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LEACH AND SOIL COLUMN TESTS
WITH STORED REDOX SLUDGE

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LEACH AND SOIL COLUMN TESTS
WITH STORED REDOX SLUDGE

INTRODUCTION

Radioactive solids (sludge) precipitated from neutralized Redox process high-level waste are currently stored in 26 tanks at Hanford. Alkaline supernatant liquid is present in many of these tanks. As part of the overall Hanford Waste Management program, the supernatant liquid will be pumped from these tanks and processed in the B Plant to remove ^{137}Cs . The radioactive sludge in the bottom of the tanks will not be removed, however, but will be stored there for an indefinite time.

Detailed knowledge of the physical and chemical properties of the stored Redox sludge is essential to specify proper conditions for its prolonged safe storage in the underground tanks. A laboratory program to provide the necessary data has been under way for some time. Studies to identify and measure the decomposition products of the Redox sludge stored in Tank 108-SX have recently been completed.⁽¹⁾ Experiments to determine the thermal conductivity of the sludge in Tank 108-SX are in progress.⁽²⁾

Studies to characterize the behavior of typical Redox sludge solids upon exhaustive water leaching are also being performed as part of the laboratory effort. Preliminary results of such leach tests with solids from Tank 108-SX are presented in this report. Soil column tests (with representative Tank Farm soil) were also performed to establish uptake of various radionuclides from the leach liquor. To some extent at least, these leach-soil column tests simulate behavior to be expected if large amounts of water were inadvertently

introduced into the underground tanks and then discharged to the surrounding soil.

SUMMARY AND CONCLUSIONS

The leachabilities of ^{90}Sr , ^{239}Pu , and ^{137}Cs from solids in Tank 108-SX were determined. Solids from this tank were vigorously mixed with 3 volumes of water or 0.1M NaNO_3 at 25 C and the leachate replaced every four hours with fresh water or dilute sodium nitrate solution. Through 21 leach cycles covering a 3.5 day period, 63 volumes of leachate removed 1.4, 4.6, and 19.2 percent of the ^{239}Pu , ^{90}Sr , and ^{137}Cs , respectively, from the 108-SX solids. With the exception of the first few leachates, the ^{239}Pu concentrations in the leachates were less than MPC_w^* .

A composite of the TK-108-SX leachate was passed through a column containing typical Hanford soil. Column distribution coefficients with 108-SX leachate of 14, 24, and 20 were obtained for ^{239}Pu , ^{90}Sr , and ^{137}Cs , respectively, indicating appreciable sorption of all these radionuclides by the soil. The radionuclides were not readily removed from the loaded soil columns with 12 column volumes of dilute leachate solution.

Laboratory results with the 108-SX sludge were carefully evaluated to determine their significance to the overall analysis of the safety of long-term storage of Redox sludge in existing underground tanks. The *primary* deterrent to movement of the radionuclides from such solids to the water table, which would be located from 215 to 230 feet below the tanks following termination of low and intermediate level liquid waste disposal,

* MPC_w = maximum permissible concentration in drinking water for 168-hour per week exposure.

is the absence of moving water in the storage zone. Incident precipitation does not migrate to the storage depth. The leaching characteristics of the solids and the ion exchange properties of the soil are *secondary* factors in preventing dispersion of the radionuclides to the environment. Based upon leaching and soil column results with 108-SX sludge, very large volumes of water would be required to move small amounts of radionuclides from the Redox sludge in the tanks to the deep water table. For instance, percolation of 250 feet of water through the solids in the 75-foot diameter 108-SX tank to the water table would transport less than 0.01 percent of the ^{239}Pu , ^{90}Sr , and ^{137}Cs in the solids into the water table. The maximum concentrations of ^{239}Pu and ^{137}Cs in the liquid entering the water table would be less than their respective MPC_w values while the maximum concentration of ^{90}Sr would be about 2000 times its MPC_w . A major flood or continued percolation downward of *all* incident precipitation for 500 years (based upon the present annual precipitation rate of 0.5 foot/year) would be required to obtain this volume of water.

