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

PRESENTATION TO
THE NUCLEAR WASTE TECHNICAL REVIEW BOARD

SUBJECT: MODELS OF WASTE PACKAGE
BEHAVIOR IN A REPOSITORY
ENVIRONMENT

PRESENTER: DR. THOMAS H. PIGFORD

PRESENTER'S TITLE
AND ORGANIZATION: DEPARTMENT OF NUCLEAR ENGINEERING & LAWRENCE
BERKELEY LAB, UNIVERSITY OF CALIFORNIA, BERKELEY, CA.

PRESENTER'S TELEPHONE NUMBER: (415) 642-6469

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MAY 16-17, 1989
SCOPE OF PRESENTATION

- OBJECTIVES
- DEVELOPMENT OF MODELS
- RESULTS OF CURRENT WORK
- FUTURE ACTIVITIES
THE DESIGN OBJECTIVE

TO DESIGN A REPOSITORY THAT PERFORMS SUFFICIENTLY WELL WHEN MEASURED AGAINST CRITERIA FOR SATISFACTORY LONG-TERM PROTECTION OF PUBLIC HEALTH AND SAFETY

THE PURPOSE OF PERFORMANCE PREDICTIONS

- TO MAKE RELIABLE PREDICTIONS THAT PERFORMANCE WILL NOT FALL OUTSIDE THE CRITERIA FOR ACCEPTABILITY
- PREDICTING ALL DETAILS OF REPOSITORY PERFORMANCE IS NEITHER NECESSARY OR ACHIEVABLE

PREDICTIVE RELIABILITY

PREDICTIVE RELIABILITY IN DESIGN DEPENDS ON:

- CLEAR AND RELIABLE CRITERIA FOR ACCEPTABLE PERFORMANCE
- A THEORY THAT CAN RELIABLY PREDICT THE PERFORMANCE DOES NOT FALL OUTSIDE THE CRITERIA FOR ACCEPTABLE PERFORMANCE
- RELIABLE PARAMETER VALUES TO APPLY THE THEORY
CONCEPTUAL WASTE PACKAGE DESIGNS FOR HIGH-LEVEL WASTE AND SPENT NUCLEAR FUEL

HIGH-LEVEL WASTE CONTAINER

SPENT NUCLEAR FUEL CONTAINER

LIFTING FIXTURE

3/8 IN. (1 cm)

DISPOSAL CONTAINER

POUR CANISTER 24 IN. (61 cm) O.D.

10.5 ft (328 cm)

26 IN. (66 cm) O.D.

18 CONSOLIDATED BWR ASSEMBLIES
WEIGHT 13,210 LB

6 INTACT BWR ASSEMBLIES
WEIGHT 6,440 LB

3 INTACT PWR ASSEMBLIES
WEIGHT 6,970 LB

6 CONSOLIDATED PWR ASSEMBLIES
WEIGHT 11,640 LB

FUEL ASSEMBLIES 5.5" x 5.5" MAX.
VARIERS

0.035 IN TYP

FUEL ASSEMBLIES 8.5" x 8.5" MAX.
VARIERS

0.035 IN TYP

FUEL ASSEMBLIES 10.5" x 10.5" MAX.
VARIERS

0.035 IN TYP

FUEL RODS

HARDWARE
THREE GENERAL MODELS FOR WASTE PACKAGE RELEASES

1. DRY

2. WET-DRIP

3. WET-CONTINUOUS
1. DRY CASE FOR WASTE PACKAGE RELEASES

A. SOME CONTAINERS FAIL, ALLOWING RADIOACTIVE GASES TO ESCAPE
   (e.g., $^3$H, $^{14}$C, $^{85}$Kr, $^{129}$I)

B. NO WATER ENTERS THE PENETRATIONS IN THE FAILED CONTAINERS. NO NON-VOLATILE RADIONUCLIDES ARE RELEASED
2. WET-DRIP CASE

- Bounding calculations of radionuclide release rates to ground water for individual waste packages at Yucca Mountain
- Assume no cladding barrier
- Assume ground water enters and leaves defective waste package
- Bathtub and flow-through models (assuming no diffusive pathway to surrounding rock)

(a) Bulk-flow solubility-limited (e.g., U, Pu, Np, Am)

\[ \dot{m}_i = Q \frac{n_i}{n_e} N^*_e, \quad f_i = \frac{\dot{m}_i}{M_i} \]

