Source Term Model for USNRC Iterative Performance Assessment, Phase 2

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Introduction

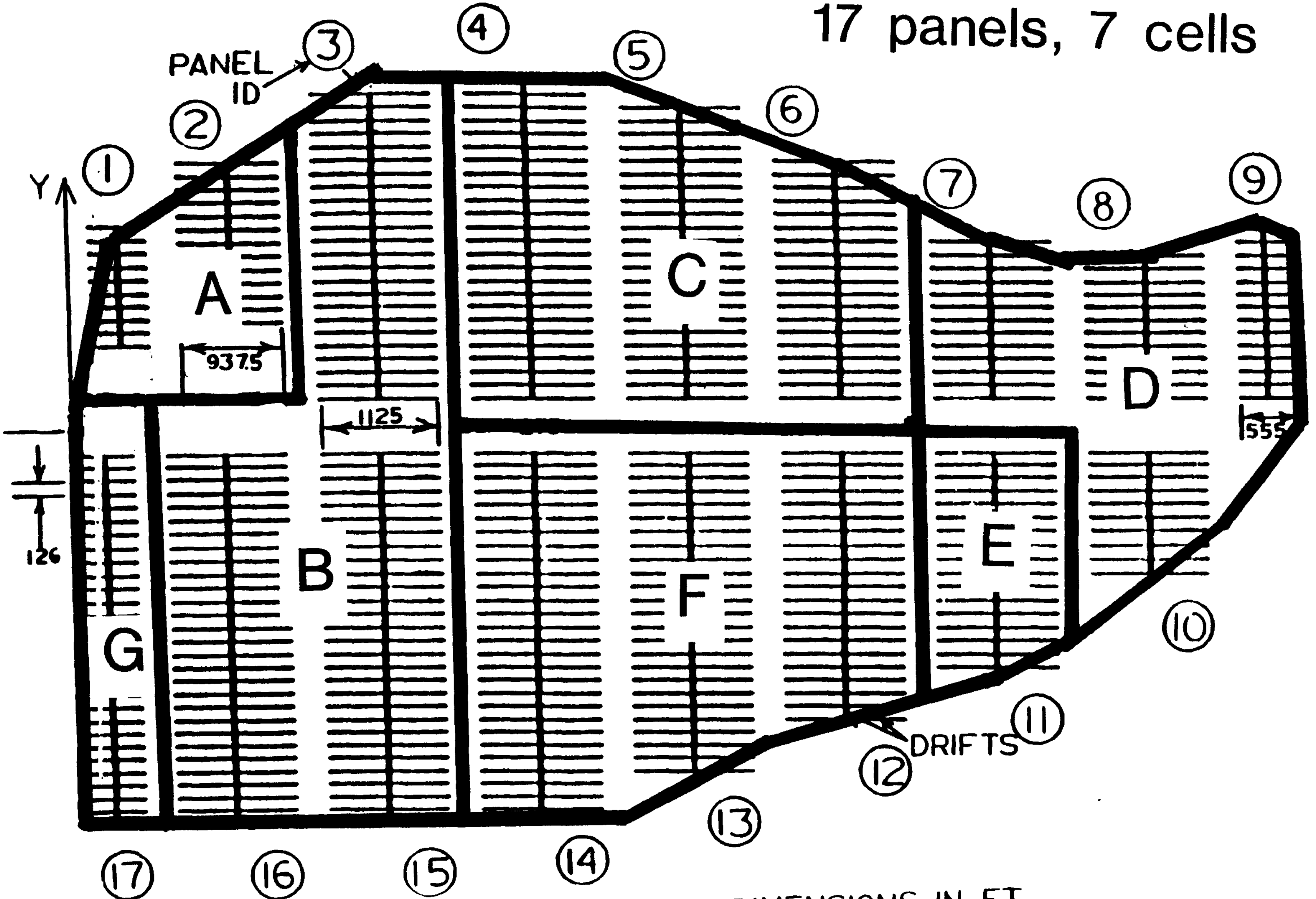
- Temperature Model
- Waste Package Failure Model
- Liquid Radionuclide Release
- C-14 Gaseous Release
- Kinetic Effects
- Disruptive Releases
- Support for Source Term Modeling

Canister Temperature Model

- Semi-analytical, conduction only
- Uniform heat transfer properties
- Heat load can vary in time and space
- Mainly used to determine time to canister failure

Zonation of Waste

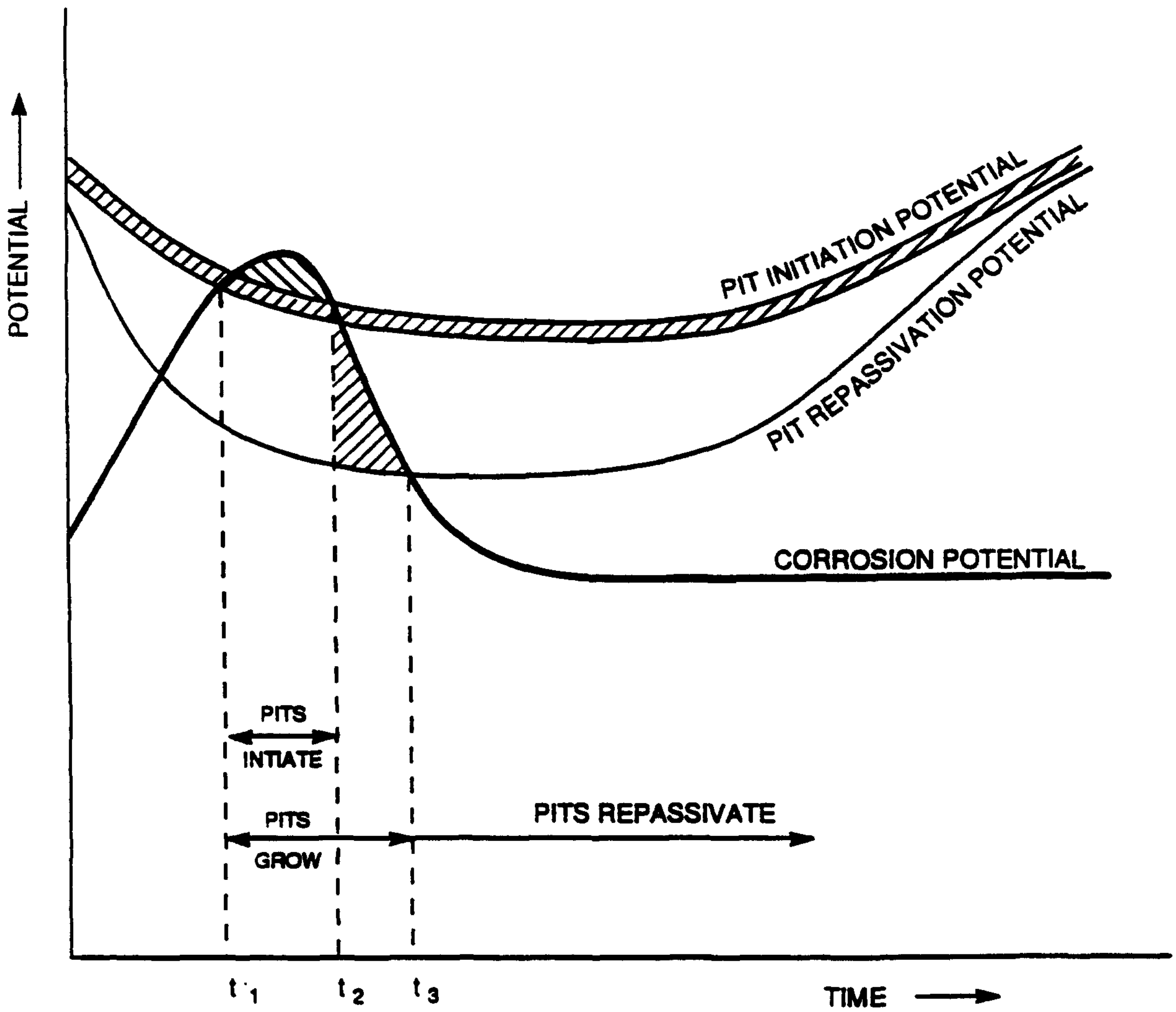
17 panels, 7 cells



DIMENSIONS IN FT

Canister Corrosion Model

- General corrosion
- Crevice corrosion
- Pitting



Schematic illustration of the use of corrosion, pitting, and repassivation potential to predict the performance of a container

