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A Leaching Technique For Studying Actinide-Contaminated Hanford Sediments

S. A. Gallagher
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Prepared for the United States
Department of Energy
Under Contract DE-AC06-77RL01030

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PRELIMINARY REPORT

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INTRODUCTION

For more than 30 years, liquid wastes containing low concentrations of actinides from chemical processing plants have been disposed directly to the ground of the Hanford Site via trenches. These liquid wastes contained trace amounts of the actinides plutonium and americium. Because these elements are biologically harmful to man, they must remain isolated from the biosphere. Therefore, the ability of the sediments to provide this protection must be studied. Assessment of the stability of the actinides in the trench sediments is necessary to evaluate the need for long-term radionuclide stabilization measures.

Three possible mechanisms exist for the transport of plutonium and americium in the sediments. These are leaching, diffusion, and particulate transport. Compared to particulate transport, diffusion is generally a slow method of transport, and leaching can occur at the same time as solutions percolate through the sediments. For this reason, three leaching techniques were reviewed, and small column leaching was chosen as the best method for assessing the stability of the actinides in contaminated sediments. This method most closely approximates natural conditions and employs a realistic solution/soil ratio. Procedures are given for implementing this technique. Both the stability and the chemical form of plutonium will be studied. Data will be input to the modeling of transport of radionuclides in sediments and also input to the ongoing assessment of waste sites.

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DISCUSSION

KNOWN LEACHING TECHNIQUES

Soxhlett- or Paige-Type Leaching Technique

The Soxhlett (Mendel and Warner, 1973, and Mendel et al., 1977), the Paige (Paige, 1966), and a number of similar leach techniques were reviewed by Mendel (Mendel et al., 1977, and Mendel, 1973). These techniques can be applied to the determination of a leach rate for a variety of materials such as solidified radioactive wastes, sediments, or rocks.

Each leach technique employs either a sieved powder contained in a wire envelope or a fused sample in a variety of forms such as blocks or spheres. These samples are then contacted by a leaching solution - water was the exclusive leaching solution in Mendel's review (Mendel et al., 1977, and Mendel, 1973).

Both Soxhlett and Paige leach techniques employ a reflux condenser to circulate distilled water at approximately 90°C over the sample. In the Soxhlett leach technique, the sample is sieved to -40 +60 U.S. mesh, and one gram of this sample is enclosed in a 200-mesh stainless steel envelope. This envelope is placed in a bucket equipped with a siphon, and the bucket is hung from a reflux condenser. Distilled water drips from the condenser onto the sample in the bucket until the bucket is full. The water is then drained from the bucket by means of the siphon (approximately every 3 minutes). The solution contained in the distillation flask after a period of 72 hours is analyzed for the element of interest. The sample remaining in the wire envelope is also dried and weighed to determine the weight loss. Here, the leach rate may be expressed in terms of percent weight loss for the entire sample or in terms of $g/cm^2/day$ for a particular ion in solution.

The Paige leach technique is used to predict the long-term leaching behavior of solids. Powdered or bulk samples are leached in recirculated distilled water. Various temperatures are maintained with a heating mantle. The leaching solution is changed daily or weekly initially and at longer intervals toward the end of the leaching test. The solutions are again analyzed for the elements of interest and a leach rate is calculated.

Another leach technique is performed simply by allowing the sample to sit in the leaching solution for a given time. The solution is then analyzed for the element of interest. A slight variation of this leach test involves changing the leaching solution at various intervals with analyses of these solutions after each exchange.

Water leach rates may be obtained by any of these methods in the temperature range of 25°C to 100°C. These leach rates expressed in terms of $g/cm^2/day$ are calculated according to the following formula:

$$\text{Leach rate} = (A/A_o)\ (W/St)$$

where:

A = amount of ion leached
A_o = amount of ion originally present
W = weight of the sample, g
S = surface area, cm^2
t = time, days

No data were collected by the Soxhlett- or Paige-type leach techniques for contaminated sediments. Therefore, no previous research will be referenced for this technique.