Where

- \( f_i \) is the fractional release rate of species \( i \)
- \( n_i \) is the concentration of element containing species \( i \) in waste
- \( n_e \) is the concentration of isotope \( i \) in waste
- \( \dot{m}_i \) is the release rate of species \( i \)
- \( M_i \) is the 1,000-year inventory of species \( i \)
- \( N^*_e \) is the solubility of element containing isotope \( i \)
- \( Q \) is the volumetric flow rate of ground water per waste package
(b) LIMITED BY WASTE-WATER REACTION RATE (e.g., SOLUBLE Tc-99)

\[ \dot{m}_i = j_{\eta} A \frac{n_{i}}{n_{e}}, \quad f_i = \frac{\dot{m}_i}{M_i} \]

WHERE \( j_{\eta} \) IS THE RATE OF SOLID-SOLID ALTERATION OF THE WASTE MATRIX, (e.g., \( UO_2 (s) \rightarrow U_3O_7 (s) \)), PER UNIT AREA

\( A \) IS THE SURFACE AREA OF WASTE EXPOSED TO GROUND WATER
(c) READILY SOLUBLE SPECIES (e.g., Cs, I, not in UO$_2$ MATRIX)

\[ \dot{m}_{is} = M_{is} \frac{d\psi}{dt}, \quad f_{is} = \frac{\dot{m}_{is}}{M_i} \]

WHERE
\[ \frac{d\psi}{dt} \]
IS THE FRACTION OF SOLUBLE INVENTORY $M_{is}$ EXPOSED TO WATER PER UNIT TIME

KEY PARAMETERS: $M_{is}, M_i, \frac{d\psi}{dt}$
WASTE PACKAGE IN UNSATURATED TUFF WET-DRIP CASE
(POST-THERMAL PERIOD)

\[ Q \geq 0 \]
THE "BATHTUB" SCENARIO INVOLVES BREACHES THAT ALLOW WATER TO ENTER AND FILL THE PACKAGE, IMMERSING THE WASTE FORM.
RELEASE RATE OF Pu-239 & Pu-240, BATHTUB MODEL
SOURCE: O'CONNELL, 1989

Q = 1 LITER/YEAR

CURIE PER YEAR

TIME, YEARS

4.0x10^-9

5.0x10^-9

6.0x10^-9

7.0x10^-9

8.0x10^-9

0 2000 4000 6000 8000 10000

PU-239

PU-240
Q = 1 LITER/YEAR

UO₂ ALTERATION RATE 0.001 PER YEAR

FRACTIONAL RELEASE RATE, PER YEAR

6.0x10⁻⁴

4.0x10⁻⁴

2.0x10⁻⁴

0.0

0 2000 4000 6000 8000 10000

TIME, YEARS

FRACTIONAL RELEASE RATE OF Tc-99 BATHTUB MODEL
SOURCE: O'CONNELL, 1989
3. WET-CONTINUOUS CASE

- DIFFUSIVE-ADVECTIVE MASS TRANSFER - DETAILED ANALYTIC SOLUTIONS

- PRESENT ANALYSES ASSUME WASTE CONTAINER AND FUEL CLADDING ARE NOT PRESENT

- ASSUMES A CONTINUOUS LIQUID DIFFUSION PATHWAY FROM THE WASTE TO THE TUFF

(a) SATURATION CONCENTRATION OF SPECIES i AT WASTE SURFACE

COMPLETE TIME-DEPENDENT ANALYTIC SOLUTIONS FOR SINGLE SPECIES OR DECAY CHAINS, FOR WASTE IN CONTACT WITH TUFF, OR SURROUNDED BY BACKFILL AND TUFF, NO ARBITRARY OR ADJUSTABLE PARAMETERS
(a) SATURATION CONCENTRATION OF SPECIES \( i \) AT WASTE SURFACE

(CONTINUED)

KEY PARAMETERS FOR THE LOW-FLOW AT YUCCA MOUNTAIN, ASSUMING GROUND WATER IN POROUS MEDIA ONLY

- \( D \) COEFFICIENT OF MOLECULAR DIFFUSION IN PORE WATER
- \( N^* \) ELEMENTAL SOLUBILITY FOR THE SPECIES
- \( R \) RADIUS OF SPHERICAL-EQUIVALENT WASTE SOLID
- \( n_i \) CONCENTRATION OF ELEMENT CONTAINING SPECIES \( i \) IN WASTE
- \( n_{i\text{iso}} \) CONCENTRATION OF ISOTOPE \( i \) IN WASTE
- \( \dot{m}_i \) RELEASE RATE OF SPECIES \( i \)
- \( M_i \) 1,000-YEAR INVENTORY OF SPECIES \( i \)
- \( \varepsilon \) POROSITY
- \( \lambda \) DECAY CONSTANT
- \( K \) RETARDATION FACTOR DUE TO SORPTION
- \( \ell \) THICKNESS OF BACKFILL, IF PRESENT
- \( S \) MOISTURE CONTENT

EXTENDED SOLUTIONS INCLUDE EFFECT OF TEMPERATURE CHANGES DUE TO REPOSITORY HEATING AND SUBSEQUENT COOLING.