Other Canister Failure Modes

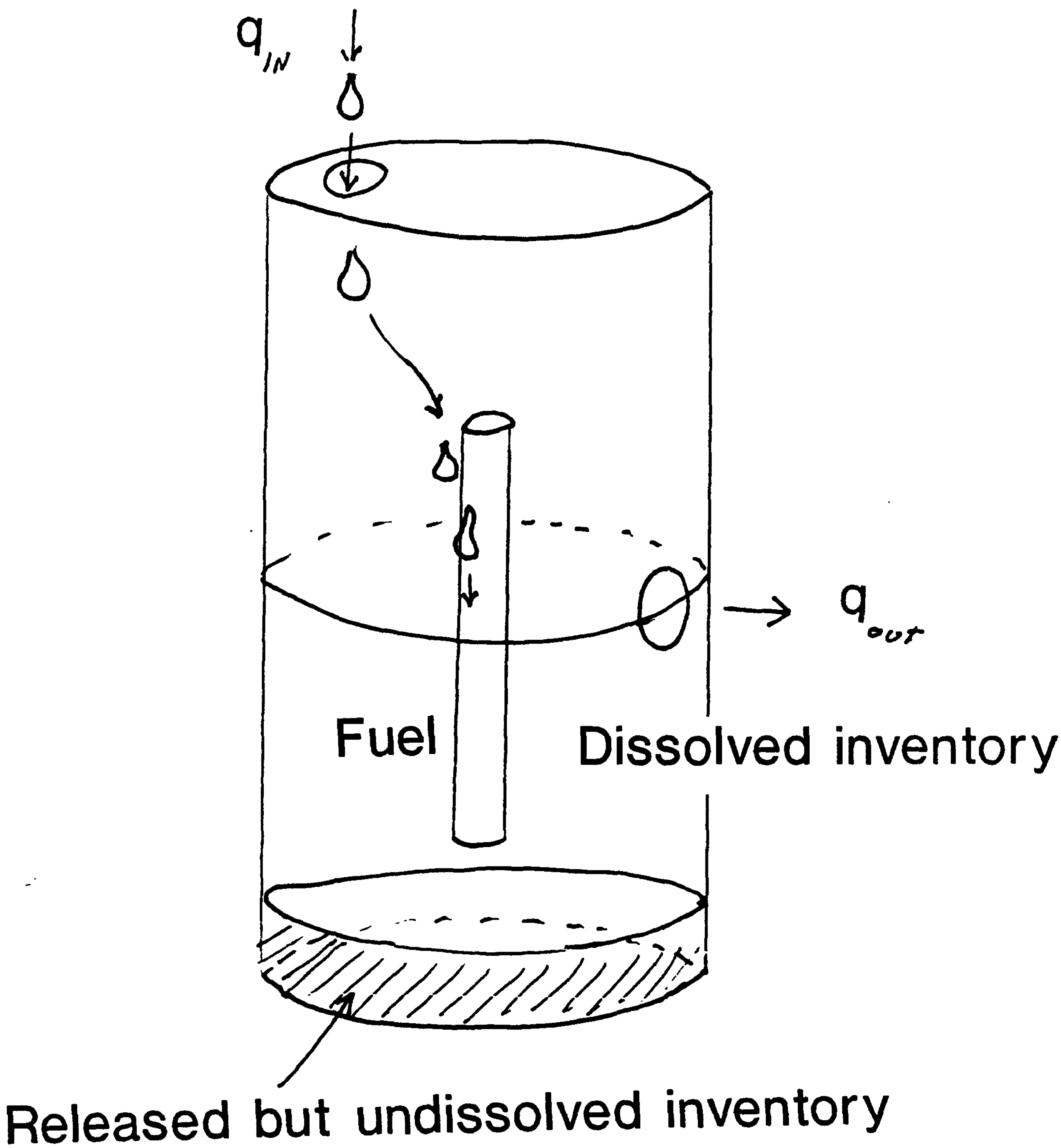
- Buckling
  - SCP design, 304L Stainless
  - Long cylinder
  - Thickness decreases by corrosion
  - No air gap
  - No stiffening
- Initial defectives
- Disruptive scenarios
  - Seismic failure
  - Volcanism
  - Drilling

Release Rate Models in SOTEC

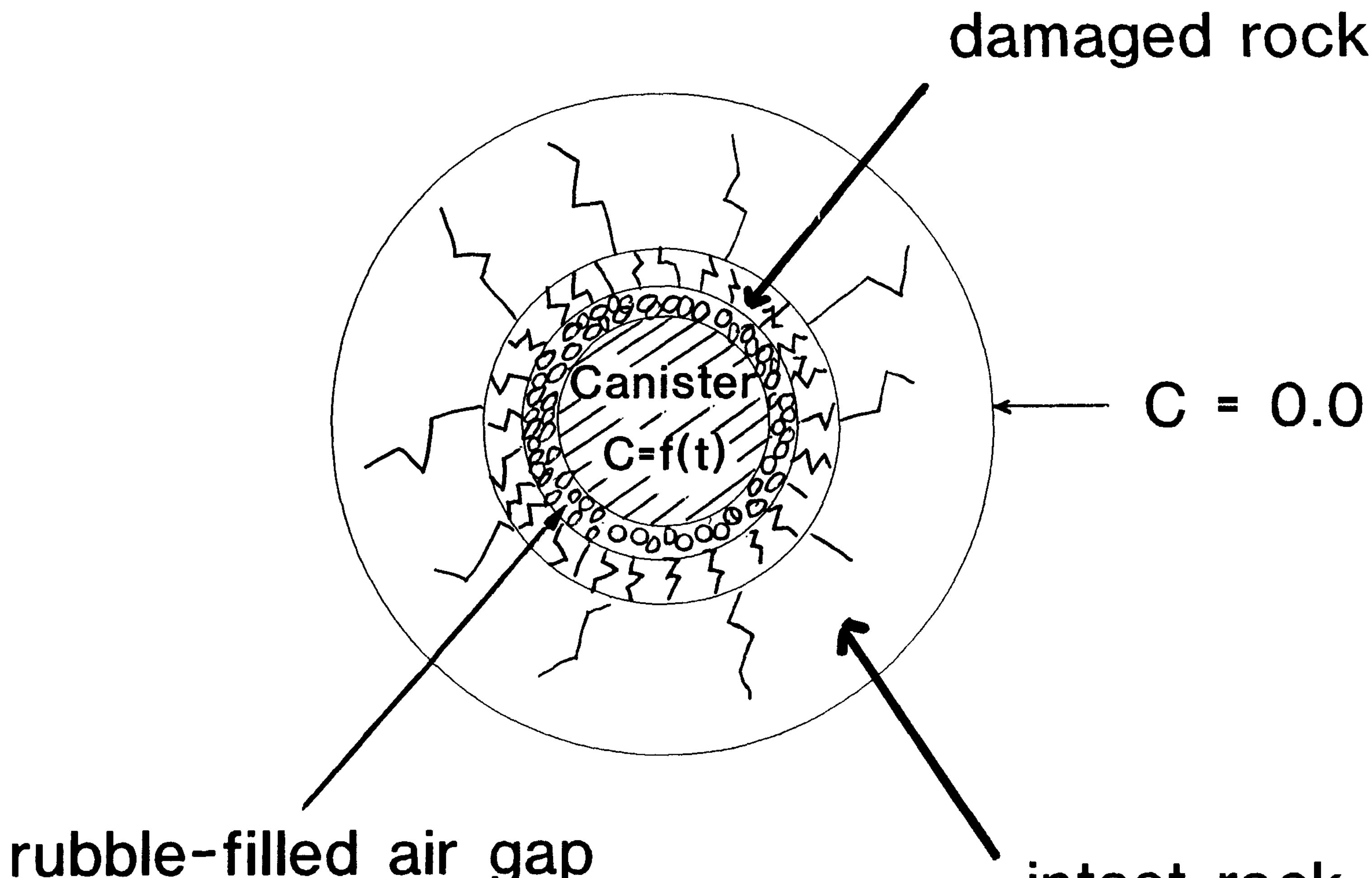
1. Dissolved and Colloidal
   a. Advective release
   b. Diffusive release
   c. Kinetic effects and colloids