Batch Leaching Technique

The batch leach technique was discussed in reviews by Benson (1960) and Routson (1973). This technique has been applied extensively to adsorption of radionuclides on soils.

Batch leaching is an equilibrium experiment in which a known volume of solution is shaken with a soil or sediment sample until equilibrium is obtained. This technique can be applied equally well to both adsorption or desorption of radionuclides on uncontaminated or contaminated soils. Both the soil and the solution are analyzed for the amount of activity present. An equilibrium distribution coefficient (K_d) in mℓ/g can then be calculated according to the following equation:

$$K_d = \frac{(A_s/W)}{(A_l/V)}$$

where:

A_s = equilibrium concentration or activity on the solid phase
W = weight of the solid phase
A_l = equilibrium concentration or activity of the solution phase
V = volume of solution phase

Ames, Rai, and Serne (1976) reviewed a limited number of batch leach studies performed on contaminated and spiked sediments. Data obtained in these studies which may be of interest are presented in Table 1. References for these data are also presented in the table.

Rai and Serne (1977) also performed a batch leaching study which was not included in the review by Ames, Rai, and Serne (1976). They sequentially leached sediments from the 216-Z-9 and 216-Z-12 cribs located in the 200 West Area of the Hanford Site with H_2O, 1.0M $MgCl_2$, 0.05M Na diethylenetriaminepentaacetic acid (DTPA), 30% H_2O_2, acid ammonium oxalate (pH = 3), and 8.0M HNO_2. The percent ^{239}Pu and ^{241}Am extracted by each of these leaching solutions is shown in Table 2. The sediment sample Z-9, 4-5A was obtained from a depth of 4 to 15 cm. It was grayish, gravelly, coarse sand with a pH of 4.3 (1:10 soil to water). Sample Z-9, 4-11A was coarse sand from 5 to 15 cm with a pH of 3.5, and sample Z-12, 1D was a reddish brown, medium to coarse sand collected at 30 to 45 cm with a pH of 6.8.

Column Leaching Technique

Column leaching is an experiment in which various influent solutions are pumped or percolated through a packed soil column. The effluent is analyzed to determine the concentration of radionuclides as a function of throughput volume (Routson, 1960). Radionuclide distribution on the soil column can also be determined by either sectioning the column or surveying the intact column with scintillation detection (Hajek, 1968). Column leaching can also be applied to both adsorption and desorption of radionuclides from uncontaminated soils.

TABLE 1. Percent Plutonium Leached From Various Soils Using a Number of Leaching Solutions.

Leaching Solution	Soil	Plutonium Removed, %	Reference
8M HNO_3	Spiked bentonite (fired at <40°C)	100	(Tamura et al., 1972)
8M HNO_3	Spiked bentonite (fired at 950°C)	12	(Tamura, 1974, 1975a and 1975b)
8M HNO_3	NTS[a] surface soil	58-91	(Tamura, 1974, 1975a and 1975b)
8M HNO_3	NTS	10-15	(Tamura, 1974, 1975a and 1975b)
8M HNO_3	ORNL[b]	60-75	(Tamura, 1974, 1975a and 1975b)
8M HNO_3	ML[c]	80-85	(Tamura, 1974, 1975a and 1975b)
0.1M citric acid (pH = 2.0)	NTS	0.4-1.5	(Tamura, 1974, 1975a and 1975b)
0.1M citric acid (pH = 2.0)	ORNL	23-24	(Tamura, 1974, 1975a and 1975b)
0.1M citric acid (pH = 2.0)	ML	43-59	(Tamura, 1974, 1975a and 1975b)
0.1M sodium citrate (pH = 8.5)	ORNL	7-8	(Tamura, 1974, 1975a and 1975b)
0.1M sodium citrate (pH = 8.5)	ML	8-14	(Tamura, 1974, 1975a and 1975b)
sequential: 0.1M NaCl	Hanford U-Pond	1-3	(Emery et al., 1974)
sequential: 0.1M oxylic acid-2.0M $(NH_4)_2C_2O_4$	Hanford U-Pond	3-5	(Emery et al., 1974)
sequential: 0.01M EDTA	Hanford U-Pond	2-22	(Emery et al., 1974)
sequential: CCl_4	High salt, acidic, waste contaminated soil	0.01-3	(Swanson, 1973)
sequential: ethyl alcohol	High salt, acidic, waste contaminated soil	0.1-9	(Swanson, 1973)
sequential: 0.1M $MgCl_2$	Lake Michigan sediment	None or small	(Edgington et al., 1975)
sequential: Sodium dithionite-0.3M sodium citrate	Lake Michigan sediment	almost all of extractable plutonium	(Edgington et al., 1975)
sequential: 0.1N NaOH	Lake Michigan sediment	None or small	(Edgington et al., 1975)
sequential: fusion	Lake Michigan sediment	None or small	(Edgington, et al., 1975)
1.0N ammonium acetate	$Pu(NO_3)_4$ spiked soil	0.06	(Wilson and Cline, 1966)
0.1N HNO_3	$Pu(NO_3)_4$ spiked soil	0.14-0.64	(Wilson and Cline, 1966)
1.0N HNO_3	$Pu(NO_3)_4$ spiked soil	0.25-0.32	(Wilson and Cline, 1966)
2×10^{-4}N DTPA	$Pu(NO_3)_4$ spiked soil	0.03-0.1	(Wilson and Cline, 1966)
H_2O_2 (80°C, pH = 5.8)	Bombay sediments	No release	(Pillai and Mathew, 1975)
Alkali	Bombay sediments	0.4-7.5	(Pillai and Mathew, 1975)
Seawater	Bombay sediments	No release	(Pillai and Mathew, 1975)