EXPERIMENTAL - RESULTS

SOLIDS SOURCE AND COMPOSITION

Tank 108-SX was filled with neutralized Redox Process high-level waste in January, 1959. In March, 1967, the tank was found to be leaking and the bulk of the supernatant liquid was pumped to another tank. The sludge together with some supernatant liquid was left in the tank. Since March, 1967, air has been blown continuously across the surface of the sludge to dissipate radioactive heat; the maximum temperature within the salt cake is about 90 C.⁽³⁾

The Redox sludge used in these studies was an air-dried composite of several samples taken in 1968 from the top layer of the salt cake in Tank 108-SX. Table I lists the concentration

of plutonium, ^{90}Sr , and gamma-emitting radionuclides in our sample of Tank 108-SX sludge. [Leach conditions and data used to establish the composition of the Redox sludge are listed in the Appendix.] This material contained about 6.5 wt percent carbonate and its density as determined by displacement with Normal Paraffin Hydrocarbon (NPH) was 1.58 g/ml.

TABLE I
RADIONUCLIDE CONTENT OF SOLIDS
FROM TANK 108-SX

<u>Radionuclide</u> ^(b)	<u>Concentration, $\mu\text{Ci/g Solid}$</u> ^(a)
^{137}Cs	670
^{134}Cs	12
$^{106}\text{Ru}^{106}\text{Rh}$	470
$^{144}\text{Ce}^{144}\text{Pr}$	420
$^{152-154}\text{Eu}$	16
^{90}Sr	2600
Plutonium ^(c)	0.035

a Calculated from data in Appendix.

b Concentration of other radionuclides not measured.

c Milligrams Pu (as ^{239}Pu)/g solid.

LEACH STUDIES

Results of numerous leaches of a portion of Tank 108-SX sludge with water and 0.1M NaNO_3 are listed in Table II. To obtain these data, 5.49 grams of solids were leached at 25 C with successive 11-ml portions of either water (first 7 leaches) or 0.1M NaNO_3 (last 14 leaches). Solids and liquid (contained in a 40-ml centrifuge cone) were vigorously mixed by an overhead glass stirrer throughout each 4-hr leach period.

Twenty-one successive leaches of the 108-SX sludge removed 1.4, 19.2, and 4.6 percent of the Pu, ^{137}Cs , and ^{90}Sr , respectively. Of this amount of plutonium, about 93 percent was removed in the first five water leaches; the other 16

TABLE II
WATER LEACHING OF TANK 108-SX SLUDGE

Leach No.	Vol Leach ml	Vol Solids ml	Concentration in Leach			% in Leach ^a		
			¹³⁷ Cs μCi/ml	⁹⁰ Sr μCi/ml	Pu ^c d/m/ml	⁹⁰ Sr ^b	¹³⁷ Cs	Pu
1	6.5	7	d	d	23,400	d	d	0.58
2	10	7	d	d	8,200	d	d	0.31
3	10	7	d	d	4,400	d	d	0.17
4	10	8	d	d	2,440	d	d	0.095
5	9	9	d	d	3,100	d	d	0.11
6	6.5	9.5	d	d	1,620	d	d	0.040
7	5	12	d	d	464	d	d	0.0088
8	5	12	d	d	464	d	d	0.0088
9	8.5	12.5	d	d	<160	d	d	-
10	9	11	d	d	320	d	d	0.011
11	9	11	d	d	<160	d	d	-
12	10	11	d	d	<160	d	d	-
13	8	13	0.48	5.95	248	0.34	0.10	0.0075
14	9	12	0.29	5.24	320	0.34	0.07	0.011
15	10	11.5	0.20	5.26	<160	0.34	0.05	-
16	10	11	0.13	5.19	<160	0.37	0.036	-
17	8.5	11.5	0.11	6.06	<160	0.30	0.036	-
18	10.5	11.5	0.078	4.43	<160	0.34	0.022	-
19	10.5	12	0.058	4.04	<160	0.28	0.16	-
20	10	12	0.046	3.48	216	0.25	0.013	0.0082
21	9.5	11.5	0.041	3.57	<160	0.23	0.011	-
TOTAL						4.55	19.2	1.35

^a Percent of total radionuclide in 5.49 g sludge.

^b Composite of first 12 leaches contained 1.8 percent of ⁹⁰Sr and 18.8 percent of ¹³⁷Cs in the 5.49 g of sludge.

^c Lower limit detection was 160 d/m/ml.

^d Not determined.

leaches removed only minor amounts of plutonium. Similarly, of the ^{137}Cs removed, 98 percent was contained in the first 12 leaches; the last 9 leaches contained only small concentrations of cesium. The leach behavior of strontium in the 108-SX sludge was different from that of cesium and plutonium in that each leach appeared to remove a constant amount of ^{90}Sr .

