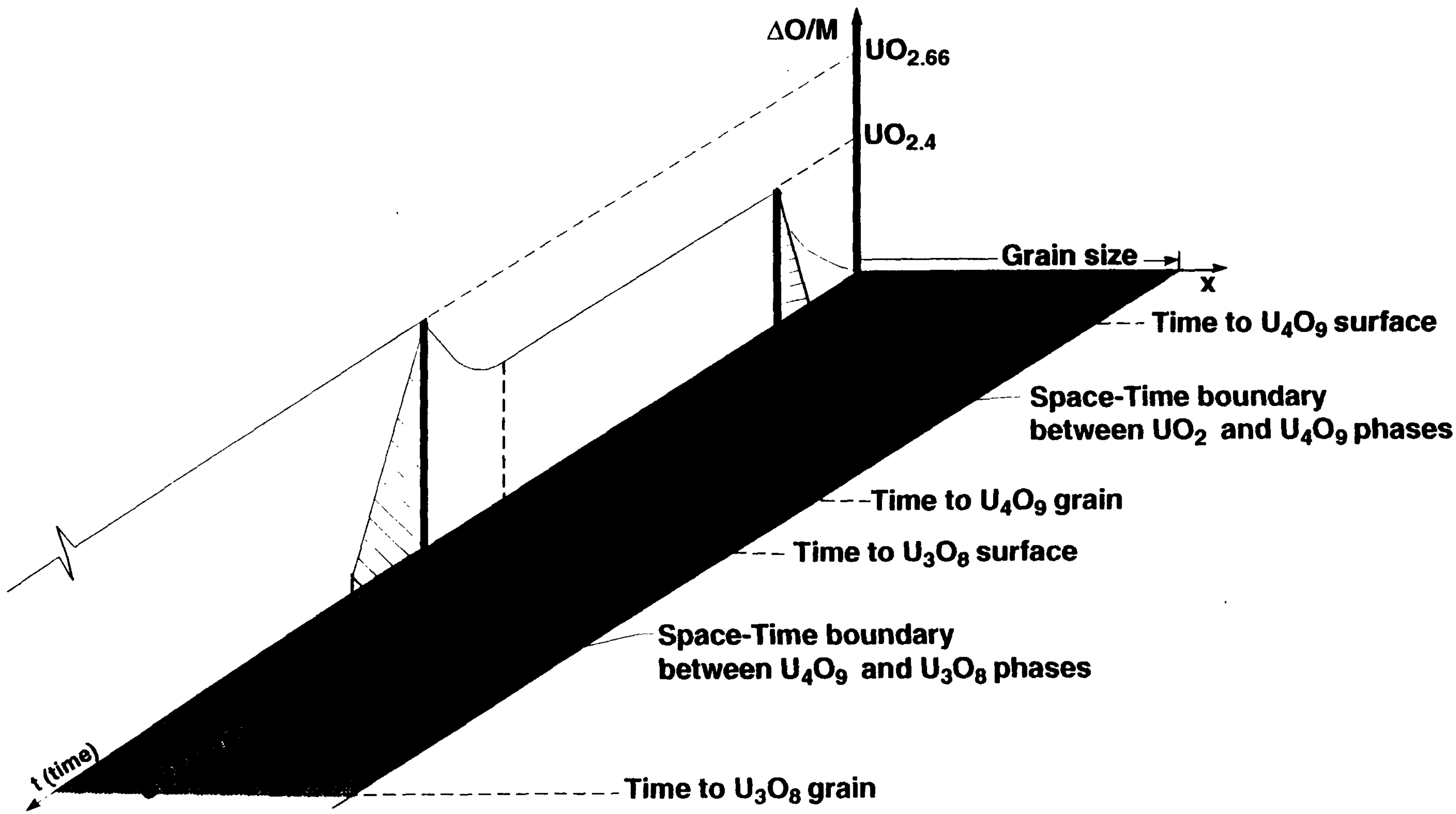
Spent Fuel Oxidation Response - Conceptualization for Model



