NOTE TO R. WELLER

FROM T. AHN TA

SUBJECT : GLASS ISSUE

DATE : MARCH 9, 1993

Previously, we have considered that HLW glasses may not be important over spent fuels in assessing the source-term of radionuclides, simply because the inventory of radioactivities contained in HLW glass is only about 3 % of the total inventory. This is a very oversimplified judgment, ignoring the nature of radionuclides release from spent-fuel dissolution or from HLW-glass leaching. The attached handouts highlight results of an analysis made for the comparison of the actual release of actinides from spent-fuel dissolution to that from HLW-glass leaching, based on experimental data obtained under various conditions. From this analysis, we believe that there are important reasons why HLW glasses should be considered in the actinide source-term. This is the best judgment we can make at the present time, based on the currently available experimental data.

The actinide source-term should be assessed with the actual release of actinides in leachates. For instance, in the simplest case of solubility limit as the Pu release mechanism, the amount of Pu release will be proportional to the flow rate of groundwater, because the Pu solubility limit in each failed container (or borehole) is considered to be maintained under near static conditions. Therefore, the total release of Pu from the repository is proportional to the number of containers, not the radioactivity inventory. Only a small fraction of the total inventory of actinides will be released through this process. In reality, there appear to be circumstances under which the actinide activities released (per unit groundwater volume) from HLW-glass leaching are much larger than those from spent-fuel dissolution. This makes HLW glasses contribute to the overall release of actinides from the repository significantly. However, we need to investigate many uncertainties involved in obtaining this conclusion. An example of such uncertainties is the actinide composition of simulated HLW glasses.

In addition to the aforementioned issue of the actual release of actinides, there are other reasons why HLW glasses cannot be ignored. Si and Fe released from HLW-glass leaching can enhance the interaction rates of spent fuel with groundwater. These two are the primary elements responsible for the formation of the secondary phases during the spent-fuel dissolution. On the other hand, B is a neutron poison if there is the criticality problem in the performance of spent-fuel packages. Also, release rates of actinides from HLW-glass leaching can be very fast compared to those from spent-fuel dissolution, which may give rise to the potential problem of dose rate effects. The last concern in HLW glasses is the time frame imposed on solidifying Liquid HLW stored at four Sites. Due to the leakage of Liquid HLW, the solidification is an urgent task and the first hot operation is scheduled for 1994 at the Savannah River Site. For the proper solidification, the performance related to product consistency must be studied.

The attached handouts are particularly for the discussion of the issue on the actual release of actinides. As a first step to resolve the glass issue, a tele-

Legacy - ZO (Letter not entered into system)

conference will be made within the HLWM and the CNWRA.

The Attachment : As Stated.

CC : R. Ballard
C. Interrante, EBS
K. Chang, EBSPAC
M. Lee, IPA
P. Nair, CNWRA

IMMERSION TESTS (Activity in Leachates, pCi/mL)

	Glass (*1)	SF (*2)
Pu-(239+240)	910 (3,640)	1 (3,000 ~ 20,000)
Am-241	640 (2,360)	0.1 ~ 1 (5,000 ~ 30,000)
Np-237	35 (80)	0.1 (3 ~ 10)
Cm-244		1 (10,000 ~ 30,000)

*1 : All are approximate terminal values at 90° C in J-13 groundwater (Parentheses are for maximum values which were followed by drastic decreases).

[1] W. L. Ebert and J. K. Bates, Proceeding of the Third International High-Level Radioactive Waste Management Conference, pp.934, Las Vegas, NV, April 12-16, 1992.

[2] W. L. Ebert, J. K. Bates and T. J. Gerding, The Reaction of Glass during Gamma Irradiation in a Saturated Tuff Environment, Part 4, Argonne National Laboratory Report, ANL-90/13, 1990.

*2 : All are approximate terminal values at 85° C in J-13 groundwater (Parentheses are for maximum values which were followed by drastic decreases).

[3] C. N. Wilson, Mat. Res. Soc. Symp. Proc., 212, 197 (1991).

[4] C. N. Wilson, Results from NNWSI Series 3 Spent Fuel Dissolution Tests, Pacific Northwest Laboratory Report, PNL-7170, 1990.

DRIPPING GROUNDWATER TESTS (Activity in Leachates, pCi/mL)

	Glass (*1)	SF (*2)
Pu-(239+240)		$(2 \sim 4) \times 10^4$
Am-241	$(0.8 \sim 8) \times 10^5$	$(4 \sim 9) \times 10^4$

*1 : Assumed that activities come from Am-241 only. ATM-(1-c) and ATM-8 glasses have approximately 4 ~ 5 times more Pu-239 than SRL glasses, and the composition of Am-241 for these glasses are not reported. The above data are from ATM-10 glass of which compositions are not reported. Note that the WVDP inventory of Am-241 is over 20 times greater than that of Pu-(239+240).