Residues resulting from the sixth and, particularly, the seventh water leaches peptized severely, making it difficult to separate solid from liquid by centrifugation. Peptization was avoided in succeeding leaches by using 0.1M NaNO_3 as the leachant. The volume of the leached Tank 108-SX solids remained constant at about 12 ml over the last 14 leaches.

SOIL COLUMN TESTS

Table III gives conditions and results of soil column tests with the leach liquors obtained from exhaustive leaching of Tank 108-SX sludge. Data for the loading step are correlated in Figure 1, where C/C_0 (concentration activity in effluent/concentration activity in feed) is plotted on log probability paper as a function of the number of bed volumes loaded. As expected from theoretical considerations,⁽⁴⁾ such plots are straight lines. Approximately 17 to 19 column volumes were loaded before 50 percent breakthrough of either ^{137}Cs or ^{90}Sr ; but 50 percent breakthrough of plutonium occurred after passage of about 10 column volumes.

Passage of 12 column volumes of the wash liquor eluted about 5.5 percent of the ^{137}Cs on the loaded soil column. The concentration of plutonium in both the influent and effluent wash solutions was below the detection limit of 160 d/m/ml indicating that little, if any, plutonium was washed from the loaded soil column. Strontium, rather than being desorbed, actually was taken up from the wash liquor.

TABLE III
SOIL COLUMN TESTS WITH WATER LEACH
OF TANK 108-SX SLUDGE

I. CONDITIONS

1. Leach: As listed in Table II.
2. Load: Composite of first 12 leaches (Table II) passed downflow at 0.6 ml/min through a 4 cm³ bed (1 cm diam x 5 cm high) of soil from the 241-SX Tank Farm area.
3. Wash: Composite of last 9 leaches (Table II) passed downflow at 0.6 ml/min through loaded bed.

II. RESULTS

A. Load^a

Bed Volumes	Effluent			C/C ₀ ^b		
	¹³⁷ Cs μCi/ml	⁹⁰ Sr μCi/ml	Pu d/m/ml	¹³⁷ Cs	⁹⁰ Sr	Pu
3	-	0.0045	192		0.0178	0.0571
6	0.0044	0.030	1100	0.0014	0.119	0.327
9	1.13	0.061	1730	0.0362	0.241	0.515
12	3.72	0.084	2000	0.119	0.443	0.604
15	6.59	1.01	2130	0.211	0.398'	0.634
18	3.30	1.17	2340	0.106	0.461	0.696

^a Feed: 3360 d/m/ml Pu, 254 μCi/ml ⁹⁰Sr, 31.2 μCi/ml ¹³⁷Cs.

^b C/C₀: Concentration in Effluent/Concentration in Feed

B. Wash^a

Bed Volumes	¹³⁷ Cs μCi/ml	⁹⁰ Sr μCi/ml	Pu d/m/ml	¹³⁷ Cs % Washed from Bed
3	3.31	0.035	<160	1.83
6	2.24	0.069	<160	1.21
9	2.32	1.00	<160	1.25
12	2.35	1.43	<160	1.27

^a Wash: 160 d/m/ml Pu, 4.56 μCi/ml ⁹⁰Sr, 0.16 μCi/ml ¹³⁷Cs.

FIGURE 1

SOIL COLUMN TEST WITH WATER LEACH OF TK 108-SX
SLUDGE - CORRELATION OF LOADING DATA

ANALYTICAL PROCEDURES

The concentrations of gamma emitters in the various solutions generated in this study were determined by conventional (NaI) gamma energy pulse height analyses. A procedure by Kiba and Mizukami was used to analyze for ^{90}Sr in these solutions.⁽⁵⁾

Plutonium was determined by triiso-octylamine and thenoyl-trifluoroacetone extractions and alpha counting.⁽⁶⁾ This procedure would not detect less than about 160 d/m/ml plutonium in the solutions analyzed in this work.

Conventional techniques were used to determine carbonate in solid samples of the sludges.

DISCUSSION

LEACHABILITY OF SOLIDS

Twelve leaches of the 108-SX solids removed 19 percent of the cesium, 1.4 percent of the plutonium, and 1.8 percent of the strontium. High leachability of cesium was expected since the solids are composed principally of highly-soluble sodium salts. It is interesting to note, however, that the leachability of cesium, and also of strontium, from the Tank 108-SX solids are lower than similar values (Table IV) with calcine produced from fluidized bed calcination of acidic aluminum wastes at the Waste Calcining Facility.