[a]NTS - Nevada Test Site

[b]ORNL - Oak Ridge National Laboratory

[c]ML - Mound Laboratory

TABLE 2. Percent ^{239}Pu and ^{241}Am Extracted From Sequential Treatment of Contaminated Sediments with Various Chemicals (Rai and Serne, 1977).

Treatment	^{239}Pu			^{241}Am		
	Z-12, 1-1D[a]	Z-9, 4-5A[a]	Z-9, 4-11A[a]	Z-12, 1-1D[a]	Z-9, 4-5A[a]	Z-9, 4-11A[a]
H_2O	0.44	0.15	1.29	0.47	0.35	0.82
$MgCl_2$	0.03	0.05	1.34	0.23	6.17	3.09
DTPA	11.90	34.55	11.43	18.09	38.40	10.41
H_2O_2	0.96	4.13	15.30	0.92	1.35	5.16
AAO[b]	10.33	43.38	23.08	0.97	15.74	8.55
HNO_3	71.02	0.48	0.55	70.08	2.57	7.40
Subtotal	94.68	82.74	52.99	90.76	64.58	35.43
Residual (sediment)	5.34	17.26	47.01	9.27	35.42	64.56

[a]The numbers Z-12 and Z-9 are crib numbers. The complete definition requires the number 216- preceed these abbreviated titles. The number 1-1D, 4-5A, and 4-11A are sample designations.

[b]Acid ammonium oxalate.

Previous research performed on contaminated and spiked sediments using the column leaching technique has been reviewed (Ames, Rai and Serne, 1976). One of these studies was the column study of the mobility of plutonium in a contaminated sediment from the 216-Z-1A crib conducted by Hajek (1966). Leaching of the sediment with groundwater and 1.0M $NaNO_3$ removed 0.1% of the total sediment plutonium after 13 column volumes and 3.5% after 100 column volumes, respectively. Knoll (1969) leached spiked sediments in columns with various organic wastes. He found that 130 column volumes of TBP-CCl_4 (tributylphosphate-carbon tetrachloride) solution leached 5.0% of the plutonium, 30 column volumes of DBBP (dibutylbutyl phosphonate) leached 40.0%, and D2EHPA (di-(2-ethylhexyl) phosphoric acid) and hydroxyacetic acid removed 30.0 and <40.0%, respectively.

SELECTION OF A LEACHING TECHNIQUE

To assess the stability of the actinides in Hanford sediments, it is desirable to reproduce natural conditions as nearly as possible. That is the percolation of groundwater, rainwater, or waste solution through sediments at ambient temperature must be simulated in the laboratory.