IF GROUND WATER EXISTS IN THE FRACTURES AND IN POROUS ROCK, ADDITIONAL PARAMETERS ARE:

- FRACTURE SPACING
- FRACTURE APERTURE

GROUND WATER FLOW IS NOT A PARAMETER, UNLESS IT BECOMES MANY ORDERS OF MAGNITUDE GREATER THAN CURRENTLY ESTIMATED FOR YUCCA MOUNTAIN

NWFRDSP A135-16, 17-89 14
3. WET-CONTINUOUS CASE

(continued)

(b) READILY SOLUBLE SPECIES (e.g., Cs, I, NOT IN UO₂ MATRIX)

COMPLETE TIME-DEPENDENT ANALYTIC SOLUTIONS FOR SINGLE SPECIES FOR WASTE IN CONTACT WITH TUFF, OR SURROUNDED BY BACKFILL AND TUFF, NO ARBITRARY OR ADJUSTABLE PARAMETERS

KEY PARAMETERS FOR THE LOW-FLOW AT YUCCA MOUNTAIN

D  COEFFICIENT OF MOLECULAR DIFFUSION IN PORE WATER
ε  POROSITY
V  VOLUME WITHIN WASTE PACKAGE THAT CAN BE FILLED WITH WATER
λ  DECAY CONSTANT
K  RETARDATION FACTOR DUE TO SORPTION
l  BACKFILL THICKNESS, IF PRESENT
S  MOISTURE CONTENT (FRACTION)
Mᵢₛ  INVENTORY OF SPECIES i IN READILY SOLUBLE FORM
EXTENDED SOLUTIONS INCLUDE EFFECT OF TEMPERATURE CHANGES DUE TO REPOSITORY HEATING AND SUBSEQUENT COOLING
3. WET-CONTINUOUS CASE

(CONTINUED)

(c) DISSOLUTION OF SPECIES $i$ CONGRUENT WITH THAT OF A DOMINANT LOW-SOLUBILITY SPECIES $m$

$$f_i = \frac{\dot{m}_i}{M_i} = f_m = \frac{\dot{m}_m}{M_m} , \dot{m}_i = \dot{m}_m \left( \frac{M_i}{M_m} \right)$$

KEY PARAMETERS FOR $\dot{m}_m$ GIVEN IN 3(a)

(d) DISSOLUTION OF SPECIES $i$ CONGRUENT WITH THE RATE OF ALTERATION OF THE WASTE MATRIX

$$\dot{m}_i = j_m A \frac{n_i}{n_m} , f_i = \frac{\dot{m}_i}{M_i}$$
WET-CONTINUOUS CASE
A WASTE PACKAGE IN SATURATED TUFF
STREAMLINES IN POROUS ROCK

PORE VELOCITY

PLUME OF CONTAMINATED GROUNDWATER

CONCENTRATION ISOPLETH

WASTE-FORM SURFACE

CONCENTRATION IN GROUNDWATER

VELOCITY AND CONCENTRATION PROFILES FOR GROUNDWATER FLOWING AROUND A WASTE CYLINDER
3. WET-CONTINUOUS CASE
MASS TRANSFER BY DIFFUSION AND ADVECTION
(NO RADIOACTIVE DECAY)

\[ f_j = \frac{8 N_j^* D^{\frac{3}{2}} \epsilon V^{\frac{1}{2}} (1 + R/L)}{(\pi R)^{\frac{3}{2}} n_j}, \quad \frac{VR}{D} > 4 \]

- \( f_j \) = FRACTIONAL DISSOLUTION RATE OF SPECIES \( j \)
- \( N_j^* \) = SATURATION CONCENTRATION
- \( D \) = DIFFUSION COEFFICIENT IN GROUND WATER
- \( \epsilon \) = POROSITY
- \( V \) = GROUNDWATER PORE VELOCITY
- \( R \) = CYLINDER RADIUS
- \( L \) = CYLINDER LENGTH
- \( n_j \) = CONCENTRATION IN WASTE SOLID
NOTATION FOR FIGURES

d DIAMETER
D DIFFUSION COEFFICIENT
\( f_j \) FRACTIONAL DISSOLUTION RATE OF SPECIES \( j \)
h MASS-TRANSFER COEFFICIENT
\( j_0 \) FORWARD REACTION RATE
K RETARDATION COEFFICIENT
r RADIUS
t TIME
V GROUND WATER PORE VELOCITY
\( \alpha \) BACKFILL PERMEABILITY
\( \beta \) ROCK POROSITY
\( \epsilon \) POROSITY
\( \lambda \) RADIOACTIVE DECAY CONSTANT
NOTATION FOR FIGURES
(CONTINUED)