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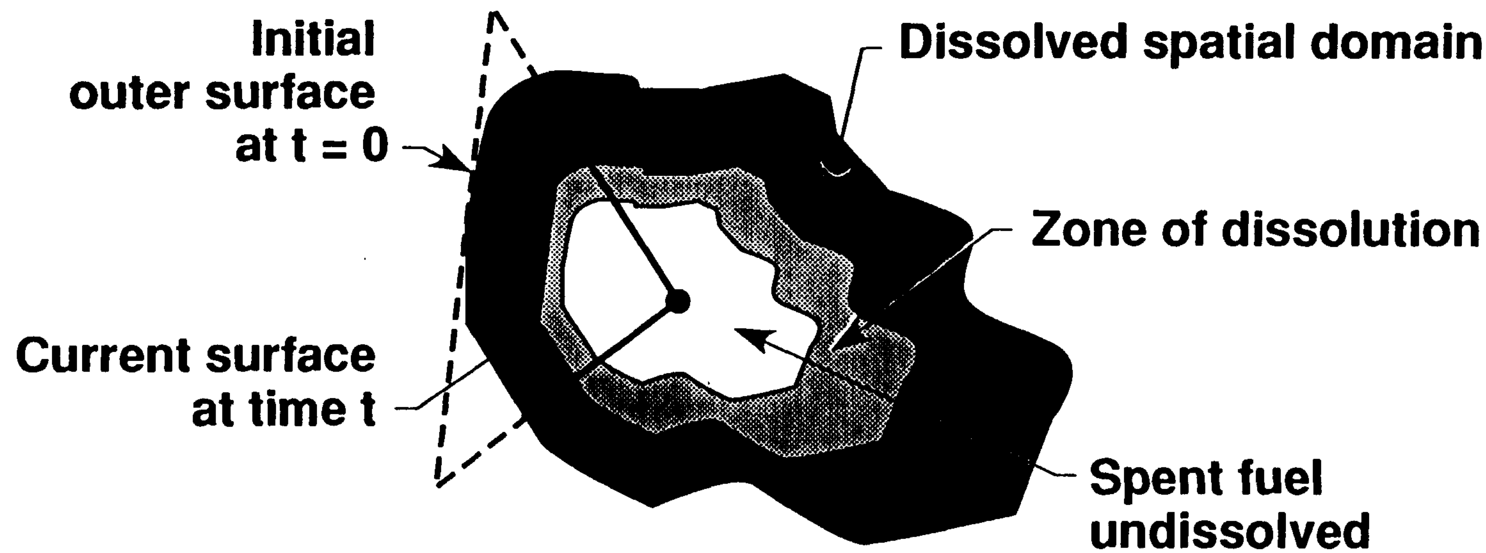
Oxidation Front and Dissolution Front Analog

Grain boundary/volume oxidation front motion through a pellet fragment has geometrical and model development similarities to an idealized dissolution surface motion progressing into a fragment.

This means that model development concepts for oxidation kinetics can be also applied to dissolution response for a distribution of fragments.