The Soxhlett, Paige, and other leaching techniques described by Mendel (Mendel et al., 1977, and Mendel, 1973) were designed primarily for the evaluation of radioactive waste products. Although these techniques require a minimum of time and equipment, the conditions under which they are performed provide a poor approximation of natural conditions. Both the Soxhlett and the Paige techniques are restricted to the use of distilled water. All of these techniques require alteration of the soil by crushing, grinding, or forming. An unrealistically high solution/soil ratio is also employed in these techniques. Leach rates are often found to depend on the duration of contact with the leaching solution. The effects of flow rate and degree of saturation cannot be studied with this technique.

Batch leaching is a simple, low-cost test. A variety of leaching solutions can be employed, and numerous samples can be run at one time.

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This technique involves an equilibrium condition which is not always achieved in a sediment column. Some of the disadvantages of this technique are alteration of the sediments through grinding or crushing, the unrealistic solution/soil ratio, and the difficulty in separating the solid and liquid phases. Again, the effects of flow rate and degree of saturation cannot be studied.

The column leaching technique most closely approximates natural conditions. With this technique, it is possible to leach the sediment with a variety of solutions at a realistic solution/soil ratio. The sediment sample itself is not altered appreciably before it is packed into the columns, and the sediments in the columns are contacted by fresh solution throughout the experiment. Flow rates which approximate natural percolation of rainwater or groundwater through the sediments can be simulated, and adsorption as well as desorption experiments can be performed. Some of the disadvantages of this technique are that only a single experiment requiring elaborate equipment can be performed and are sometimes affected by channeling. However, the advantages of being able to closely simulate natural conditons far outweighs any of the disadvantages.

Because the small column leaching technique most closely approximates natural conditons, this method has been selected for studying the stability of actinides in the Hanford sediments. Methods for implementing this study will now be presented.

METHODOLOGY

Column Design and Set-Up

Two types of columns can be employed in small column leaching. These are the saturated and unsaturated columns as shown in Figures 1 and 2. For saturated columns, the leaching solution, which is contained in a graduated cylinder is forced upflow through the soil column, and equal volumes of the effluent are collected by an automatic fraction collector. For unsaturated columns, the influent solution is applied to the top of the sediment column with a syringe pump, and the effluent

samples are again collected by an automatic fraction collector. Solution flow through the column is maintained by the use of a fritted glass disk attached to the bottom of the column. By positioning the column directly over a vacuum chamber containing the fraction collector, the vacuum chamber and fritted glass disk will maintain a constant suction on the lower end of the column. The most fundamental difference between these two techniques is the length of time required to perform a given experiment. The unsaturated column which closely approximates conditions present in a desert environment becomes a longer term study.