PECLET NUMBER

\[ Pe = \frac{V_r}{D} \]

\[ \lambda \text{ Kr} \]

DAMKÖHLER NUMBER

\[ Da = \frac{V}{D} \]

THIELE MODULUS

\[ \Lambda = \left( \frac{\lambda d^2 K}{D} \right)^{1/2} \]

FOURIER MODULUS

\[ T = \frac{D t}{K d^2} \]

SHERWOOD NUMBER

\[ Sh = \frac{hd}{D \varepsilon} \]

REACTION-RATE MODULUS

\[ R = \frac{j_o r}{\varepsilon DN^*} \]
NORMALIZED SOLUBILITY-LIMITED RELEASE RATE AS A FUNCTION OF TIME FLOW ACROSS A WASTE CYLINDER

\[ da = \frac{K \lambda R}{U} = 50 \]

- \( U = 1 \text{ m/yr} \)
- \( R = 0.15 \text{ m} \)
- \( D = 10^{-5} \text{ cm}^2/\text{sec} \)
- \( \varepsilon = 0.01 \)
- \( K = 10 \)

TIME, YEARS

\( f_n/N^*, \text{ yr}^{-1} \)
3. WET-CONTINUOUS CASE
MASS TRANSFER BY DIFFUSION
(NO RADIOACTIVE DECAY)

\[ f_j = \frac{\beta eD_n^*}{n_j}, \text{ as } V \to 0 \]

SPHERE: \( \beta = \frac{3}{R^2} \)

PROLATE SPHEROID: \( \beta = \frac{3e}{b^2 \ln \left( \coth \frac{\alpha_s}{2} \right)} \)

\( e = \text{ ECCENTRICITY} \)
\( b = \text{ SEMI-MINOR AXIS} \)
\( \alpha_s = \cosh^{-1} \left( \frac{1}{e} \right) \)

TIME FOR 99% OF STEADY STATE:
\[ t = \frac{10^4 K b^2}{\pi D} \left[ \sinh \alpha_s \ln \left( \coth \frac{\alpha_s}{2} \right) \right]^2 \]

\( K = \text{ RETARDATION FACTOR} \)
NORMALIZED MASS-TRANSFER RATE AS A FUNCTION OF TIME AND RETARDATION COEFFICIENT (K);
DIFFUSION FROM SPHERICAL WASTE FORM

\[ U = 0 \]
\[ \lambda = 0 \]
\[ R = 44 \text{cm} \]
\[ \varepsilon = 0.01 \]
\[ D = 10^{-5} \text{cm}^2/\text{sec} \]
RELEASE RATE AS A FUNCTION OF GROUND WATER VELOCITY
DIFFUSIVE-ADVECTIVE MASS TRANSFER
(UO₂ IN AN OXIDIZING ENVIRONMENT)

FRACTIONAL DISSOLUTION RATE, YR⁻¹

GROUND WATER PORE VELOCITY, cm/yr

10⁻⁹

10⁻⁸

10⁻⁷

10⁻⁶

10⁻⁵

10⁻⁴

10⁻³

10⁻²

10⁻¹

10⁰

10¹

10²

10³

10⁴

DIFFUSION-ADVECTION

100 YR, K = 100

DIFFUSION

STEADY-STATE

NOT VALID

L = 300cm
R = 15cm
N₅ = 5 X 10⁻⁵ g/cm³ (Kerrisk)
D = 32 cm²/yr
ε = 0.1
R = 48.6cm
COMPARISON OF WET-DRIP AND WET-CONTINUOUS CASES
FRACTIONAL RELEASE RATES FOR UO₂ IN AN
OXIDIZING ENVIRONMENT

\[ f_i = \frac{\pi R_i^2 V_i N_i^-}{3 \pi R_i^2 n_i} = \frac{3 V_i \epsilon N_i^-}{4 R_i n_i} \]