[5] J. K. Bates, J. P. Bradley, A. Teetsov, C. R. Bradley, M. Buchholtz ten Brink, Science, 256, 649 (1992).

- Flow Rate Conversion : 5 mL/year x (60,000)/14 = ~ 20 L/year.
- Most of them can be trapped in tuff matrices but can be redissolved too.

*2 : For (20 ~ 40) μg/mL of UO_2 from 238 weeks' tests. Fractions of actinides are for Robinson fuels.

[6] D. J. Wronkiewicz, J. K. Bates, T. J. Gerding and E. Veleckis, J. Nuclear Materials, 190, 107 (1992).

[7] Site Characterization Plan, Department of Energy, DOE/RW-0199, 1988.

- Flow Rate Conversion : (2 ~ 8) mL/year x (500,000)/(20 ~ 500) = ~ (2 ~ 200) L/year.

UNCERTAINTIES

[A] S/V (Immersion Tests)

Glass :

Laboratory : 20 ~ 200 cm^{-1} [1]
Repository : ~ 0.4 cm^{-1} without cracks [7]

SF :

Laboratory : 0.2 cm^{-1} [3]
Repository : ~ 1 cm^{-1} without cracks [7]

[B] F/V (Immersion Tests)

Glass :

Laboratory : 0 (No Replenishment) [1]
Repository : 10^{-3} ~ 10^{-1} $year^{-1}$ [7]

SF :

Laboratory : 0.25 ~ 0.6 $year^{-1}$ [3]
Repository : 10^{-4} ~ 10^{-2} $year^{-1}$ [7]

[C] FACTORS IN THE INCREASE OF ACTIVITY FROM ACTINIDES SOLUBILITIES

25° C :

Pu-(239+240) : ~ 400 from SF results.
Am-241 : ~ 5 to ~ 6,000 (Carbonate Well Water) from SF results.

60° C :

Pu-(239+240) : ~ -15 (decrease) from 25° C results.
Am-241 : ~ 50 from 25° C results.

[8] H. Nitsche, Mat. Res. Soc. Symp. Proc., 212, 517 (1991).

[D] FRACTURE

Glass : ~ 50 from radiation without initial cracks.

SF : ~ 40 for transgranular cracking of U_4O_9.

[E] IRON EFFECTS

Glass :

Pu-239 (pCi/mL, terminal values)

	No Iron	With Iron
DI Water	45	990
Basalt	10	380
Brine	30	150

[9] Final Report of the Defense High-Level Waste Leaching Mechanisms Program, compiled by J. E. Mendel, Pacific Northwest Laboratory Report, PNL-5157, 1984.

SF :

- Most of Uranium released was plated out on iron coupons at 25° C in brine [10].

[10] W. J. Gray and G. L. McVay, in Proceedings of the Third Spent Fuel Workshop, edited by L. Werme, SKBF/KBS TR-83-76, 1984.

[F] CUMULATIVE EFFECTS

- The amount of particles can be accumulated in proportion to V/F.
- For SF, it can be ~ 350 at the flow rate of 1 L/year.

[G] AGITATION : Glass

Pu : 5,550 pCi/mL

Am : 3,500 pCi/mL

- PNL 76-68 Glass in DI water and silicate water at 90° C.
- Terminal Si concentration in particulate form is converted into Pu and Am using the actinide fraction in SRL 202A glass.

[11] J. W. Shade and D. M. Strachan, Am. Ceram. Soc. Bull., 65, 1568 (1986).

INVENTORY I

SF (year 2,020) : Ci for 116,921 MTHM (Parentheses : Ci/MTHM)

	Oversby [12]			Robinson [13]	Turkey Point [13]	EPA Limits
Ci/mL	10	300	1,000			
Pu-239	3.66×10^{7} (313)		3.57×10^{7} (305)			1.17×10^{4} (0.1)
Pu-240	6.16×10^{7} (527)		5.59×10^{7} (478)	8.70×10^{7} (744)	8.23×10^{7} (704)	1.17×10^{4} (0.1)
Am-241	1.98×10^{8} (1,690)	3.22×10^{8} (2,750)	1.04×10^{8} (893)	2.07×10^{8} (1,770)	1.77×10^{8} (1,510)	1.17×10^{4} (0.1)
Np-237	3.74×10^{4} (0.32)		1.17×10^{5} (1.0)	2.81×10^{4} (0.24)	2.57×10^{4} (0.22)	1.17×10^{4} (0.1)
Cm-244	1.54×10^{8} (1,320)	2.34×10^{3} (0.02)		1.50×10^{8} (1,280)	1.16×10^{8} (990)	

[12] V. M. Oversby, Important Radionuclides in High Level Nuclear Waste Disposal: Determination Using a Comparision of the EPA and NRC Regulations, Lawrence Livermore National Laboratory Report, UCRL-94222, 1986.

[13] C. N. Wilson, Results from NNWSI Series 2 Bare Fuel Dissolution Tests, Pacific Northwest Laboratory Report, PNL-7169, 1990.