2. C-14 Gaseous Release
   a. Metal oxide
   b. Cladding oxidation
   c. Grain and gap release
   d. Fuel oxidation

Dissolved Radionuclide Release Model



Liquid Diffusion Model



EVIDENCES FOR KINETIC EFFECTS

- MULTI-PHASE FORMATION
- PARAGENESIS OF SECONDARY PHASES
- UNSTABLE SECONDARY PHASES
- NONPROTECTIVE SECONDARY PHASES
- ENVIRONMENTAL CHANGES

Releases of C-14 from Spent Fuel

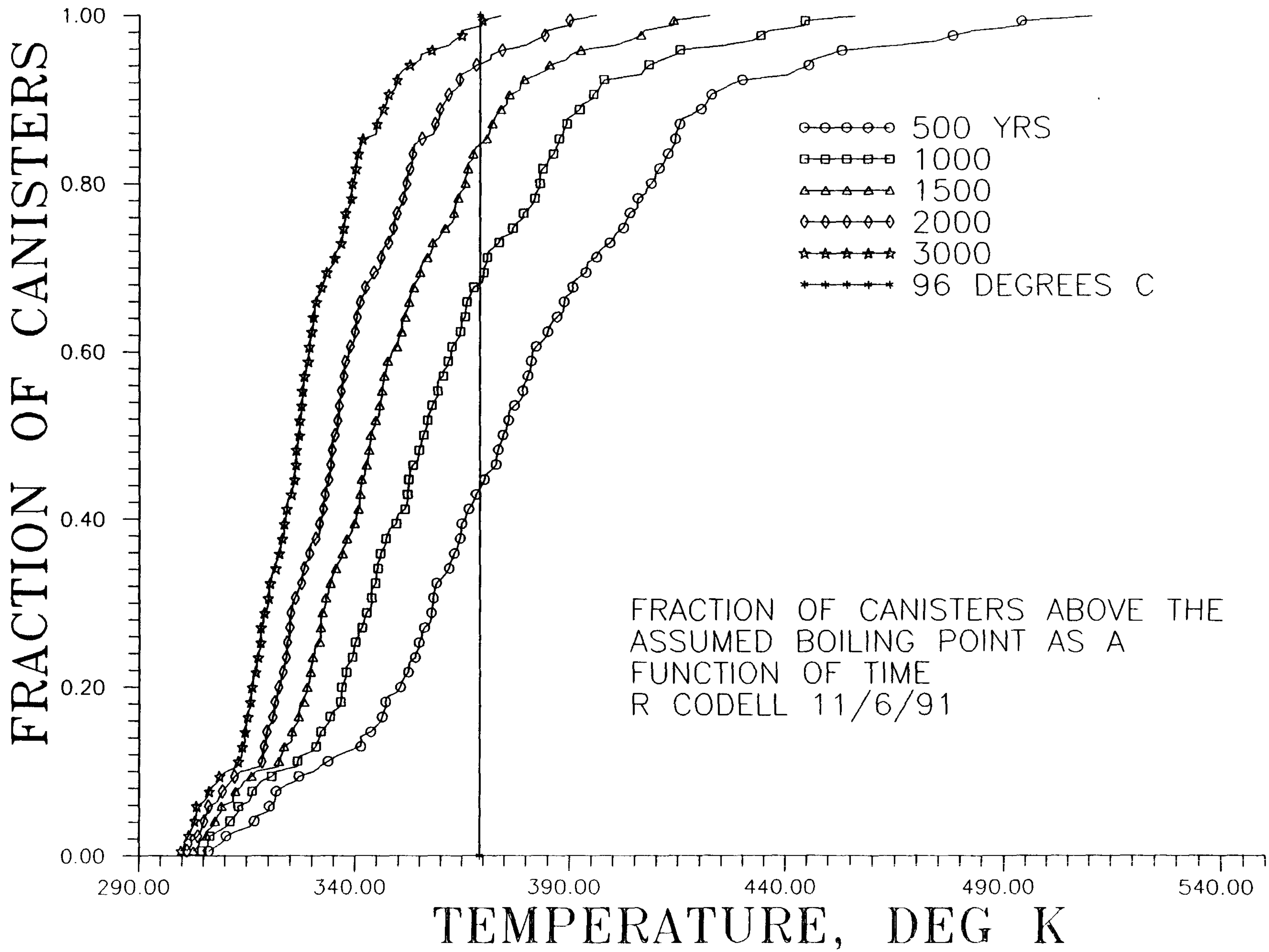
- Initial cladding oxide and crud
- Grain boundary and cladding gap
- Zircaloy oxidation
- Oxidation of fuel