Fragment cross-section

Grain Boundary Attack on Bare Fuel

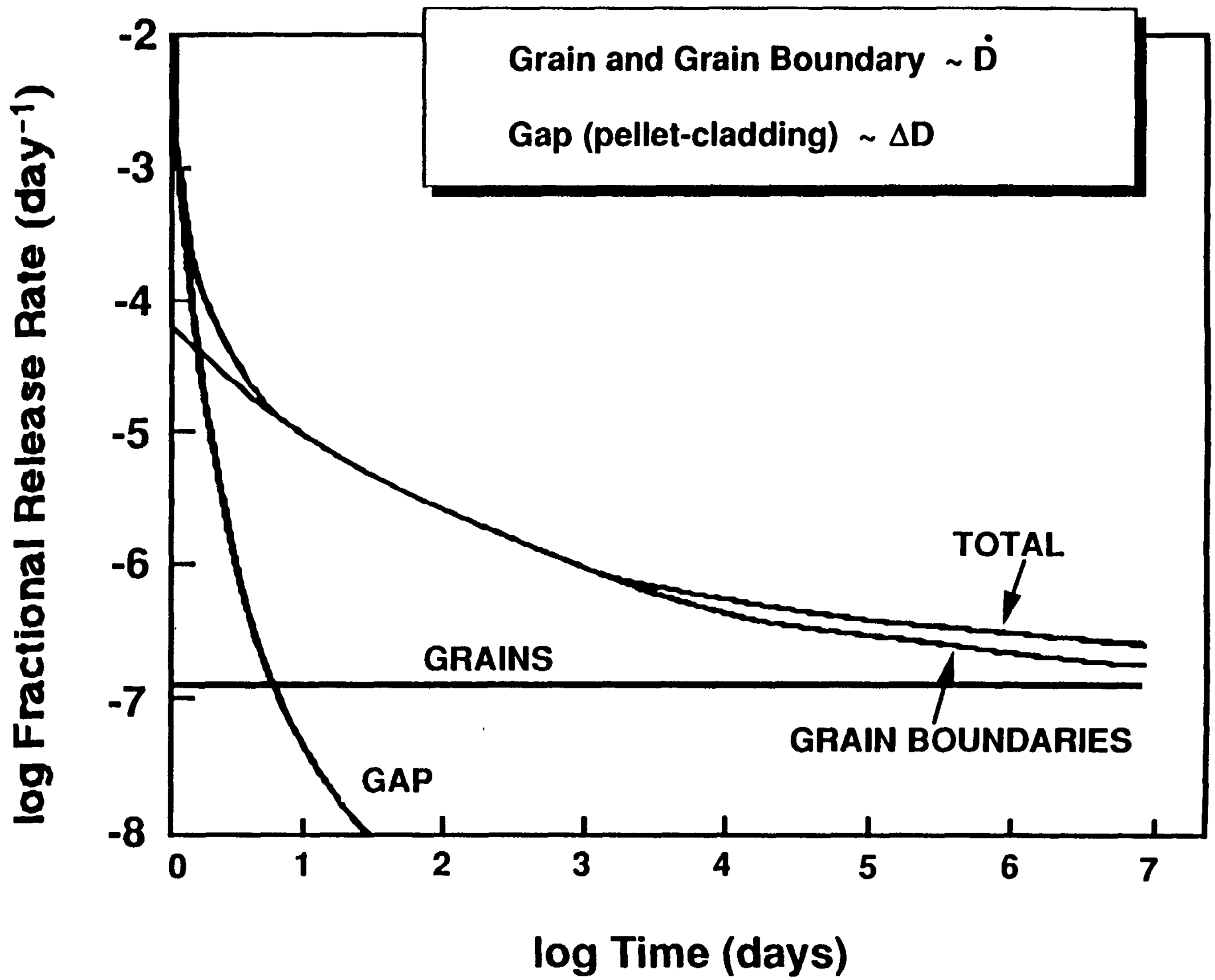
Turkey Point Fuel in Deionized Water for 1 Year