- **$L = 300$ cm**
- **$R_e = 15$ cm**
- **$N_i^- = 5 \times 10^{-5}$ g/cm³ (Kerrisk)**
- **$D = 32$ cm²/yr**
- **$\epsilon = 0.1$**
- **$R_e = 48.6$ cm**
WET-CONTINUOUS CASE, LOW-FLOW RELEASE RATES FOR DEFENSE WASTE BOROSILICATE GLASS

<table>
<thead>
<tr>
<th>ELEMENT</th>
<th>CONCENTRATION IN WASTE(^a) (g/cm(^3))</th>
<th>SATURATION CONCENTRATION(^b) (g/m(^3))</th>
<th>FRACTIONAL RELEASE RATE(^c) (PER YEAR)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>7.6 x 10(^{-1})</td>
<td>3.2 x 10(^{2})</td>
<td>7.6 x 10(^{-7})</td>
</tr>
<tr>
<td>U</td>
<td>5.4 x 10(^{-2})</td>
<td>4.7 x 10(^{-2})</td>
<td>1.6 x 10(^{-9})</td>
</tr>
<tr>
<td>Np</td>
<td>5.3 x 10(^{-5})</td>
<td>3.0 x 10(^{-4})</td>
<td>1.0 x 10(^{-8})</td>
</tr>
<tr>
<td>Pu</td>
<td>3.0 x 10(^{-4})</td>
<td>3.8 x 10(^{-10})</td>
<td>2.3 x 10(^{-15})</td>
</tr>
<tr>
<td>Am</td>
<td>4.1 x 10(^{-7})</td>
<td>1.5 x 10(^{-3})</td>
<td>6.6 x 10(^{-6})</td>
</tr>
<tr>
<td>Cs</td>
<td>2.0 x 10(^{-3})</td>
<td>6.2 x 10(^{-1})</td>
<td>5.6 x 10(^{-7})</td>
</tr>
</tbody>
</table>

NOTES:
A. DATA FROM BRUTON, 1987; WASTE DENSITY = 3 g/cm\(^3\)
B. DATA FROM BRUTON, 1987, AT 90 C; EXCEPT Cs SOLUBILITY FROM APTED, 1989
C. WASTE CYLINDER 0.3m RADIUS, 2.5m LONG; POROSITY = 0.16; DIFFUSION COEFFICIENT 10\(^4\) cm\(^2\)/s; LOW-FLOW STEADY STATE MASS TRANSFER ANALYSIS
EVOlUTION OF SOLUBILITY DATA FOR SPENT FUEL
SOLUBILITY (MOLES/L) AT 25 C

<table>
<thead>
<tr>
<th>ELEMENT</th>
<th>EA 1986</th>
<th>SCP 1988</th>
<th>LLNL 1989</th>
</tr>
</thead>
<tbody>
<tr>
<td>Am</td>
<td>$1 \times 10^{-8}$</td>
<td>$1 \times 10^{-6}$</td>
<td>$1.6 \times 10^{-10}$</td>
</tr>
<tr>
<td>Np</td>
<td>$3 \times 10^{-3}$</td>
<td>$1 \times 10^{-3}$</td>
<td>$6.3 \times 10^{-6}$ to $10^{-9}$</td>
</tr>
<tr>
<td>Pu</td>
<td>$1.8 \times 10^{-6}$</td>
<td>$1 \times 10^{-5}$</td>
<td>$4 \times 10^{-13}$ to $1.6 \times 10^{-4}$ NOTE a. $5 \times 10^{-5}$ to $2 \times 10^{-6}$ NOTE b.</td>
</tr>
<tr>
<td>U</td>
<td>$2.1 \times 10^{-4}$</td>
<td>$4 \times 10^{-3}$</td>
<td>$6.3 \times 10^{-5}$ NOTE c.</td>
</tr>
</tbody>
</table>

NOTES:
A. AS PuO$_2$
B. AS AMORPHOUS Pu(OH)$_4$
C. IN EQUILIBRIUM WITH ATMOSPHERIC OXYGEN
FRACTIONAL RELEASE RATES FROM SPENT FUEL INTO ROCK AS A FUNCTION OF TIME
(10cm OF VOID WATER, 1% OF TOTAL CESIUM AND IODINE IS IN GAP, D = 0.12 M²/YR, NON-OXIDIZING ENVIRONMENT)
FRACTIONAL RELEASE RATES FROM SPENT FUEL INTO ROCK AS A FUNCTION OF TIME
(10 cm OF VOID WATER, 1% OF TOTAL CESIUM AND IODINE IS IN GAP,
D = 0.12 m²/yr, OXIDIZING ENVIRONMENT)