- All inventory are calculated values (Ci per MTHM) using ORIGEN-2. Oversby results are 10, 300, and 1,000 years after discharge, Robinson fuels assumed 12 years from discharge, and Turkey Point fuels assumed 10.5 years from discharge. Measured inventory is only available for Robinson fuel, which is close to the calculated values. Cm-244 has the half life of 18.1 years and become Pu-240.

Glass (year 2,020) : Ci for 29,297 MTHM (Parentheses : Ci/MTHM)

	DWPF [7]	HWVP [14]	ICPP (14,*1)	WVDP [7]
Pu-239	74,000 (6.4)	25,303 (5.2)	23,162 (1.9)	1,700 (2.4)
Pu-240	47,000 (4.1)	6,197 (1.3)	14,711 (1.2)	1,300 (1.8)
Am-241	83,000 (7.2)	101,052 (20.9)	25,972 (2.1)	72,000 (100.4)
Np-237	68 (0.00592)	48 (0.00995)	21 (0.00171)	11 (0.0153)
Cm-244	1,300 (0.113) {44,356 (3.9) [14]}	1,648 (0.342)	407 (0.0332) {13,883 (1.1) [14]}	22,000 (30.7){7,436 (10.4) [14]}

*1 : Total activity conversion including fission products = 0.313 x (DWPF values).

[14] Integrated Data Base for 1991 : U. S. Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics, Department of Energy, DOE/RW-0006, Rev.7, 1991.

INVENTORY II

Glass (year 2,020) :

	DWPF	HWVP	ICPP	WVDP	TOTAL
MTHM [15]	11,483	4,824	12,273	717	29,297
Container Numbers [14]	5,282	2,280	5,800	300	13,662

[15] DWPF Orientation and Tour, August 7-8, The Department of Energy, 1991.

- 2.174 MTHM/DWPF-Canister [15]. Other canister weights are obtained by the conversion of the volumes of canisters from reference [14] using the conversion factor of 2.174 MTHM/DWPF-Canister : 2.116 MTHM/HWVP-Canister, 2.264 MTHM/ICPP-Canister, and 2.389 MTHM/WVDP-Canister.

SF (year 2,020) :

	BWR	PWR	TOTAL
MTHM [7]	45,850	71,071	116,921
MTIHM [14]	26,200	49,700	75,900
Number of Containers [7]	24,317 (24,259)	38,533 (36,014)	62,850 (60,273)

- 3 PWR fuel assemblies or 6 BWR fuel assemblies were used for each container. MTIHM for a BWR assembly is 0.18, and MTIHM for a PWR assembly is 0.46. Factors of 1.75 for BWR fuel and 1.43 for PWR fuel were used in the conversion of MTIHM to MTHM.
- The number of containers is the number of total fuel assemblies divided by the number of assemblies per container (Parenthese are values from the MTIHM conversion).

CUMULATIVE RELEASE OF ACTINIDES

CUMULATIVE RELEASE FOR 10,000 YEARS (CUMREL) =

[ACTIVITY PER UNIT LEACHATE VOLUME] x [FLOW RATE] x [NUMBER OF CONTAINERS] x [10,000 YEARS]

EPA LIMIT FOR TOTAL INVENTORY (EPAL) =

[EPA LIMIT PER 1000 MTHM] x [MTHM] x [0.001]

	ALL				Without ICPP			
pCi/mL for glass	300	1,500	3,000	30,000	300	1,500	3,000	30,000
EPAL	14,622 Ci				13,395 Ci			
CUMREL at flow rate 1 L/year	230	394	598	4287	212	307	424	2547
Equivalent CUMREL from Glass Only	41	205	410	4099	24	118	236	2359
Percentage of Glass	18	52	68	96	11	38	56	93

- Flow rates span from 0.1 L/year to 20 L/year.
- Used reference activity per unit volume of leachate is 300 pCi/mL for SF and the values for glass in the Table were varied with respect to this reference value. As shown previously, these values of leachate activity can be increased significantly to exceed the EPA limits even at lower values of flow rates.

REFERENCE ACTIVITY IN LEACHATE (pCi/mL) [3,4]