The technique used by Paige⁽⁷⁾ for determining calcine leach rates shown in Table IV was considerably less severe than that used in this study. He continuously circulated water through a bed of calcine particles rather than violently agitating the water and solids to hasten the leach rate as was done in this study.

TABLE IV
COMPARISON OF LEACHABILITIES OF
REDOX SLUDGE AND CALCINE
[% Leached in Initial 3.5 Days]

<u>Radionuclide</u>	<u>Redox Sludge Tank 108-SX</u>	<u>Calcine</u> ⁽⁷⁾
⁹⁰ Sr	4.6	25
¹³⁷ Cs	19.	80
²³⁹ Pu	1.4	-

SOIL COLUMN TESTS

Absorption Behavior

Sorption of plutonium from the Tank 108-SX leachate by the soil yielded a typical breakthrough curve showing an increase in plutonium concentration in the effluent with increased throughput. Assuming the breakthrough curve to be linear on a log probability plot to 100 percent breakthrough, a plutonium column distribution ratio (τ'') of 14 was calculated using ion exchange principles, where the column distribution ratio is defined by Helfferich⁽⁸⁾ as follows:

$$\tau'' = K_d e_b \quad \text{and}$$

K_d = equilibrium distribution coefficient

$$= \frac{\text{amount Pu associated with soil per gram of soil}}{\text{amount Pu in the equilibrium solution per ml of solution}}$$

e_b = bulk density of the soil, g/ml

By comparison, Hajek and Knoll⁽⁹⁾ obtained a plutonium K_d of 1659 ($\tau'' \sim 2600$) when alkaline solutions resulting from water leaches of sludges formed from neutralized Z Plant high-salt waste were contacted with soil in batch equilibration studies. Carbonate present in the 108-SX solids was probably removed by the leachates and hindered plutonium sorption by the soil.

The behavior of strontium in adsorption studies with Tank 108-SX leachate is not unusual. A column distribution coefficient of 24 was calculated. Hajek⁽¹⁰⁾ obtained a column distribution coefficient of 73 for river water traced with strontium. Knoll⁽¹¹⁾ determined the strontium capacity of soil to be from 4×10^{-4} to 5.2×10^{-4} meq/100 grams soil with a solution containing 1000 ppm calcium and 0.1 ppm strontium. Our results indicate a strontium capacity of about 6×10^{-4} meq/100 grams soil, which is in reasonable agreement with Knoll's results even though the two systems are somewhat dissimilar.

Adsorption of cesium by the soil from Tank 108-SX leachate was as expected based upon other work. We obtained a column distribution ratio of 20, while Hajek⁽¹⁰⁾ determined a value of 712 for river water traced with cesium. Routson⁽¹²⁾ has estimated a cesium K_d of 0.7 ($\tau \sim 1.2$) for a high-salt waste system ($8M Na^+$). The distribution ratio for Tank 108-SX appears reasonable in light of these data since the salt content of the leachate would be intermediate to the salt contents of solutions used by Hajek and Routson.

Desorption Behavior

Following the sorption step, a composite of the last nine leaches of the 108-SX solids was passed through the column. In this step, only cesium desorbed from the loaded soil column; strontium, instead of being desorbed, actually continued to load onto the soil. The plutonium concentration in the effluent solution from the desorption cycle was below the analytical detection limit. Strong affinity of the soil for sorbed plutonium is inferred from the latter observation.

SIGNIFICANCE OF LEACHING AND SOIL COLUMN RESULTS TO SAFETY OF LONG-TERM STORAGE OF REDOX SLUDGES

The leaching and soil column data indicate that water will

leach radionuclides from Redox sludge and that appreciable sorption of these radionuclides from the leachate onto Hanford soil can be expected. These data need to be analyzed and applied to field conditions to assess their significance for safe, long-term storage of Redox sludges in existing tanks.

The *primary* deterrent to the movement of radionuclides from the sludge through the soil column beneath the storage tanks to the water table is the absence of flowing water in the storage zone. Previous field experience and results of studies now under way show that incident precipitation (rainfall) does not migrate to the depth where the Redox sludges are stored. The leaching characteristics of the sludges and the ion exchange properties of the soil for sorbing and retaining radionuclides are *secondary* factors in preventing dispersion of radionuclides to the environment. It is of interest to scale up the laboratory leaching and soil column tests to determine the additional safety afforded by the secondary factors.