FRACTIONAL DISSOLUTION RATE, yr⁻¹

TIME, YEARS

10⁻⁷ 10⁻⁶ 10⁻⁵ 10⁻⁴ 10⁻³ 10⁻²

TOTAL

MATRIX

GAP

CS-135

10⁰ 10¹ 10² 10³ 10⁴ 10⁵ 10⁶
RELEASE RATES OF SOLUBLE SPECIES THROUGH 0.3m OF BACKFILL

FRACTIONAL RELEASE RATE INTO ROCK, YR⁻¹

TIME, YEARS

10⁻²  10⁻¹  10⁰  10¹  10²  10³  10⁴  10⁵  10⁶  10⁷

1-129
Cs-135
Cs-137
RELEASE RATES OF SOLUBLE Cs-137 SPECIES AS A FUNCTION OF BACKFILL THICKNESS

FRACTIONAL RELEASE RATE YR\(^{-1}\)

NO BACKFILL

15 cm BACKFILL

30 cm

70 cm

TIME, YEARS

10\(^0\) 10\(^{-1}\) 10\(^{-2}\) 10\(^{-3}\)
OTHER USUAL ASSUMPTIONS IN MASS-TRANSFER THEORY

1. EFFECT OF A LIQUID-FILLED ANNULUS BETWEEN WASTE & ROCK
2. EFFECT OF FLOW DIRECTION AND GEOMETRY
3. HYDRODYNAMIC DISPERSION
4. EFFECT OF RADIOACTIVE DECAY
5. LOCAL SORPTION EQUILIBRIUM
6. LINEAR SORPTION: CONSTANT DISTRIBUTION COEFFICIENT, CONSTANT RETARDATION COEFFICIENT
7. SURFACE DIFFUSION
8. INTERFERENCE FROM OTHER WASTE PACKAGES
9. POROUS OR FRACTURED ROCK
10. CONSTANT TEMPERATURE
11. CONSTANT AND UNIFORM CHEMICAL ENVIRONMENT
12. NO RADIOACTIVE DECAY PRECURSOR
FRACTIONAL RELEASE RATES OF A RADIONUCLIDE DECAY CHAIN, NORMALIZED TO 1000 YEAR INVENTORIES

NOTE: Th-230 AND Ra-226 ASSUMED TO BE RELEASED CONGRUENTLY WITH U-234
FRACTIONAL DISSOLUTION RATE FOR SILICA NORMALIZED TO INITIAL INVENTORY - EFFECT OF REPOSITORY HEATING

\[ M^0 = 470 \text{kg} \]
\[ K = 100 \]
\[ R = 42 \text{cm} \]
\[ \epsilon = 0.01 \]

TIME AFTER EMPLACEMENT, YR

FRACTIONAL DISSOLUTION RATE, YR\(^{-1}\)

CONSTANT \( T \)

\( T(t) \)
EFFECT OF BACKFILL POROSITY ($\varepsilon$) (NO RADIOACTIVE DECAY)

BACKFILL (1) ROCK (2)

INNER RADIUS, cm 65.9 95.9
RETARDATION $10^3$ $10^3$
POROSITY $\varepsilon_1$ 0.01

INTO BACKFILL $\varepsilon_1 = 0.2$

INTO ROCK $\varepsilon_1 = 0.2$

MASS TRANSFER RATE
CONCENTRATION AT INNER SURFACE, cm$^3$/SEC

$10^{-1}$ $10^{-2}$ $10^{-3}$ $10^{-4}$ $10^{-5}$

$10^1$ $10^2$ $10^3$ $10^4$ $10^5$ $10^6$ $10^7$
EFFECT OF RETARDATION (K) IN BACKFILL AND ROCK
(NO RADIOACTIVE DECAY)

INTO BACKFILL

BACKFILL (1) ROCK (2)

INNER RADIUS, cm 65.9 95.9
POROSITY 0.2 0.01

$K_1 = 10, K_2 = 10$

TIME, YEARS

MASS TRANSFER RATE, cm$^3$/SEC

CONCENTRATION AT INNER SURFACE

$10^3, 10$

$10^3, 10^3$

$10, 10$

$10^3, 10^3$

$10^3, 10$

INTO ROCK
TRANSPORT OF RADIONUCLIDES IN THE BACKFILL SURROUNDING A WASTE FORM IN FISSURED ROCK

BACKGROUND

- In most kinds of rock, there are numerous fractures
- Fractures have larger permeability than rock