	25° C	85° C
Pu-(239+240)	300	1
Am-241	300	0.1 – 1
Np-237	0.3	0.1
Cm-244	300	1

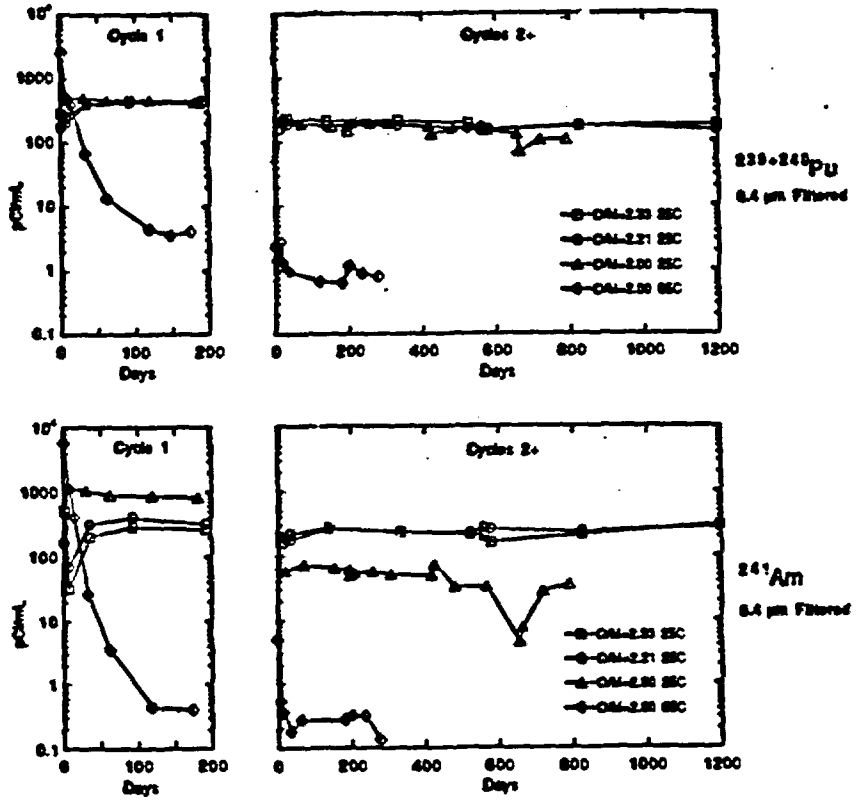
ISSUES ON PARTICULATE SEDIMENTATION

- The elemental release may be slowed down, but to the ratio of 1 L/year to 100 L/container ?
- S/V values are smaller in repositories without considering crackings.
- Tests under dripping groundwater conditions or under agitation conditions did not show the sedimentation.
- pH effects ?

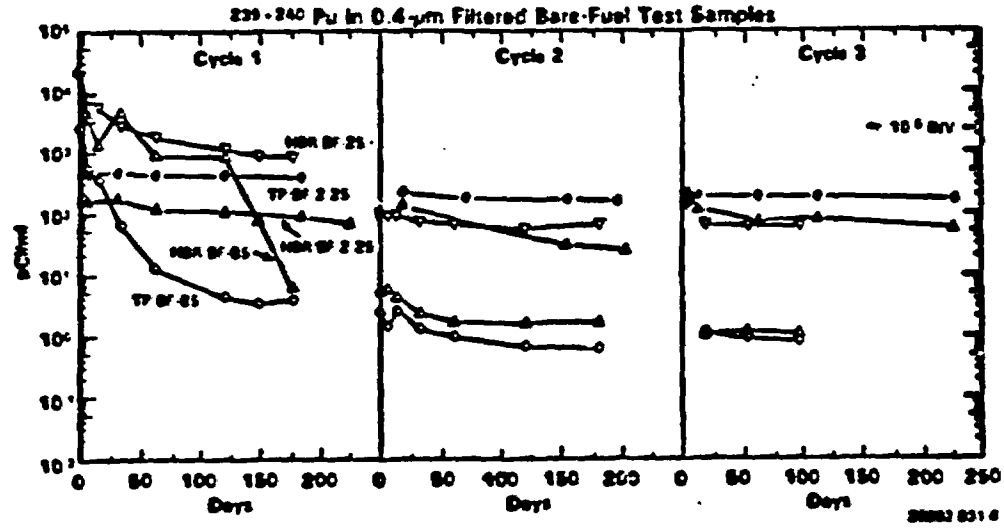
OTHER REASONS FOR THE IMPORTANCE OF HLW GLASS

- Urgent Task : Hot Operation in 1994.
- Alteration of Groundwater Chemistry to Affect SF Dissolution.
  - Si and Fe : Secondary Phase Formation.
  - B : Neutron Poison.
- Dose Rate Effects.

DATA I : SF [3,4]