* Consider the relative volume of solution (water or 0.1M NaNO_3) to solids which was used in the laboratory leaching tests. Through 21 leaches of Tank 108-SX solids, 73 volumes of leach solution were used per volume of solids to remove 1.4 percent, 19.2 percent, and 4.6 percent of the plutonium, cesium, and strontium, respectively. Tank 108-SX contains about 1.3×10^5 gallons of solids so that nearly 8.2×10^6 gallons of water would be required to leach these percentages of radionuclides from the solids in the tank assuming *vigorously agitated* contact conditions such as used in the laboratory. Of course, vigorous agitation is inconceivable as liquid would merely percolate through the solids. Percolation of 8.2×10^6 gallons through the solids would require nearly 250 feet of rain water to pass through the 75-foot diameter tank assuming no loss of rain water by evapotranspiration. At the current average incident precipitation rate of 0.5 ft/year,

500 years would be required to obtain this volume of water. Since present climatic conditions preclude migration of incident precipitation to the storage depth, a virtually infinite time would be required to leach radionuclides from the solids.

In scaling up the 108-SX soil column test results, we postulate a simple model using the following assumptions:

1. Water (8.2×10^6 gallons) from a local flood covering the tank percolates through the solids in the tank and exits from the tank uniformly through cracks in the bottom. The leachate leaving the tank contains ^{239}Pu , ^{137}Cs , and ^{90}Sr concentrations of 3.2 , 1.4×10^4 , and 1.3×10^4 $\mu\text{Ci/gallon}$, respectively. These values were obtained by compositing all the leachates given in Table II. The sample of solids from the 108-SX tank used in the leaching tests is assumed to be representative of all the solids in the tank.
2. The leachate flows under nearly saturated conditions to the water table, located 215 feet below the bottom of Tank 108-SX, through a soil column formed by projecting the area of the tank bottom to the water table. [The water table depth is based upon post-Hanford conditions; *i.e.*, after disposal of liquid wastes to swamps and cribs is terminated.] The leachate entering the water table does not appreciably raise the water table level. The flow rate of the leachate is comparable to that used in the laboratory tests (superficial velocity of 1.5 ft/hr).
3. The soil in the soil column is homogeneous and has the same properties as the soil used in the laboratory tests.
4. The specific retention capability of the soil is

ignored so that all the leachate is assumed to enter the water table.

For this simplified model, the results shown in Figure 2, page 15, would be obtained. Note that 92 percent of the ^{239}Pu , 98 percent of the ^{90}Sr , and >99.9 percent of the ^{137}Cs in the leachate would be adsorbed on the soil within the 50 feet of soil column immediately beneath the tank after 8.2×10^6 gallons of leachate had passed through the column. Less than 0.1 percent of the radionuclides in the leachate would enter the water table, which corresponds to less than 0.01 percent of the radionuclides in the solids. The plutonium and cesium concentrations in the last portion of liquid entering the water table would be less than their respective MPC_w levels and the strontium concentration would be a factor of about 2,000 above its MPC_w . Thus, even for the very large quantity of leachate assumed in this analysis, only a small amount of the radionuclides leached from the solids would enter the water table.

We can extend the analysis further and assume that additional water is continuously added to the tank to cause the radionuclides adsorbed on the soil to move toward the water table. If we assume column distribution coefficient determined during the loading cycle of 20, 24, and 14 for ^{137}Cs , ^{90}Sr , and ^{239}Pu , respectively, the velocities of the radionuclides relative to the liquid velocity would be approximately 1/70, 1/80, and 1/50 for the ^{137}Cs , ^{90}Sr , and ^{239}Pu , respectively. Many millions of gallons of water would be required to move the radionuclides down the soil column to the water table. As the salt content of the water decreased with the continued leaching of the sludge, the column distribution coefficients for the radionuclides would increase, resulting in even slower radionuclide migration rates.