Plutonium Releases

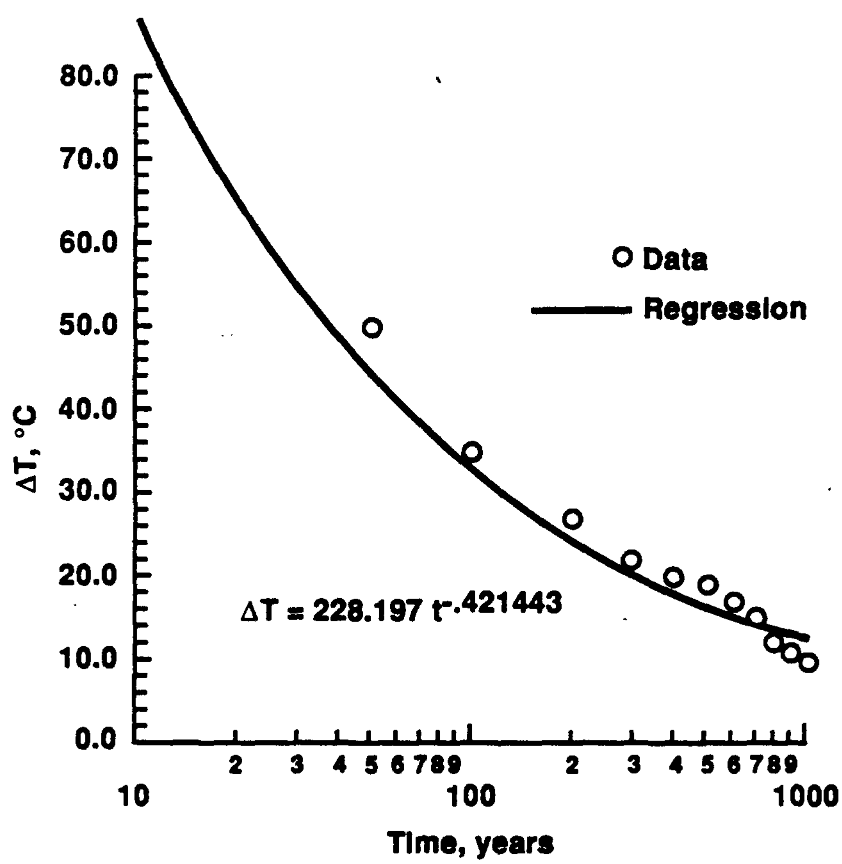
- Pu and Am dominate dose potential, but
- Very insoluble - Largely ignored in other performance assessments
- Kinetic effects may play an important part in releases of Pu and other actinides
- Potential concentrations of Pu taken from data by Nitsche et al, and Wilson et al, E-5 to E-9 M
- Speciation calculations show range reasonable at 25°C (not at 85°C)

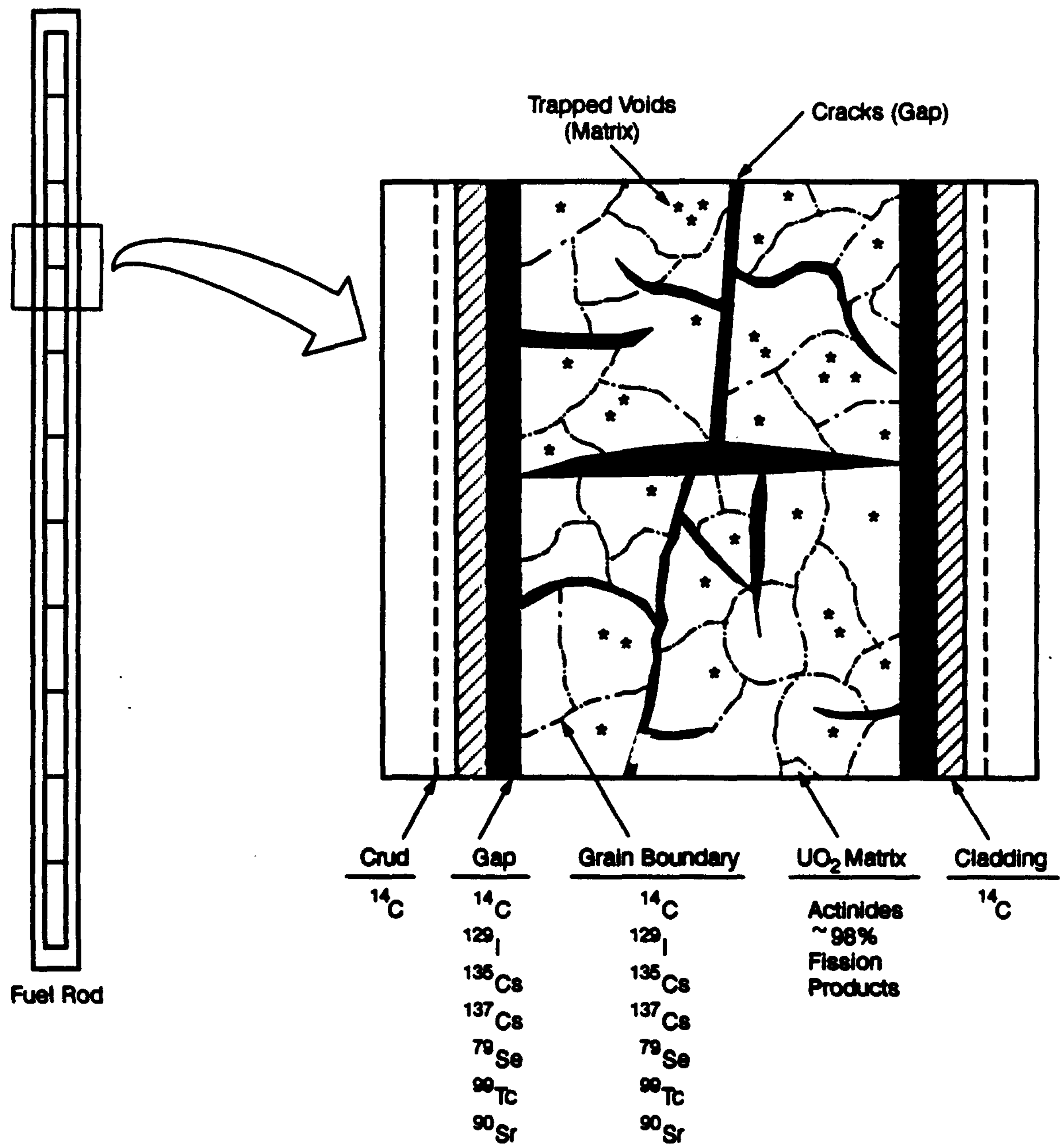
FAST RELEASE OF RADIONUCLIDES FROM HLW FORMS

- Colloid
  - Real Colloid
  - Pseudo-Colloid
- Supersaturation
- Species
- Effects of Surface Area
- Effects of Radiation, Stress, Preferential Dissolution and Microbe


FRACTION OF CANISTERS
1.00
0.80
0.60
0.40
0.20
0.00
290.00
340.00
390.00
440.00
490.00
540.00
TEMPERATURE, DEG K
500 YRS
1000
1500
2000
3000
96 DEGREES C
FRACTION OF CANISTERS ABOVE THE
ASSUMED BOILING POINT AS A
FUNCTION OF TIME
R CODELL 11/6/91

Fuel Temperature vs. Canister Skin Temperature





Location of Radionuclides in Spent Fuel and Potential Releases of C-14
(Apted, el. al., 1989)

Adjusted ^{14}C Content in Spent Fuel (Ci/MTHM)
(After Park, 1992)

Type	Burnup Mwd/MTHM	UO_2	Zirc.	Hard-ware	Total
BWR	35,000	0.69	0.48	0.13	1.3
PWR	40,000	0.73	0.22	0.26	1.21
Average		0.72	0.31	0.21	1.24

Model for Release of C-14

- C-14 is in a reduced state initially in fuel
- C-14 oxidizes to carbon dioxide as fuel oxidation front passes
- C-14 dioxide diffuses out through same two layers as oxygen diffuses in
- C-14 released quickly from grain boundaries, cladding/fuel gap, and initial zirc. oxide
- Minor releases from oxidation of cladding and other metals