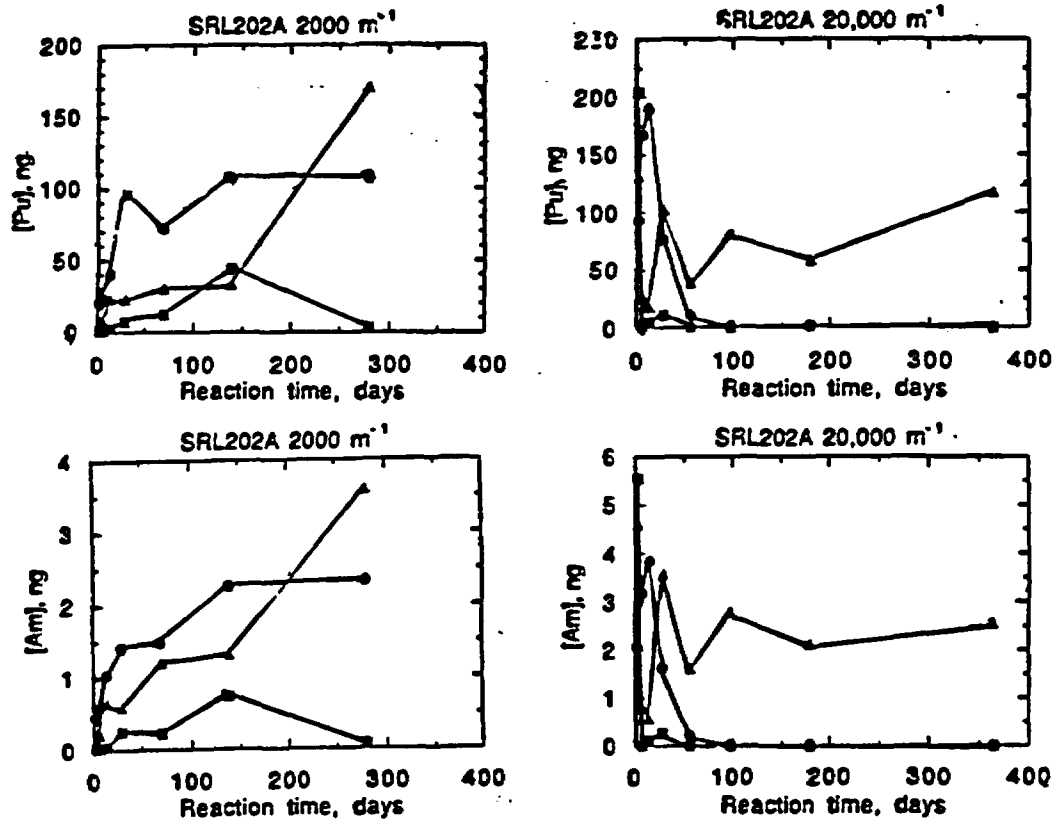


Uranium Concentrations and Activities of $^{239+240}Pu$ and ^{241}Am Measured in 0.4-μm Filtered Solution Samples. Mass basis conversion factors are 1 pCi $^{239+240}Pu$ = 1.1×10^{-5} μg Pu and 1 pCi ^{241}Am = 3.6×10^{-7} μg Am.



Comparison of $^{239+240}Pu$ Activities Measured in 0.4-μm Filtered Samples from the Three Series 3 and Two Series 2 Bare-Fuel Tests

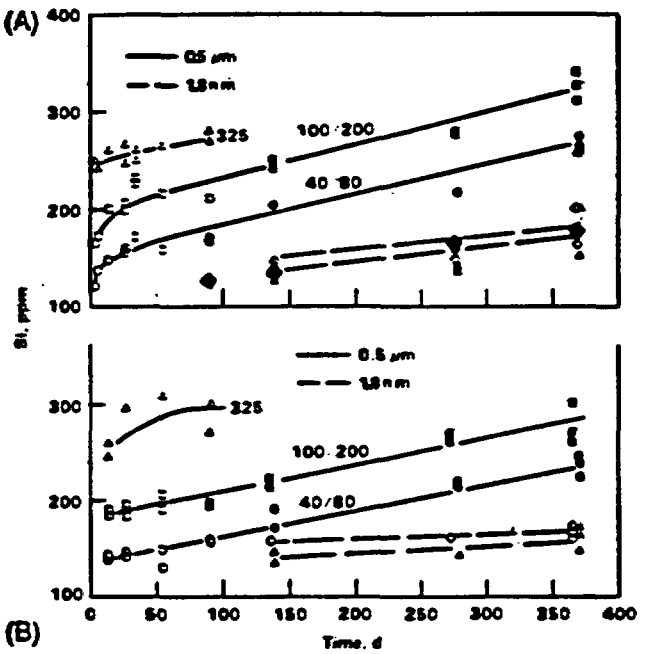
DATA II : RADIOACTIVE GLASS [1]



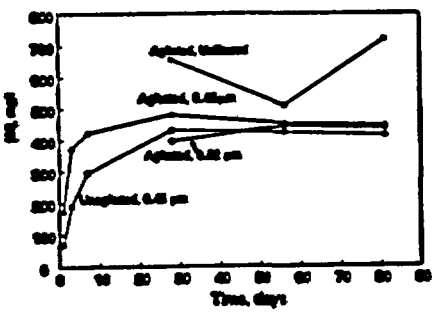
Mass actinide in (●) 0.45 μm filtrate, (■) 60 Å filtrate, and (▲) acid soak solution for tests with SRL 202A glass at 2000 m^{-1} for (a) Np, (b) Pu, and (c) Am; and at 20,000 m^{-1} for (d) Np, (e) Pu, and (f) Am.