Photo

Photo

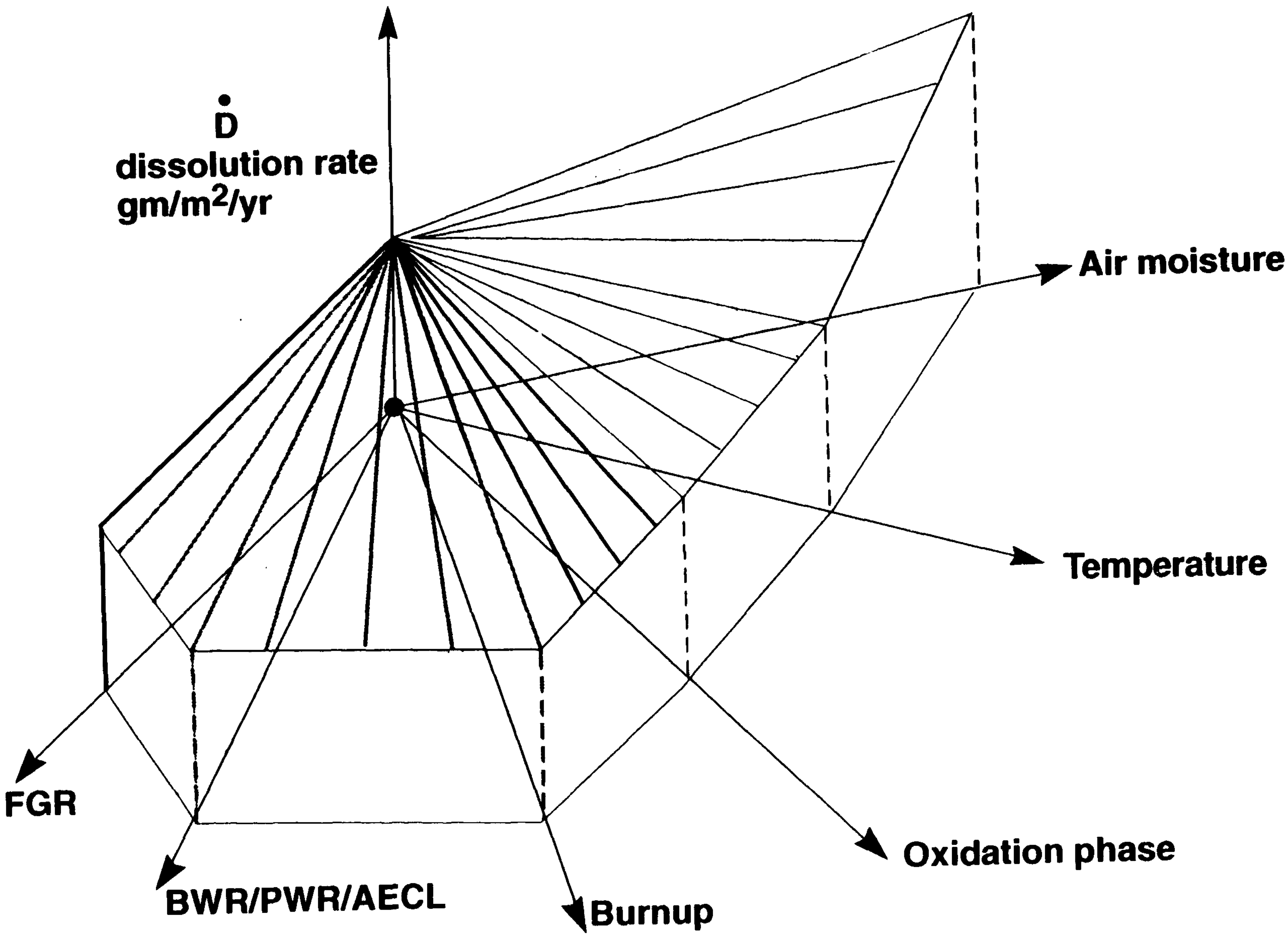
30 μm

A Schematic View of Spent Fuel Dissolution*



* L.H. Johnson and D.W. Shoesmith, "Radioactive Waste Forms for the Future," W. Lutze and R.C. Ewing, eds. Elsevier (1988) p. 686.