Time to Diffuse Most C-14 From Outer Cladding Oxide

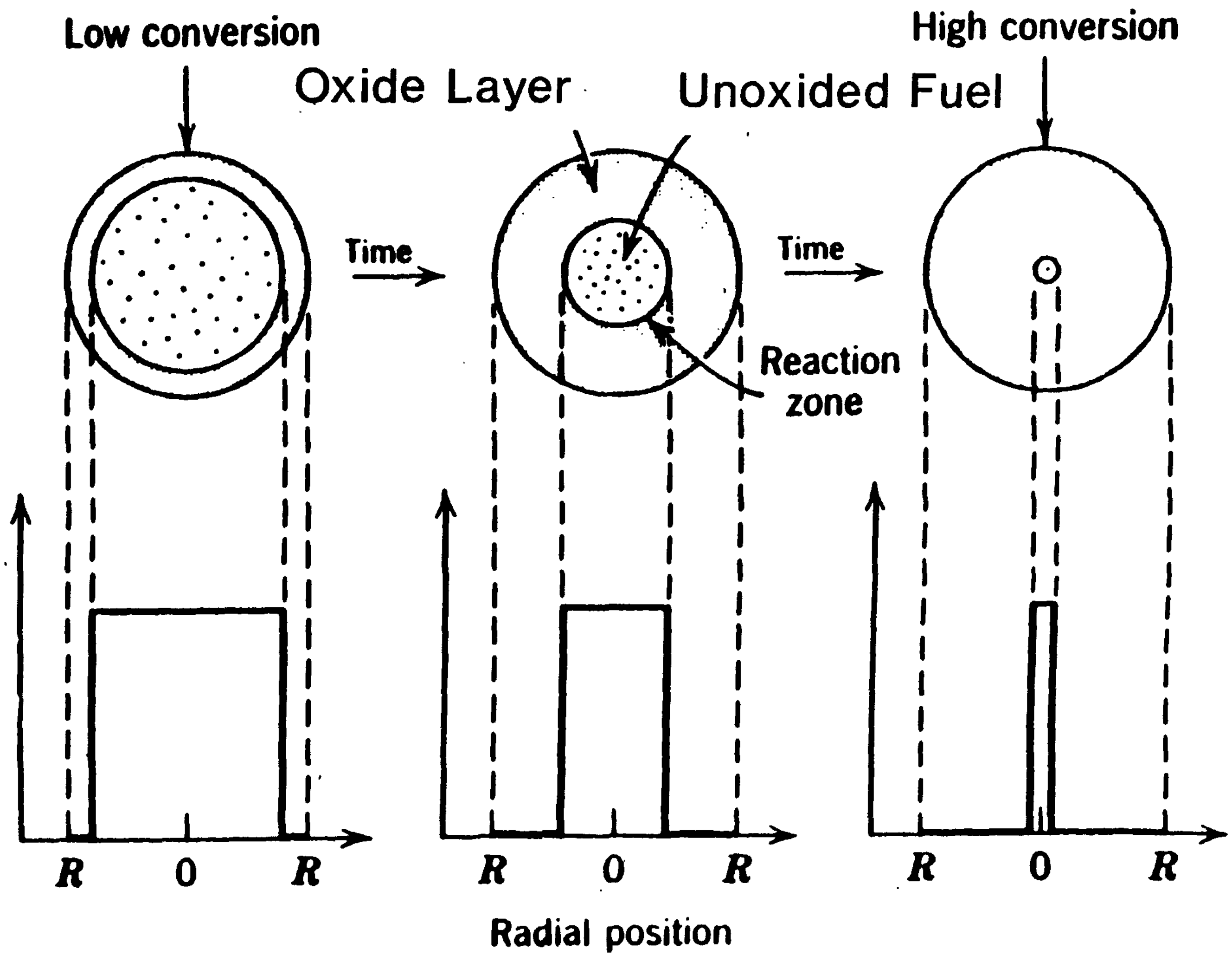
T°C	t_2, yrs E=19 Kcal/mol	t_2, yrs E=25 Kcal/mol
350	0.00091	0.00091
300	0.0035	0.0053
250	0.017	0.043
200	0.12	0.55
150	1.3	13
100	27	690
75	169	7800

Fuel Oxidation Model

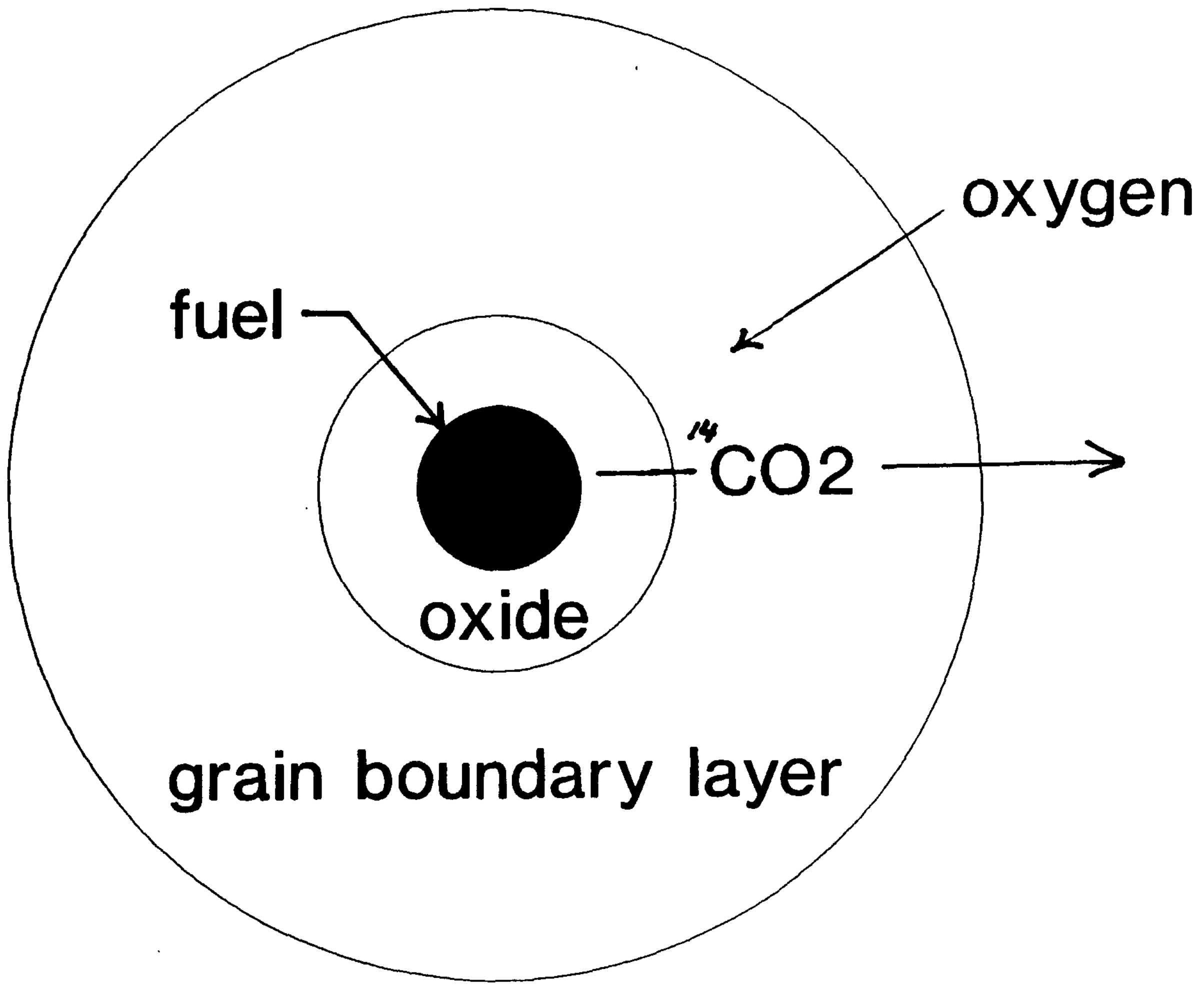
Assumptions:

- No oxidation until canister fails
- No protection of fuel by cladding
- Oxygen diffuses through two layers:
  - outer layer representing grain boundaries
  - inner layer representing oxidized fuel
- Oxide is U3O7 stochiometrically
- Oxygen concentration zero at inner boundary
- Oxygen profiles in layers are at steady state

Shrinking Core Model for Fuel Oxidation

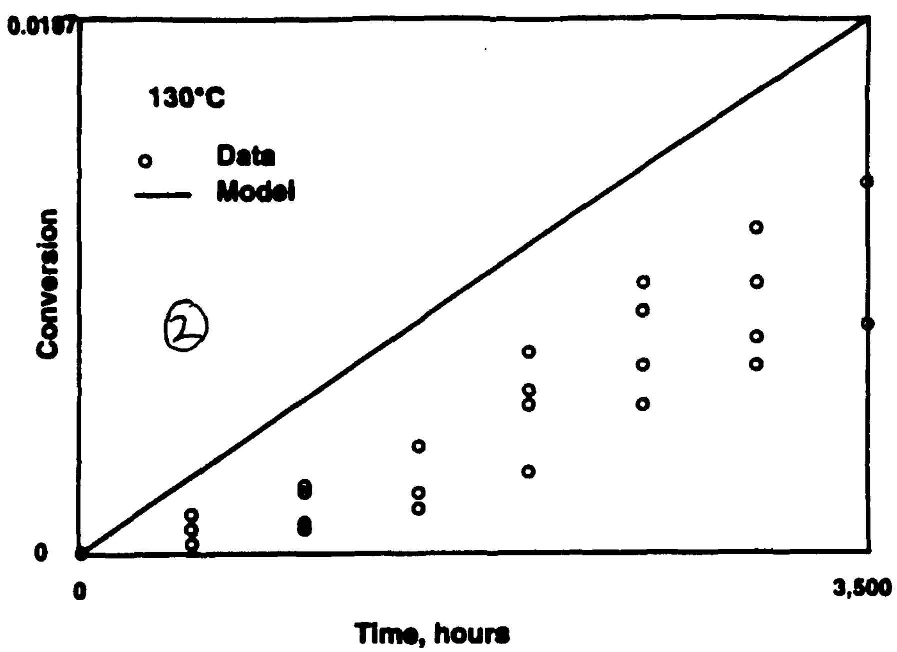


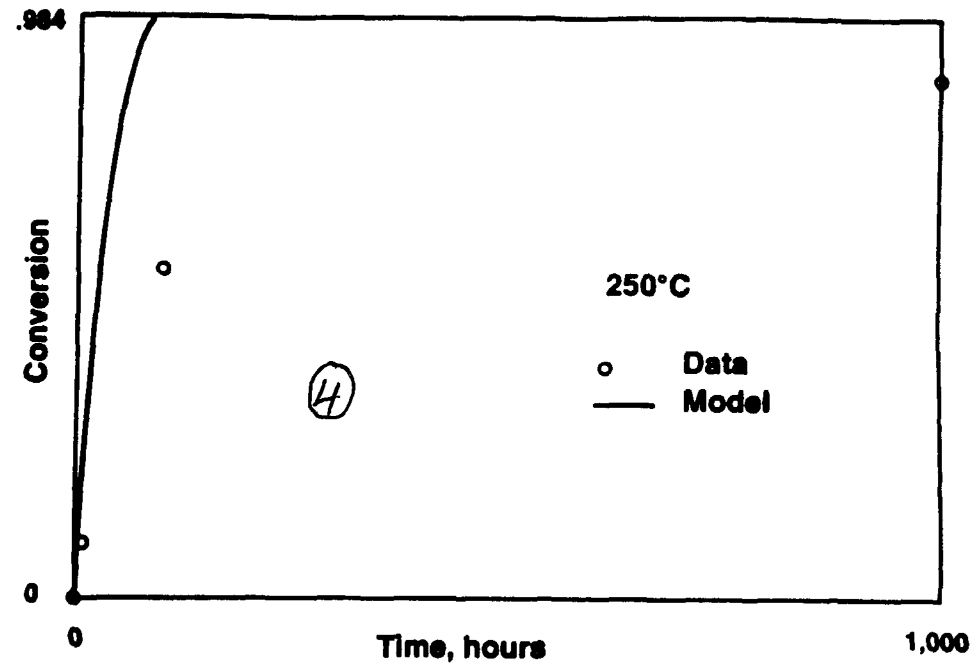
C-14 Gaseous Release Model

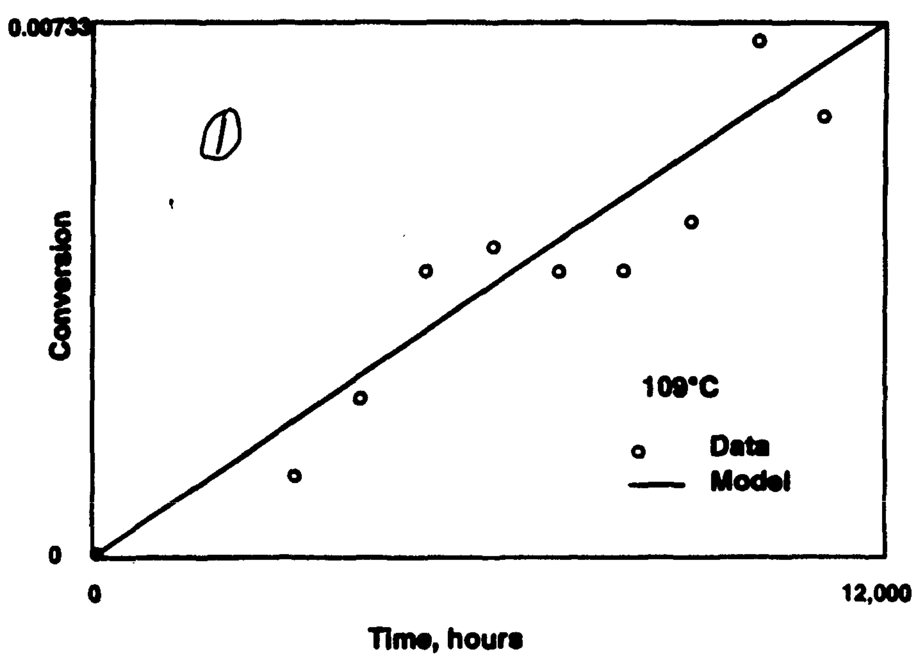


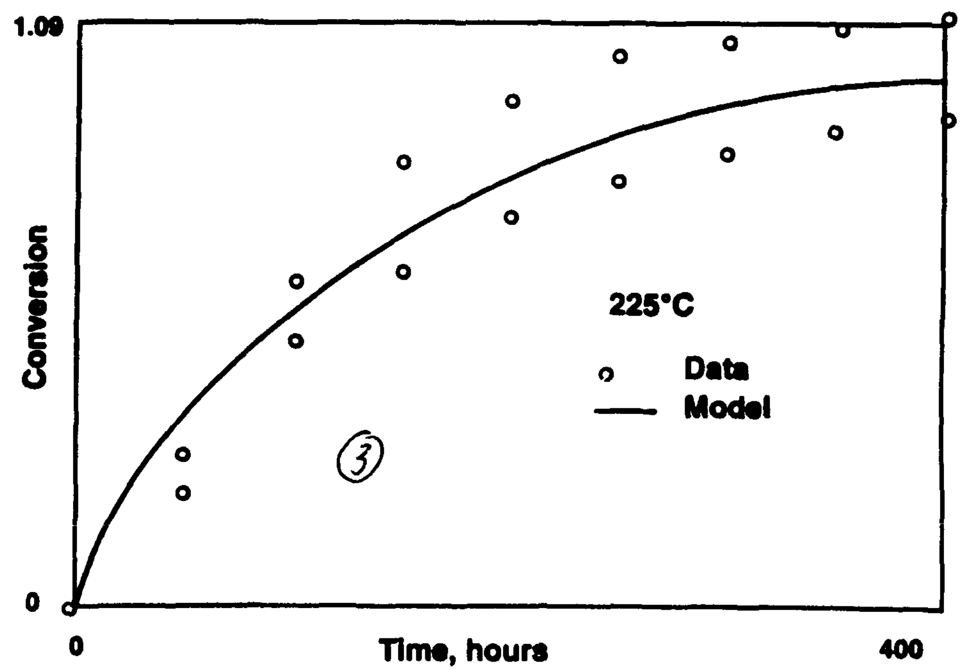
Parameter Identification and Model Verification