DATA III : NONRADIOACTIVE GLASS [11,16]

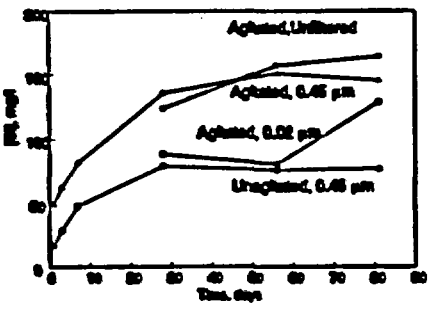
[16] I. S. Muller, A. C. Buechelle, I. L. Pegg and P. B. Macedo, Mat. Res. Soc. Symp. Proc., 257, 91 (1992)



Total Si concentrations in 76-68 glass leachates from 90°C tests: (A) deionized water, large solid diamonds are calculated silica concentrations; (B) silicate water. Solid symbols represent U-doped glass; open symbols represent non-U-doped glass.



Glass UTh116.



Glass UTh119.

NOTE TO R. WELLER

FROM T. AHN TIH

SUBJECT : SOURCE TERM OF LONG-LIVED RADIONUCLIDES

DATE : MARCH 17, 1993

The attached handouts are results from an analysis conducted for the release of long-lived radionuclides : U-234, Am-243, Cs-135, Se-79, I-129, Tc-99, C-14, Np-237 and Sn-126. From this work, it is found that the release of Tc-99 and C-14 under saturated conditions is a potential problem against their containment in natural barriers within EPA limits. This tentative conclusion is based on the assumption of the linear time-law in the growth of the altered layer of spent fuels under long-term steady-state conditions. The non-linear time-law in the growth of the altered layer may lead to the containment of C-14 and Tc-99 in natural barriers within EPA limits. Also the problem associated with Tc-99 is rather dependent on the retardation characteristics in natural barriers, because the inventory of Tc-99 is only slightly more than its EPA limit.

The annual release rates of C-14, I-129, Tc-99, Cs-135 and Se-79 may exceed the NRC criteria for the linear time-law in the growth of the altered layer, whereas those of Np-237 and Sn-126 may not (estimations from direct measurements); those of U-234 and Am-243 may not (estimation based on the congruent dissolution).

This tentative conclusion is based on our postulates of the continuous growth of the altered layer of spent fuels under long-term steady-state conditions. A consultation with PA staff will be made regarding the transport properties of these radionuclides for final conclusions.

The Attachment : As Stated.

CC : R. Ballard
: K. Chang, EBSPAC
: M. Lee, IPA

NOTICE : All handouts are for spent fuels, unless specified otherwise.

LONG-LIVED RADIONUCLIDES [1]

Species	Inventory (Ci/MTHM)	EPA Limits (Ci/MTHM)	Release Type
U-234	1.89 (2.04×10^{-2})	0.1	s
Am-243	1.54×10 (3.44×10^{-3})	0.1	s
Np-237	1.12 (5.30×10^{-3})	0.1	s
Cs-135	3.51×10^{-1} (5.92×10^{-2})	1.0	a
I-129	2.95×10^{-2}	0.1	a
Sn-126	7.15×10^{-1} (2.63×10^{-1})	1.0	s
Tc-99	1.23×10 (1.83)	10.0	a
Se-79	3.81×10^{-1} (1.01×10^{-1})	1.0	a
C-14	1.54(total) 0.58(matrix) 0.51(cladding) 0.45(hardware)	0.1	a

[1] : R. W. Barnard et al., An Initial Total-System Performance Assessment for Yucca Mountain, Sandia Report, SAND91-2795, 1992.

- Pu-(239+240) and Am-241 are assessed separately.
- a: alteration, s: solubility
- Parentheses are for DWPF glass.
- Inventories are for MTHM of SF or glass.
- 0.1 % criteria were not considered due to uncertainties associated with transport properties of Pu-(239+240) and Am-241. All species are assumed to be greater than 0.1 % of the total annual release rate limit.

Measured 10^{-5} of Inventory at 85° C [2]

Species	Cycle 1 (175 days)	Cycle 2 (180 days)	Cycle 3 (100 days)
I-129	107 ~ 306 (7.7)	25 ~ 29 (0.3)	13 ~ 17 (1.4)
Tc-99	12 ~ 89 (25.2)	39 ~ 57 (7.9)	19 ~ 34 (<8.1)
C-14	442 (235) (*1)	195 (193)	99 (120)

[2] : C. N. Wilson, Results from NNWSI Series 3 Spent Fuel Dissolution Tests, Pacific Northwest Laboratory Report, PNL-7170, 1990.

- Parentheses are for tests at room temperature.

*1 0.5 ~ 1.2 % of the inventory was released for ~ 220 days at room temperature in N2 tests [3].

COMPARISION OF FLOW-THROUGH TESTS AND IMMERSION TESTS

Temperature (° C)	Test Type	Dissolution Rate (cm/day)
85	N2 [3] N3 [2]	- ~ 10^{-6}
25	N2 N3	erratic to N3 ~ $3x10^{-9}$
25	Flow-Through [4]	~ $2x10^{-10}$

- In flow-through tests, transient effects may increase the dissolution rate slightly.