Spent Fuel Dissolution Rate Response Surface



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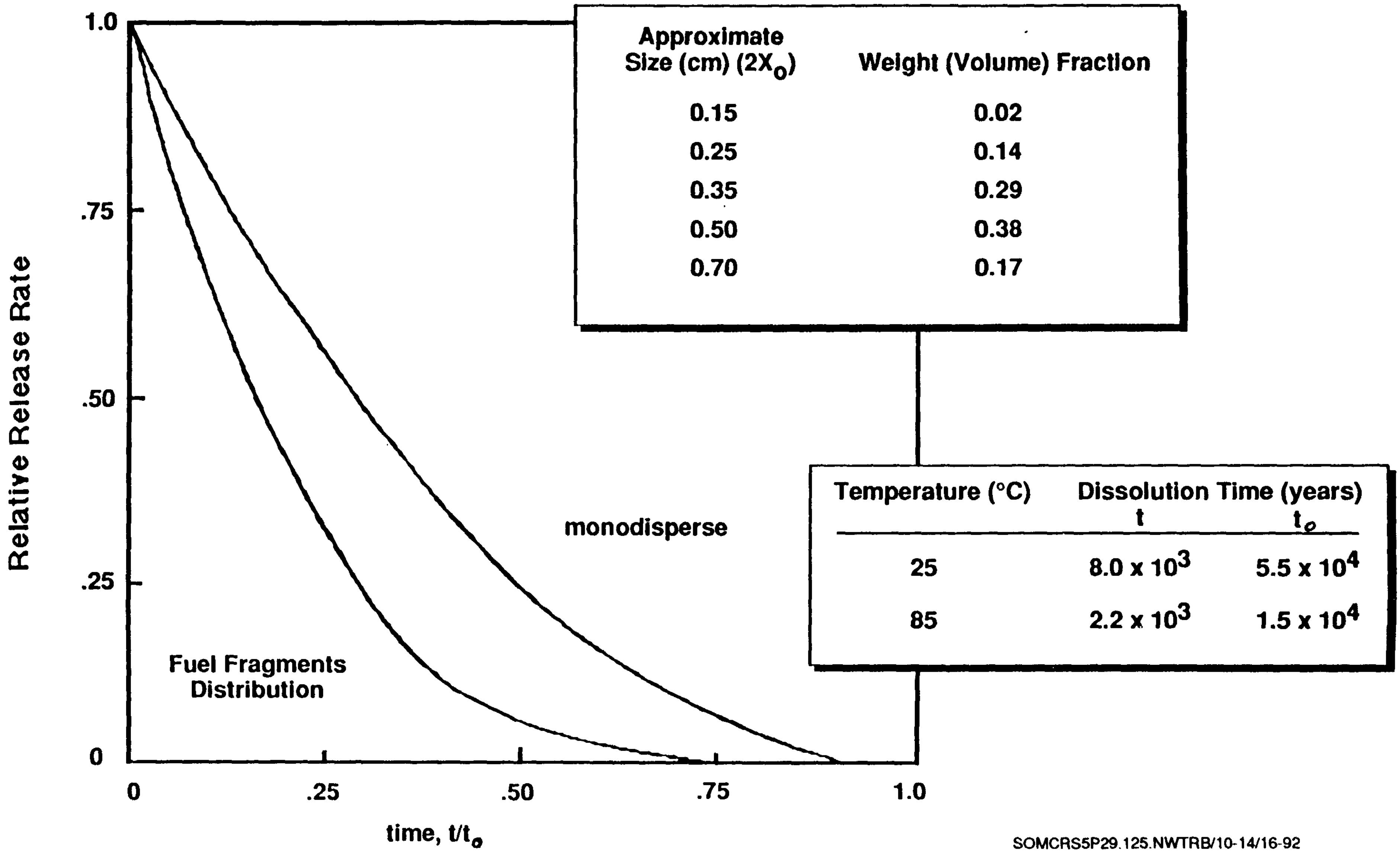
Release Rate: Volume of Fragments - Aqueous (Flow-thru Case Only)



Approximate Size (cm) ($2X_o$)	Weight (Volume) Fraction
0.15	0.02
0.25	0.14
0.35	0.29
0.50	0.38
0.70	0.17

Temperature (°C)	Dissolution Time (years) t	t_o
25	8.0×10^3	5.5×10^4
85	2.2×10^3	1.5×10^4

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Dissolution Release — Spent Fuel Pellets — Aqueous (Flow Thru)

$$\begin{aligned}\dot{R}\mathrm{elease}_{(aq)} &= C \otimes R \otimes A_e \otimes A_W \otimes V_W \otimes \dot{D} \\ &+ \dot{C} \otimes R \otimes A_e \otimes A_W \otimes V_W \otimes \Delta D \\ &+ C \otimes \dot{R} \otimes A_e \otimes A_W \otimes V_W \otimes \Delta D \\ &+ C \otimes R \otimes A_e \otimes \dot{A}_W \otimes V_W \otimes \Delta D\end{aligned}$$

C ... Container failure

R ... Rod failure

A_e ... Area exposed/rod(R)

A_W ... Area wetted/V_W/A_e

V_W ... Volume water rate

$\dot{D}$ ... gm/m^2/yr, dissolution rate

ΔD ... instantaneous dissolution

$\dot{R}\mathrm{elease}_{(aq)}$ – no precipitation (solubility limits) constraints

$\dot{R}\mathrm{elease}_{(solution)}$ = {$\dot{R}\mathrm{elease}_{(aq)}$ subject to solubility limits and colloidal response restraints}

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Release Rate Response: Hardware

Release Rate Hardware	- [Inventory] • [Dissolution Rate] • [Area]
Inventory	- Activated species - used ORIGEN2 computer code
Dissolution Rate	- Metal corrosion rate (gm / area / time)
Area	- Estimates have been made SS ~ 652,000 M^2; Inconel ~ 1,480,000 M^2; Zircaloy ~ 400,000 M^2

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Summary: Spent Fuel Response (Rate Processes)