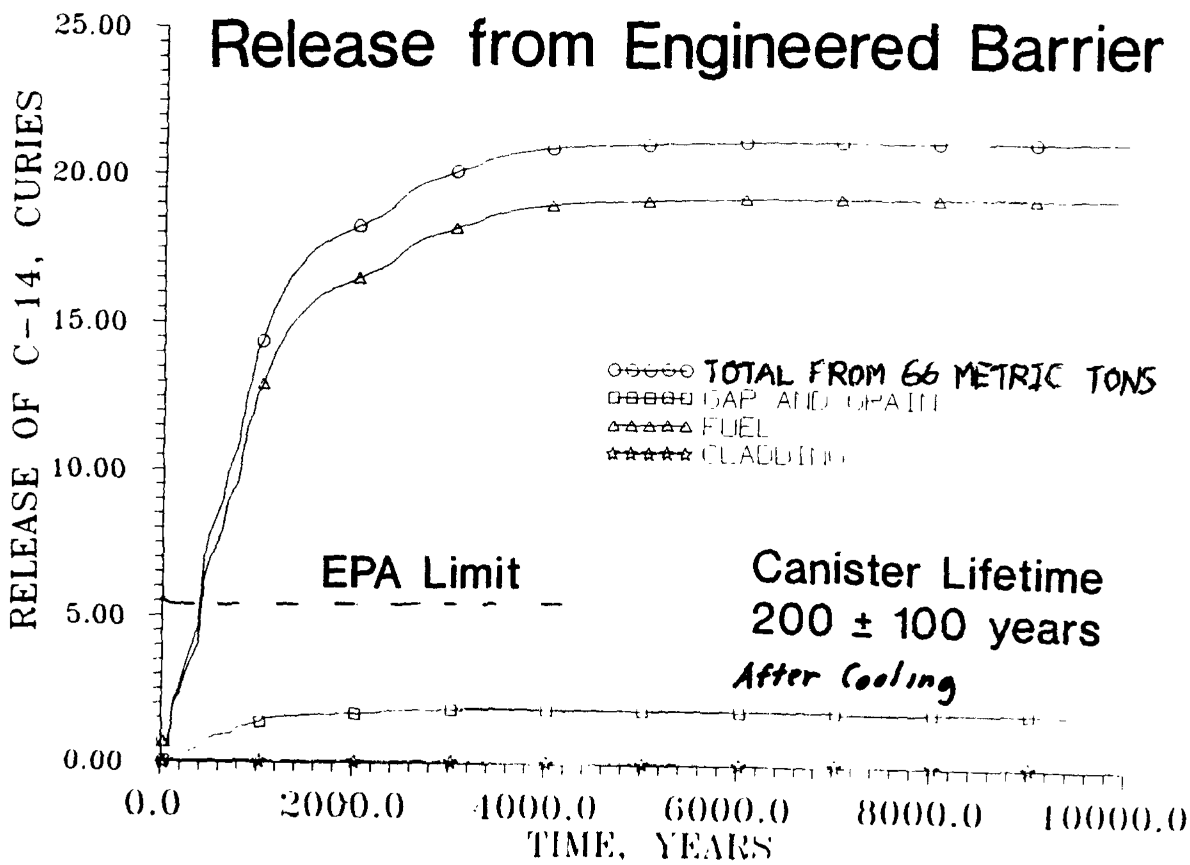
- Grain diameter from micrographs of fuel (20 microns)
- Outer layer diameter taken as fragment size (2mm)
- Weight gain from thermal gravametric analysis and dry bath experiments between 110 and 250 degrees C (PNL)
- Activation energy and diffusion coefficients adjusted for best fit to oxidation data on fuel fragments in 8 temperature ranges
- Little data found on C-14 releases from fuel oxidation