Once the radionuclides reached the water table, they would migrate toward the Columbia River. The present travel time for

MAXIMUM CONCENTRATION IN LIQUID
ENTERING WATER TABLE:

$$\begin{aligned}
 {}^{239}\text{Pu} &= 5 \times 10^{-6} \text{ } \mu\text{Ci/ml} \text{ (0.1 MPC}_w\text{)} \\
 {}^{90}\text{Sr} &= 2 \times 10^{-3} \text{ } \mu\text{Ci/ml} \text{ (2X10}^3\text{ MPC}_w\text{)} \\
 {}^{137}\text{Cs} &= < 2 \times 10^{-4} \text{ } \mu\text{Ci/ml} \text{ (<MPC}_w\text{)}
 \end{aligned}$$

*PERCENT OF THE TOTAL AMOUNT LEACHED FROM THE SOLIDS

FIGURE 2

DISTRIBUTION OF RADIONUCLIDES BENEATH TANK 108-SX
FROM HYPOTHETICAL FLOODING INCIDENT

water to flow from beneath the 108-SX tank to the Columbia River is more than 1500 years.⁽¹³⁾ This travel time would increase considerably after low and intermediate level liquid waste disposal is stopped. The travel times for the radionuclides would be many times longer due to ion exchange with the soil along the flow path. Radioactive decay and dilution would cause concentrations of all the radionuclides to be reduced to innocuous levels before reaching the river.

Many of the assumptions used in this simplified analysis are highly conservative so that the quantities of radionuclides reaching the water table as a function of volume of liquid passing through the sludge are biased high. For example, percolation of liquid through the sludge would not remove radionuclides as readily as vigorous agitation of the liquid-solids mixture. Percolation of the liquid for a very long time (several years) would probably be required to equal the quantity of radionuclides removed by 3.5 days of vigorous agitation. Assumption two requires that the liquid flow rate be 1.5 ft/hr. This rate is nearly 100 times higher than that observed for water percolating to the water table below swamps. A flow rate of 0.015 ft/hr would probably sharpen the breakthrough curve shown in Figure 1; resulting in a lower quantity of radionuclides reaching the water table for the same volume of liquid.

A previous safety analysis⁽¹⁴⁾ has shown that the potential for local flooding, such as from a flash flood, which would provide 250 feet of water for percolation through the sludge, is extremely remote. There has been no evidence of previous flash floods during the last 10,000 years in the 200 Area plateau where the wastes are buried. There is no mechanism capable of creating a major river flood of sufficient magnitude to inundate the waste storage sites. Only massive floods such as that which occurred when Lake Missoula released its water during the glacial age some 50,000 to 10,000 years ago could inundate

these sites.

In summary, very large volumes of water would be required to move small amounts of ^{90}Sr , ^{137}Cs , and ^{239}Pu from the Redox sludge in Tank 108-SX to the water table. If the source of the water were incident precipitation at present rates, hundreds of years would be required to achieve the necessary volumes of water even if there were no evapotranspiration. Since incident precipitation does not migrate to the storage depth and flooding is extremely remote, the probability of leaching radionuclides from the 108-SX sludge is very low.

There is every reason to believe that Tank 108-SX sludge is typical of other stored Redox process sludges. Hence, conclusions pertaining to safety of long-term storage of sludge in Tank 108-SX apply equally well to storage of Redox sludge in other tanks.

RECOMMENDATIONS FOR FUTURE WORK

Two or three continuous core samples should be taken of the Redox sludge stored in Tank 108-SX and in various other tanks to determine variations in solids composition and leaching behavior as a function of location and depth. The leaching procedures should be modified to simulate field conditions more closely. Detailed composition of the leachates should be determined so that results of subsequent soil column tests can be compared and the effects of leachate composition on radionuclide movement in the soil can be defined.

Leaching and soil column tests should be performed with core samples taken from tanks filled with solids in the In-Tank Solidification Program. Some preliminary efforts in this direction with solids from Tank 116-TX have been started.

Methods for determining the permeability and porosity of the solids should be defined and implemented. Studies should be continued to determine the effects of temperature gradients

on water movement and to develop methods for reducing the leachabilities of the radionuclides from the solids by either reducing solubilities or permeabilities. Chemical and radiation stabilities of the solids and associated decomposition products should be defined.

Other studies aimed at determining the effects of increased rainfall, increased irrigation, and local and widespread flooding upon water table elevations and migration of the radionuclides from the solids to the water table should be continued. Data from these studies are essential to perform rigorous safety analyses of postulated incidents such as flooding.