Spent fuel attributes (SFA)

- Initial condition data have large spread (variance)
- Much remains to be acquired (significances addressed)

Repository environment (REV)

- Space-time-dependent boundary conditions that have uncertainty (Stochastic)
- Information remains to be characterized (uncertainty)

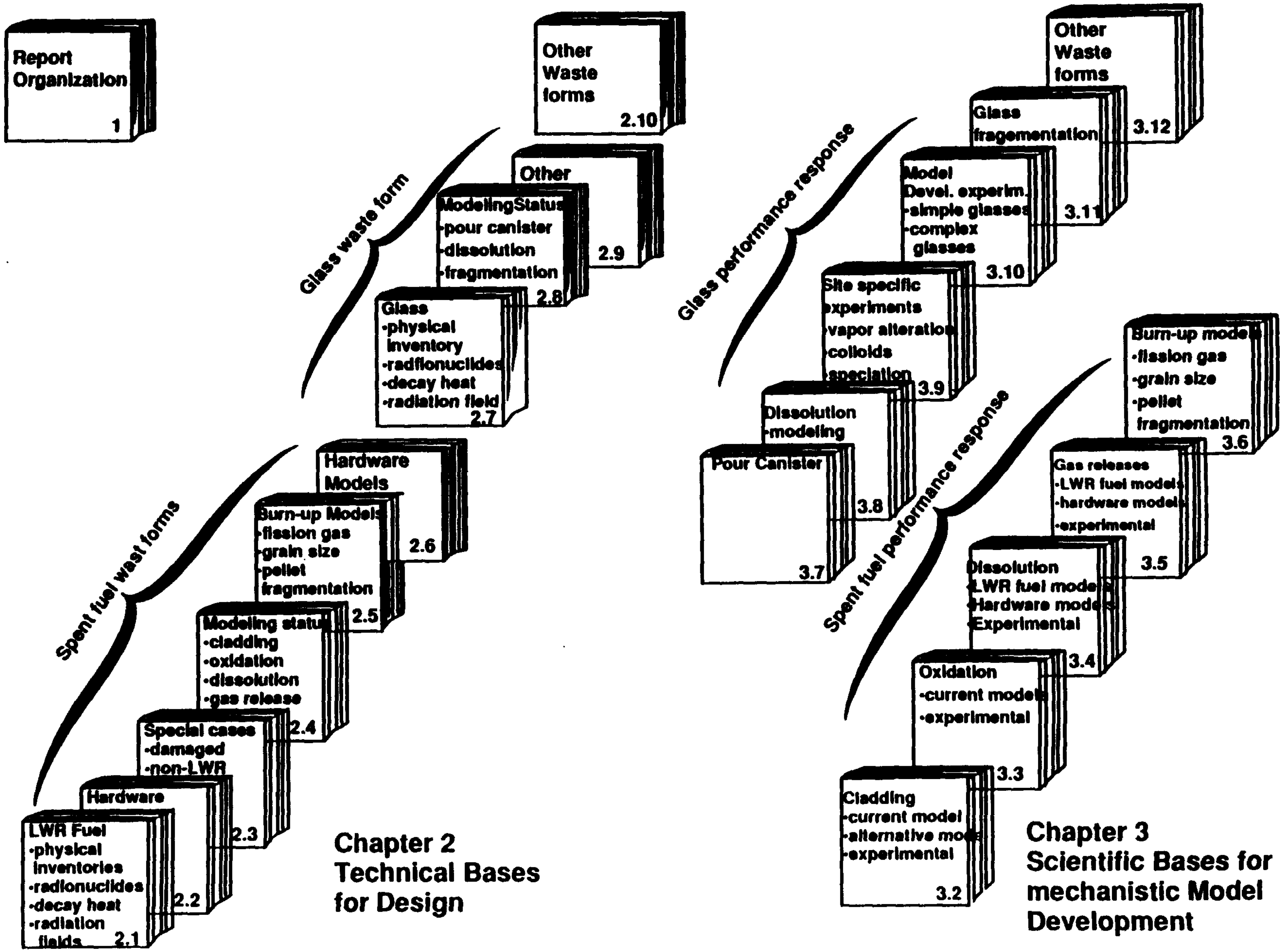
Mechanistic understanding of rate processes