[3] : C. N. Wilson, Results from NNWSI Series 2 Bare Fuel Dissolution Tests, Pacific Northwest Laboratory Report, PNL-7169, 1990.

[4] : W. J. Gray, Dissolution Testing of Spent Fuel, NWTRB Full Board Meeting, 1992.

RELEASE OF Np-237 and Sn-126

	Np-237	Sn-126
CUREL (Ci)	6.3×10^{-2}	1.6×10^{-1}
EPA Limit (Ci)	1.2×10^{4}	1.2×10^{5}
Inventory (Ci)	1.3×10^{5}	8.8×10^{4}
Fractional Release (/year)	4.9×10^{-11}	1.8×10^{-10}

- Numbers of Containers : 62,850
- Total Inventory : 116,921 MTHM
- CUREL : Total Release at the Flow Rate of 1 L/year for 10,000 Years without Retardation.
- Increasing factors in room temperature tests : ~ 1 (Np-237) and ~ 0.5 (Sn-126).

- **ENHANCEMENT FACTORS USING RESULTS OF DRIPPING GROUNDWATER TESTS [5,6]**

  Np-237 : (70 ~ 130) times (REF : ~330 times for Glass).

  Sn-126 : (50 ~ 280) times.

[5] D. J. Wronkiewicz, J. K. Bates, T. J. Gerding and E. Veleckis, J. Nuclear Materials, 190, 107 (1992).
[6] J. K. Bates, J. P. Bradley, A. Teetsov, C. R. Bradley, and M. Buchholtz ten Brink, Science, 256, 649 (1992).

LEACHATE ACTIVITIES AT 85° C (pCi/mL)

	N3 [2]	D [5]
Np-237	~ 10^{-1}	(7 ~ 13) (*1)
Sn-126	~ 3×10^{-1}	(19 ~ 39)

N3 : Immersion Tests.
D : Dripping Groundwater Tests.

*1 : Glass : 33 [6]

U-234 AND Am-243 RELEASE FOR CONGRUENT DISSOLUTION

	U-234	Am-243
pCi/mL I-25 I-85 D-90	 5 ~ 27 0.8 54 ~ 108	 $3x(10^{-2} \sim 1)$ $9x(10^{-4} \sim 10^{-3})$ 438 ~ 877
CUREL (Ci) I-25 I-85 D-90	 3 ~ 17 0.5 68	 $2x(10^{-2} \sim 1)$ $6x(10^{-4} \sim 10^{-3})$ 276 ~ 551
EPA Limit (Ci)	$1.17x10^{4}$	$1.17x10^{4}$
Inventory (Ci)	$2.37x10^{5}$	$1.81x10^{6}$
Fractional Release (/year) I-25 I-85 D-90	 $(1 \sim 7)x10^{-9}$ $2x10^{-10}$ $3x10^{-9}$	 $10^{-12} \sim 10^{-10}$ $3x(10^{-14} \sim 10^{-13})$ $(2 \sim 3)x10^{-8}$

- I : Immersion Tests at 25° C and 85° C [2,7].
- D : Dripping Groundwater Tests at 90° C [5].
- Flow Rate : 1 L/year.

[7] C. N. Wilson, Mat. Res. Soc. Symp. Proc., 212, 197 (1991).

RELEASE OF RADIOACTIVITY IN THE ALTERATION CONTROL : LINEAR GROWTH

$$\left(\frac{dC_{container}}{dt}\right) = k - \frac{F}{V} C_{container}$$

$C_{container}$: The Amount of Radioactivities Retained.
t : Time.
k : Dissolution Rate Constant.
F : Flow Rate.
V : Leachate Volume.
$C_{released}$: The Amount of Radioactivities Released.

$$C_{container} = k \frac{V}{F} \left(1 - e^{-\frac{F}{V} t}\right)$$

$$C_{released} = k\, t - k \frac{V}{F} \left(1 - e^{-\frac{F}{V} t}\right)$$

[Tc-99 Case]

$C_{released}$: 7.2×10^6 Ci (TP, 1 L/year)
1.6×10^7 Ci (HBR, 1 L/year)
4.8×10^6 Ci (TP, 0.1 L/year)
1.1×10^7 Ci (HBR, 0.1 L/year)

Inventory : 1.4×10^6 Ci

- 62,850 containers from 116,921 MTHM.
  V : 4×10^5 cm^3.
  TP : Turkey Point Fuel, HBR : Robinson Fuel.

Glass Contribution :

Glass : 1.83 Ci/MTHM with 29,297 MTHM
SF : 12.3 Ci/MTHM with 116,921 MTHM

Averaged Value : 10.2 Ci/MTHM for DWPF Glass

[Cs-135 and Se-79 Cases] : 0.0285 (Cs-135) and 0.0310 (Se-79) of $C_{released}$ in Tc-99.