ASSUMPTION

- Water-saturated backfill
- No canister
- Constant spacing fracture and perpendicular to waste form
- Constant surface concentration
- No groundwater flow in backfill
- Completely impervious rock
RELEASE INTO BACKFILL AND INTO FISSURED ROCK

\[ \lambda d^2/D = 10 \]

\[ \lambda d^2/D = 0 \]

\[ a = 2\text{m (waste length)} \]
\[ b = 1\text{cm (fracture width)} \]
\[ d = 15\text{cm (backfill thickness)} \]
\[ \text{Sh} \equiv hd/\varepsilon D_f = 1 \]
\[ D \equiv D_f/K \]

\[ \frac{\dot{m}}{2\pi t_2 b c_h} \] vs. \[ Dt/d^2 \]
EFFECT OF FLOW THROUGH BACKFILL ON RELEASE RATE
(RELEASE RATE IS PROPORTIONAL TO SHERWOOD NUMBER)

\( \beta = 0.0333 \)

\[
\begin{align*}
\text{ASSUMED REFERENCE CONDITION} & \quad \alpha = 0.01 \\
\text{NO FLOW THRU BACKFILL} & \quad \alpha = 0 \\
\text{CRUSHED ROCK} & \quad \alpha = 10
\end{align*}
\]
EFFECT OF FLOW RATE AND REACTION RATE ON RELEASE RATE (RELEASE RATE IS PROPORTIONAL TO THE SHERWOOD NUMBER)

INTEGRAL METHOD
ASYMPTOTIC - LARGE Pe
ASYMPTOTIC - SMALL Pe

R = 1000
R = 10
R = 1

PECLET NUMBER
MODEL FEATURES

- \(^{14}\text{C}\) is released as gaseous \(^{14}\text{CO}_2\) at the repository horizon
- \(^{14}\text{CO}_2\) is advected upward through a fracture
- Some of the \(^{14}\text{CO}_2\) dissolves into the pore water of the adjacent wet-rock matrix
- Equilibrium is maintained at the fracture interface between \(^{14}\text{CO}_2\) in the gas phase and dissolved \(^{14}\text{C}\) in the liquid phase
- The dissolved \(^{14}\text{C}\) diffuses transversely in the wet-rock matrix
$^{14}$CO$_2$ CONCENTRATION VS. DISTANCE FOR IMPULSE RELEASE FROM INFINITE PLANE SOURCE (0.04 m/yr)
PEAK SURFACE CONCENTRATION OF $^{14}$CO$_2$ (Z = 350M) VS. GAS DARCY VELOCITY FOR BAND (1000 yr) RELEASE FROM INFINITE PLANE SOURCE
PEAK SURFACE CONCENTRATION OF $^{14}\text{CO}_2$ ($Z = 350\text{M}$) VS. BAND - RELEASE DURATION FOR INFINITE PLANE SOURCE
CALCULATED TEMPERATURE FOR REFERENCE
WASTE PACKAGE IN TUFF

TIME AFTER EMBLACEMENT (YRS)

TEMPERATURE (°C)

PEAK FUEL
CONTAINER SURFACE
HOST ROCK AT BOREHOLE SURFACE
HOST ROCK 1M FROM BOREHOLE SURFACE

WASTE FORM - SPENT FUEL (PRESSURIZED WATER REACTOR)
LOCAL POWER DENSITY - 57.0 KW/ACRE
AREAL POWER DENSITY - 48.4 KM/ACRE
AVERAGE PACKAGE POWER AT BURIAL - 3.3 KW
(10 YRS OUT OF REACTOR)
CONTAINER DIAMETER - 0.7 M
PACKAGE SPACING - 5M
DRIFT SPACING - 46.86 M
DIRECTORY NO. - P57V3.3A
(DOE-SCP, 1988 p. 7-14)
GAS IN A WASTE CONTAINER, RESULT OF WASTE COOLING AND FLOW THROUGH PENETRATIONS

ASSUMPTIONS:  FILL PRESSURE = 1 ATM
ASSUME EARLY FAILURE

LEGEND

FILL TEMPERATURE (K)  MOLE RADIUS (μm)
298  3, 5, 10
400  10
558  10

YEARS
FURTHER WORK NEEDED

FOCUS

1. REFINE AND VALIDATE PRESENT PREDICTIVE CAPABILITY
   - METHODOLOGY
   - DATA BASE FOR PARAMETERS

2. EVALUATE GASEOUS RELEASES: $^{14}$CO$_2$, $^3$H, $^{85}$Kr, $^{129}$I

3. ASSESS THE PREDICTION OF LOWER RELEASE RATES FROM MORE REALISTIC MODELS

4. EVALUATE ADDITIONAL PHENOMENA THAT CAN AFFECT RELEASE RATES
SPECIFIC EXAMPLES OF FURTHER WORK NEEDED

1. REFINE AND VALIDATE PRESENT PREDICTIVE CAPABILITY

- DEVELOP PROGRAM-WIDE UNDERSTANDING OF PRESENT CAPABILITY AND OF DATA NEEDS VIA TIG'S NEW TEST PROBLEMS

- COMPLETE DOCUMENTATION OF PRESENT CAPABILITY

- APPLY ADDITIONAL SUBMODELS ALREADY DEVELOPED

- REFINE DATA AND ANALYSES ON EFFECTIVE SATURATION CONCENTRATIONS, INCLUDING EFFECT OF AIR INFLOW THROUGH FAILED CONTAINERS