- Knowledge base & models are being acquired with a multi-laboratory established testing program (concrete)
- Tests matrix for domain of SFA and REV (closure)
- Testing and modeling viewed as an iterative interface (checks)

Preliminary knowledge base and models

- Incomplete, but are being integrated in a "Preliminary Waste Form Characteristic Report" (document)

Organization of the Waste Form Characterization Report



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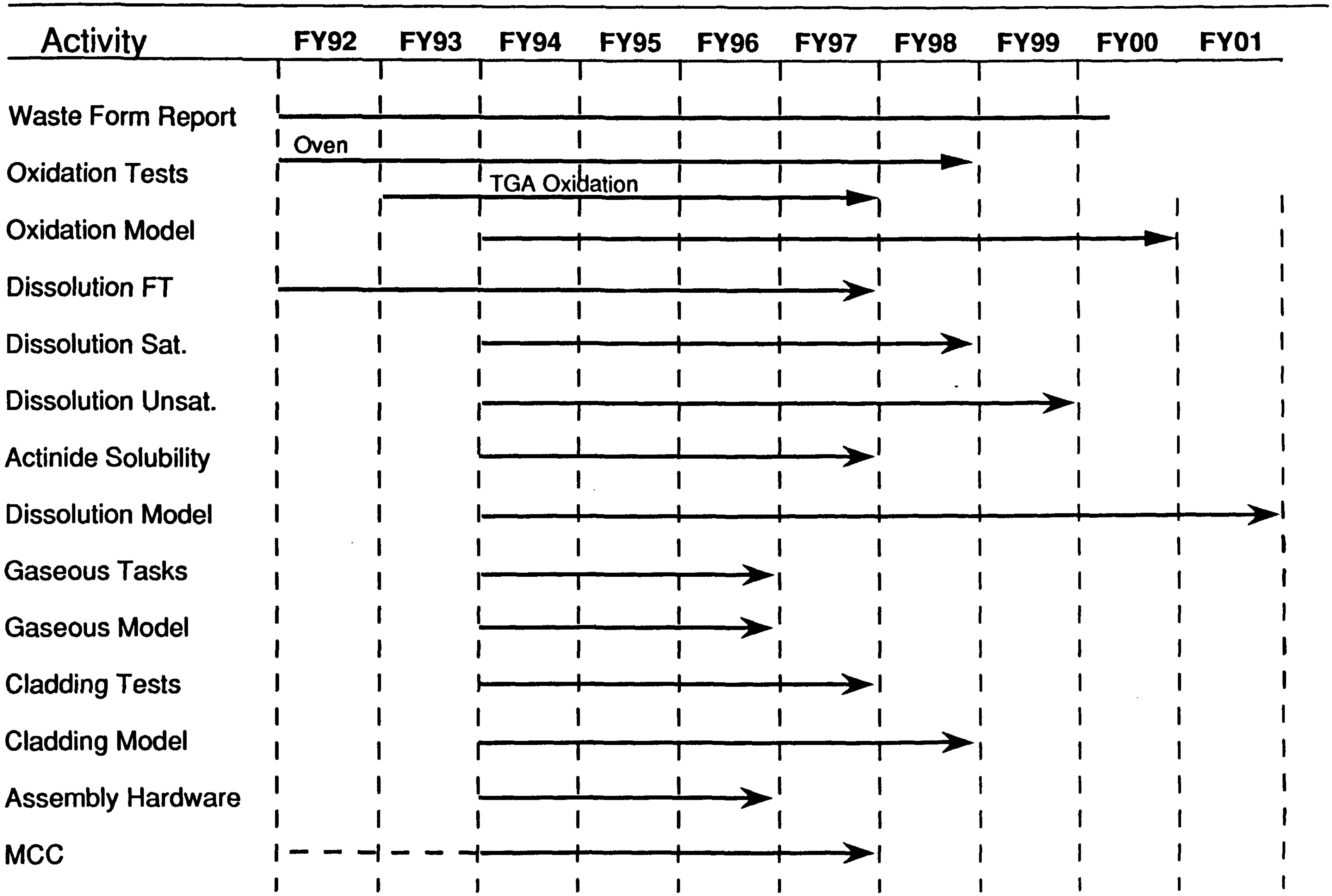
Activity Plans - Mission 2001



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