RELEASE OF RADIOACTIVITY IN THE ALTERATION CONTROL : PARABOLIC GROWTH

$$\frac{dC_{container}}{dt} = k\,(t+a)^{-\frac{1}{2}} - \frac{F}{V}\,C_{container}$$

$$C_{released} = 2\,k\,(t+a)^{\frac{1}{2}} - \left[e^{-\frac{F}{V}t}\int k\,(t+a)^{-\frac{1}{2}}\,e^{\frac{F}{V}t}\,dt\right]_0^t$$

- Alteration from Data of Flow-Through Tests Adjusted with Immersion Tests Data (Parentheses for no adjustment)
  - (1) For 0.1 cm, 5×10^{12} ($\sim10^{15}$) years.
  - (2) For 10,000 years, 3×10^{-6} (3×10^{-7}) cm.

    (1) and (2) are from a regression with two data points [4].
  - (3) From the first term of second equation : 7×10^{-3} of the value in linear-growth case.

RELEASE OF Cs-135, Se-79 AND Tc-99 IN DRIPPING GROUNDWATER

	Cs-135	Se-79	Tc-99
pCi/mL	10 ~ 20	11 ~ 22	353 ~ 705
CUREL (Ci)	6 ~ 13	7 ~ 14	222 ~ 443
EPA Limit (Ci)	1.17×10^{5}	1.17×10^{5}	1.17×10^{6}
Inventory (Ci)	4.10×10^{4}	4.45×10^{4}	1.44×10^{6}
Fractional Release (/year)	$(2 \sim 3) \times 10^{-8}$	$(2 \sim 3) \times 10^{-8}$	$(2 \sim 3) \times 10^{-8}$

MODELS FOR STEADY STATE DISSOLUTION OF SPENT FUEL

- Supporting Data :

  (1) Consistency of Immersion Tests and Flow-Through Tests [2,4].

  (2) Analogue Studies : Massive Alteration of Uraninite to Uranyl Silicate Phases at Pena Blanca Site (>10^6 years [8]).

  (3) Linear Growth Rate of the Altered Layer in Glass [9].

  (4) Offset of the Altered Layer in Dripping Groundwater Tests [5].

  - Uncertainties :

    - (a) Nonlinear Growth Rate (B Release Rate) of the Altered Layer [10,11].

    - (b) Effective Surface Area for Dissolution or Transient Effects.

    - (c) Role of Solid State Diffusion.

    - (d) Fe reduces dissolution rates by Eh change, whereas Fe and Si increase the growth rates of the altered layer.

    - (e) Other Effects : Radiation, Slough-off, Cyclic Behavior and etc. [HANDOUT, 8.13.92].

[8] B. W. Leslie, The Pena Blanca Natural Analog : Constraints on Contaminant Transport, NRC Research Geochemistry Program Review, November 3, 1992

[9] Nuclear Technology Programs Semiannual Progress Report, April-September 1990, Argonne National Laboratory Report, ANL-92/25, 1992.

[10] Final Report of the Defense High-Level Waste Leaching Mechanisms Program, Pacific Northwest Laboratory Report, PNL-5157, 1984.

[11] P. B. Macedo and A. C. Buechele, Mat. Res. Soc. Symp. Proc., Fall, 1992 (in press).

- Description:

  (1) NOTE TO R. WELLER, 2.19.1993.
  (2) HANDOUT for HLWM REVIEW, 10.29.92.
  (3) HANDOUT for HLWM REVIEW, 8.13.92.
  (3) Manuscript of Spent Fuel for IPA-2 (10.2.92) under revision.

SOLUBILITY CONTROLLED RELEASE OF RADIONUCLIDES IN GLASS

Inventory	U-234	Am-243	Np-237	Sn-126	Am-241
SF (10^5 Ci)	2.21	18.0	1.31	8.36	1,000
Glass (Ci)	598	101	155	7,705	192,080
Ci/MTHM (10^{-3})	20.4	3.44	5.30	263	6,560
Ratio 1	3.86	0.65	1	49.7	1,239
Ratio 2 (10^{-4})	31.1	5.26	8.07	401	10,000

- SF Inventory from NOTE 2.19.93 and Glass Inventory from NOTE 2.19.93 and [12].

[12] DWPF Orientation and Tour, August 7-8, The Department of Energy, 1991.

Leachate Activities (pCi/mL)

Np-237 D Test [6]	Am-241 D Test [6]	Np-237 I Test [13]	Am-241 I Test [13]
33	(0.8 ~ 8)x10^5	35	640

[13] W. L. Ebert and J. K. Bates, Proceeding of the Third International High-Level Radioactive Waste Management Conference, pp.934, Las Vegas, NV, April 12-16, 1992.

- Release Rate per Year (Ci/year) at 1 L/year of Flow Rate :

  = [ ] (pCi/mL) x 1.4x10^{-5}