- MEASURE LIQUID DIFFUSION COEFFICIENTS AND CONNECTED POROSITY IN TUFF

- PERFORM EXPERIMENTS TO VALIDATE THE THEORY UNDERLYING THE PREDICTIVE MODELS
SPECIFIC EXAMPLES OF FURTHER WORK NEEDED

2. EVALUATE GASEOUS RELEASES

- DETERMINE THE TIME-DEPENDENT SOURCE AND AMOUNT OF $^{14}\text{CO}_2$, $^3\text{H}$, $^{85}\text{Kr}$, $^{129}\text{I}$ THAT CAN BE MOBILIZED WITHIN A FAILED WASTE PACKAGE AND THE RATE OF ESCAPE THROUGH PENETRATIONS

- EVALUATE THE SPACE-TIME-DEPENDENT RETARDATION COEFFICIENT FOR CARBON DIOXIDE, AS AFFECTED BY TEMPERATURE, pH, LOCAL MOISTURE CONTENT IN TUFF, CALCITE, AND GROUND-WATER EVAPORATION

- EVALUATE THE SPACE-TIME-DEPENDENT FLOW RATE OF AIR IN THE HEATED REPOSITORY

- DETERMINE THE EXTENT TO WHICH THE PEAK CONCENTRATIONS OF GASEOUS EFFLUENTS AT THE SUBSURFACE ARE AFFECTED BY THEIR RATE OF RELEASE AT THE REPOSITORY HORIZON
SPECIFIC EXAMPLES OF FURTHER WORK NEEDED

3. INVESTIGATE MORE REALISTIC MODELS THAT ARE EXPECTED TO PREDICT LOWER RELEASE RATES TO GROUND WATER

- DEVELOP A MODEL FOR THE TIME OF CONTAINER FAILURE BY LOCALIZED PENETRATIONS AND THE SIZE, GEOMETRY, AND GROWTH OF THOSE PENETRATIONS

- DETERMINE THE FLOW OF GROUND WATER THROUGH THE SMALL PENETRATIONS IN A FAILED CONTAINER

- DETERMINE THE RATE OF DIFFUSION AND CONVECTION OF DISSOLVED RADIONUCLIDES OUT OF CONTAINER PENETRATIONS

- DETERMINE THE EFFECT OF TRANSPORT BARRIERS DUE TO SMALL PENETRATIONS IN DEFECTIVE FUEL CLADDING

- DETERMINE THE EFFECT OF UNCONNECTED POROSITY IN UNSATURATED TUFF ON DIFFUSIVE TRANSPORT

- DETERMINE THE TRANSPORT THROUGH RUBBLE IN THE CONTAINER-ROCK ANNULUS
SPECIFIC EXAMPLES OF FURTHER WORK NEEDED

4. EVALUATE THE EFFECT OF ADDITIONAL PHENOMENA THAT CAN AFFECT RELEASE RATES

- AIR INLEAKAGE THROUGH FAILED CONTAINERS
  - EFFECT ON SATURATION CONCENTRATIONS
  - OXIDATION OF ZIRCALOY CLADDING
  - OXIDATION OF URANIUM DIOXIDE
  - MOBILIZATION OF GASEOUS SPECIES

- ALPHA RADIOLYSIS
  - CAN RESULT IN HIGHER OXIDATIONS STATES AND INCREASED SATURATION CONCENTRATION
  - A REDOX FRONT NEAR THE WASTE SURFACE AND AN INCREASED CONCENTRATION GRADIENT AT THE WASTE SURFACE CAN INCREASE RELEASE RATE INTO GROUND WATER

- DIFFUSIVE-ADVECTIVE MASS TRANSFER FROM WASTE SOLID TO WATER INSIDE A WASTE CONTAINER
  - POTENTIALLY IMPORTANT IF VOID SPACE INSIDE CONTAINER IS FILLED WITH LOW-POROSITY MATERIAL