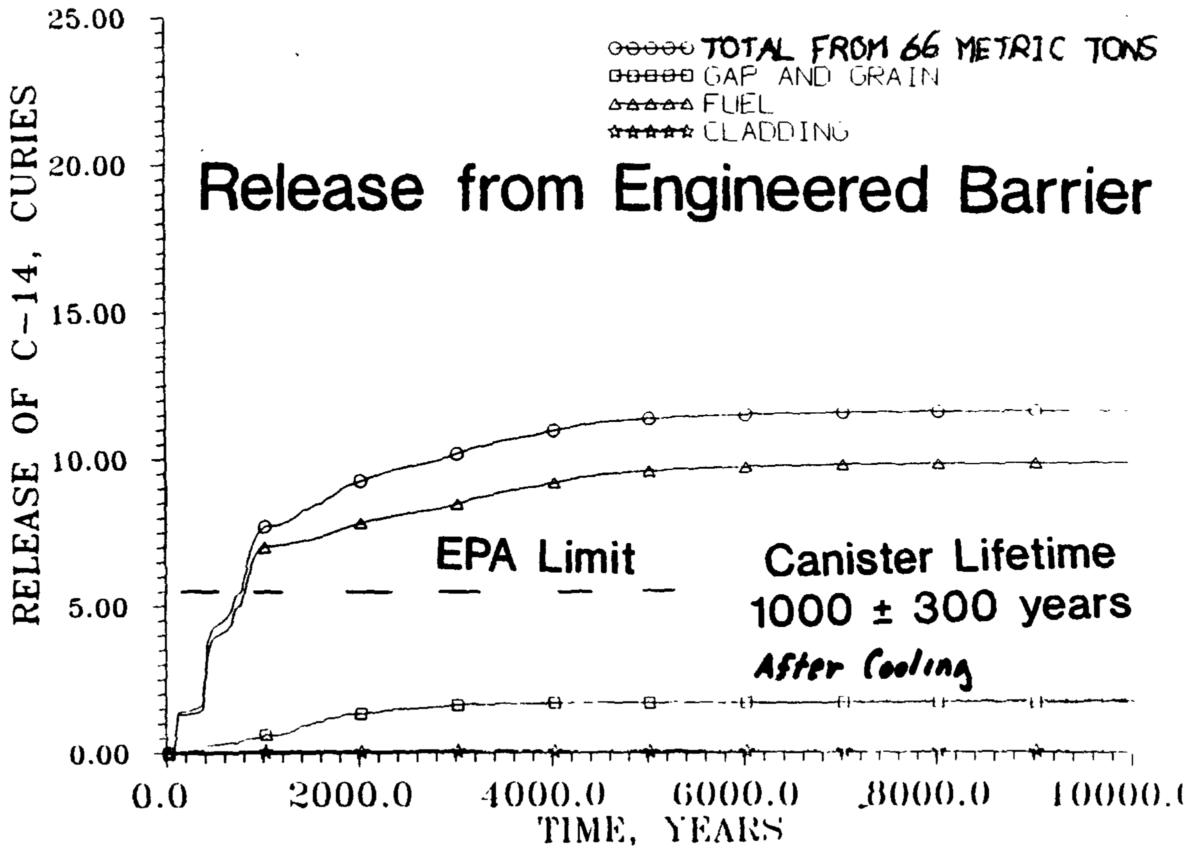

0.0187
130°C
Data
Model
Conversion
②
0
0
3,500
Time, hours


.984
Conversion
250°C
Data
Model
④
0
0
Time, hours
1,000


0.00733
①
Conversion
109°C
Data
Model
0
0
12,000
Time, hours


1.09
Conversion
225°C
Data
Model
③
0
0
Time, hours
400


Release from Engineered Barrier
RELEASE OF C-14, CURIES
TIME, YEARS
TOTAL FROM 66 METRIC TONS
GAP AND GRAIN
FUEL
CLADDING
EPA Limit
Canister Lifetime
200 ± 100 years
After Cooling
25.00
20.00
15.00
10.00
5.00
0.00
0.0
2000.0
4000.0
6000.0
8000.0
10000.0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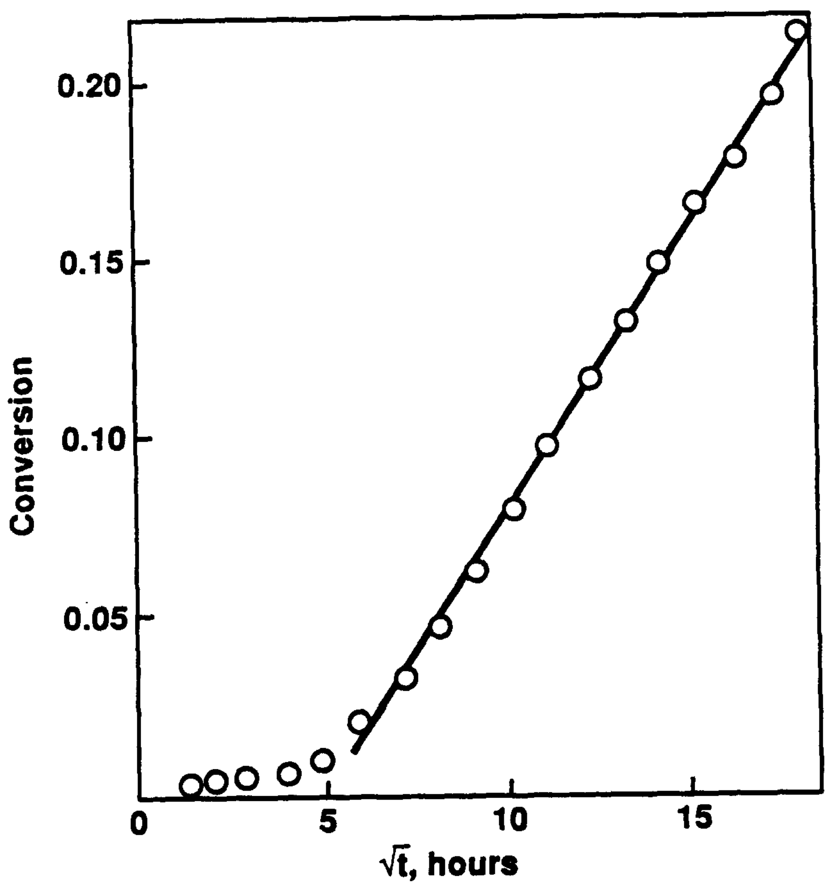
Canister Lifetime
1000 ± 300 years
After Cooling
25.00
20.00
15.00
10.00
5.00
0.00
0.0
2000.0
4000.0
6000.0
8000.0
10000.0

Transient Oxidation Effects



Not included in fuel oxidation model

Increase in Surface Area

(liquid and Gas Models)

- STORED ENERGY

$$\epsilon = \frac{E_Y}{2\ (1 - 2\ \nu)} \left(\frac{\Delta V}{3\ V}\right)^2$$

E_y : YOUNG'S MODULUS
ν : POISSON'S RATIO
ΔV : VOLUME CHANGE
V : VOLUME

- CONVERSION TO FRACTURE ENERGY

Disruptive Releases

- Intrusive Volcanism
  -Dike 1000 - 4000 m long, 1-10 m wide
- Extrusive Volcanism
  - Cindercone, 25 - 100 m radius
- Drilling intercepts repository
  - Brings up contents of waste package
  - Brings up contaminated rock

COMPARISON OF INVENTORY TO CURRENT EPA 10,000-YEAR CUMULATIVE RELEASE LIMIT AT ACCESSIBLE ENVIRONMENT AND NRC 10CFR60.113 MAXIMUM RELEASE RATES FROM THE ENGINEERED BARRIER SYSTEM*

GASEOUS RADIONUCLIDES	INVENTORY AT 1,000 YEARS (Ci)	EPA 10,000-YEAR CUMULATIVE RELEASE LIMIT, Ci (ANNUAL AVG. Ci/YR)	R	NRC POST-CONTAINMENT PERIOD RELEASE LIMIT FROM EBS (Ci/YR)
^{14}C	62,000	6,200 (0.62)	(10)	**1.07
^{129}I	1,950	6,200 (0.62)	(.31)	**1.07

SEMI-VOLATILE RADIONUCLIDES				
^{79}Se	25,050	62,000 (6.2)	(.4)	**1.07
^{99}Tc	806,000	620,000 (6.2)	(1.3)	8.06
^{135}Cs	21,390	62,000 (6.2)	(.34)	**1.07

*BASED ON 62,000 MTIHM SPENT FUEL
**NUCLIDES FOR WHICH THE MAXIMUM RELEASE RATE [illegible] GREATER THAN 1 X 10^{-5} PER YEAR INVENTORY BECAUSE OF THEIR SMALL INVENTORIES

(Park, 1991)

VAPOR PRESSURES

SPECIES	VAPOR PRESSURE (ATMOSPHERES)	
	100°C	200°C
CO_2	> 2,000	> 12,000
I_2	6×10^{-2}	3.7
SeO_2	9.1×10^{-4}	5.4×10^{-2}
Tc_2O_7	1.2×10^{-4}	3.7×10^{-2}

(FROM LANGE'S HANDBOOK OF CHEMISTRY, 13TH EDITION, 1985)

(Park. 1991)

Biggest Information Needs for Source term

- Integrity of canisters?
- How does water get into canisters?
  - drying of rock
  - saturation and rewetting
  - flow rate back to canisters
- How does water interact with fuel?
- Does cladding offer protection?
- Are kinetic effects important? Will colloids form as fuel disintegrates, and are they important for transport of randionuclides?

Conclusions

NRC SOTEC model includes:

- Waste package failure
- Release of liquid (dissolved) radionuclides
  - Colloids treated by empirical data
- Gaseous releases of C-14

Other analyses and codes for:

- Other gaseous nuclides
- volcanic intrusions
- drilling

Work at NRC and CNWRA in Support of Source Term

- EPSPAC - Detailed Source Term Model
- Natural Analogs
  - Alligator Rivers
  - Pena Blanca
  - Santorini
  - Cigar Lakes
  - Oklo
- Kinetic Effects of Fuel Dissolution
- Thermodynamic Properties of Actinides in High-Temperature Aqueous Solutions
- Metallic Phases in Spent Fuel