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REFERENCES

1. G. F. Schiefelbein. *Thermal Characteristics of Tank 108-SX Sludge*, BNWL-CC-2042. April, 1969.
2. D. G. Bouse. Personal Communication, Pacific Northwest Laboratory, Richland, Washington. (1969)
3. W. L. Godfrey. Personal Communication. Atlantic Richfield Hanford Company, Richland, Washington. (1969)
4. *Waste Management Technical Manual*, J. S. Buckingham, Ed., Chapter 4 *Process Chemistry and Engineering*, p. 4206, ISO-100. Isochem Inc, Richland, Washington. August, 1967.
5. T. Kiba and S. Mizukami. "Radio-Separation of Radioactive Strontium by Solvent Extraction with TTA-Hexone," *Bull. Chem. Soc. Japan*, vol. 31, pp. 1007-1013. (1958)

6. *Analytical Technical Manual*, edited by R. A. Schneider and K. M. Harmon, HW-53368, General Electric Company, Richland, Washington. (1961)
7. B. E. Paige. *Leachability of Alumina Calcine Produced in the Idaho Waste Calcining Facility*, IN-1011. July, 1966.
8. F. Helfferich. *Ion Exchange*, McGraw-Hill Book Co., Inc. (1962)
9. B. F. Hajek and K. C. Knoll. *Disposal Characteristics of Plutonium and Americium in a High Salt Acid Waste*, BNWL-CC-649. June 10, 1966.
10. B. F. Hajek. *Adsorption, Migration, and Dispersion of Strontium and Cesium in an N-Area Soil*, BNWL-CC-208. July 7, 1965.
11. K. C. Knoll. *Adsorption of Strontium by Soils under Saturated and Unsaturated Flow Conditions*, HW-67830. General Electric Company, Richland, Washington. December 1, 1960.
12. Letter, R. C. Routson to G. L. Ritter. *Estimated Removal of Sr and Cs from High Salt Alkaline Wastes by Sorption on Soil*. March 4, 1970.
13. D. B. Cearlock. *Personal Communication*, Pacific Northwest Laboratory, Richland, Washington. March, 1970.
14. C. M. Unruh. *A Preliminary Safety Analysis of Near Surface Storage of Radioactive Waste as Salt Cakes*, BNWL-1194. January, 1970.

APPENDIX

TABLE A-1

LEACH STUDIES TO DETERMINE COMPOSITION OF TANK 108-SX SLUDGE

Conditions: 4.15 g of Tank 108-SX sludge leached (at 80 C) successively with 10-ml portions of solutions indicated.

No.	Composition	Final Vol, ml	Radioisotope Concentration					
			$\mu\text{Ci/liter (x } 10^{-4})$					$\mu\text{g/ml (x } 10^4)$
			^{137}Cs	$^{106}\text{Ru}-^{106}\text{Rh}$	$^{144}\text{Ce}-^{144}\text{Pr}$	$^{152-154}\text{Eu}$	^{90}Sr	Pu
1	H ₂ O	6.	15.2	a	a	a	0.32	1.19
2	H ₂ O	7.	7.09	a	a	a	0.13	0.15
3	6M HCl	9.5	4.81	a	a	a	95.4	51.0
4	5M HCl	9.5	1.37	3.72	a	a	10.1	22.2
5	11M HNO ₃ -0.1M NH ₄ F	6.5	0.43	1.55	a	a	3.66	96.4
6	2M NaOH	9.5	1.97	7.51	8.87	0.27	2.0	8.36
7	11M HNO ₃ -0.1M NH ₄ F	9.5	0.08	0.38	0.25	0.01	4.29	3.46

a Not detected in this leach liquor.

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TABLE A-2

LEACH STUDIES TO DETERMINE COMPOSITION OF TANK 108-SX SLUDGE

Conditions: 4.0 g of Tank 108-SX sludge leached (at 80 C) successively with 10-ml portions of solutions indicated.

No.	Composition	Final Vol, ml	Radioisotope Concentration, $\mu\text{Ci/liter (x } 10^{-4})$			
			^{137}Cs	$^{106}\text{Ru}-^{106}\text{Rh}$	$^{144}\text{Ce}-^{144}\text{Pr}$	$^{152}-^{154}\text{Eu}$
1	H ₂ O	10.4	22.5	a	a	a
2	H ₂ O	8.3	7.51	a	a	a
3	6M HCl	8.3	2.98	17.1	19.3	0.84
4	6M HCl	10.5	a	6.93	4.80	0.21
5	11M HNO ₃ -0.1M NH ₄ F	6.5	a	3.41	2.38	0.072
6	2M NaOH	8.0	a	0.001	a	a
7	11M HNO ₃ -0.1M NH ₄ F	9.5	a	1.10	1.94	0.016

a Not detected in this leach liquor.

ARH-1733

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