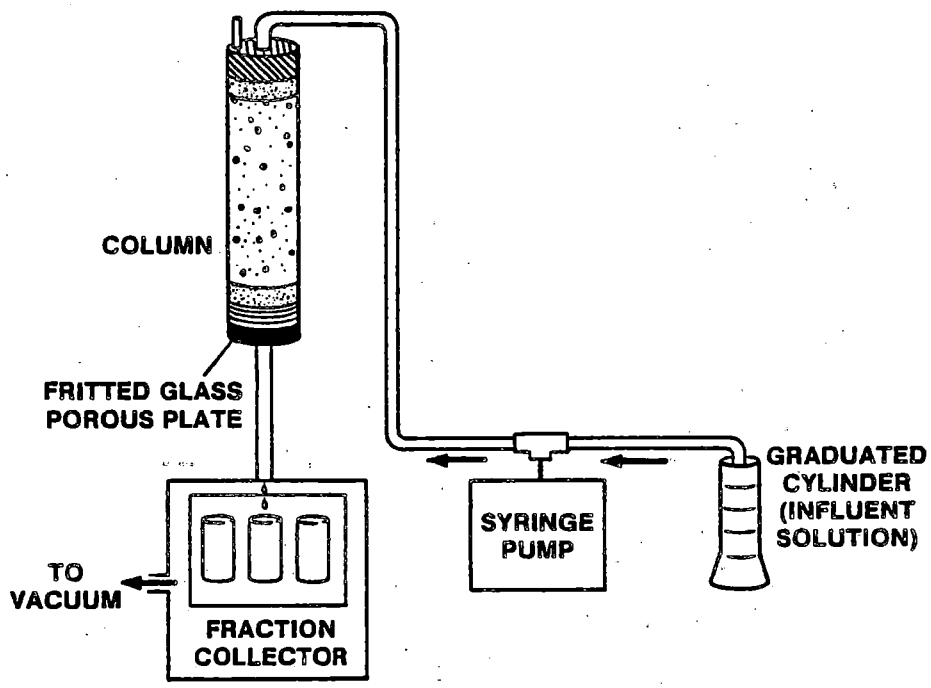

FIGURE 1. Schematic of Saturated Column Apparatus.

Initial work will be done using saturated columns. The saturated column will approximate a worst case situation such as increased rainfall, groundwater intrusion or flooding. This type of column is also an end member in the series of unsaturated columns and is a much simpler system to study. Because they more closely approximate desert conditions, unsaturated columns will be studied when confidence has been gained with the saturated column work.

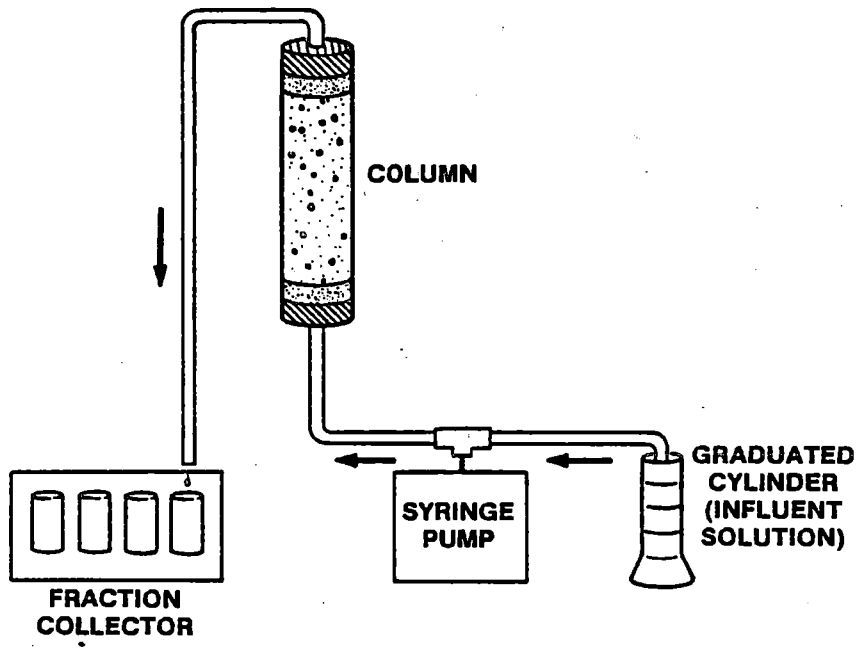

FIGURE 2. Schematic of Saturated Column Apparatus.

Columns can be made of pyrex, saran plastic, stainless steel, or other suitable material which will not react with any of the solutions to be passed through the sediments. For this study, pyrex tubing will be used for the construction of the columns because of its ready availability and its nonreactivity with the wide variety of solutions which will be used in this leaching study.

It is desirable to use the shortest column which will give reproducible results because less sediment will be required to pack a short column. Also, short columns will require less time for a given number of pore volumes of leaching solution to pass through the column. A small column diameter will help prevent wall effects such as channeling. Thus, coating the inside of the columns with silicon oil to prevent similar wall effects will be unnecessary. A column of approximately

2.54-cm ID (a standard pyrex tubing size) and approximately 20 cm in length will be used for the first leaching tests. The effect of column diameter and length on the leaching data will be studied.

Flow rates which will allow a steady flow of leaching solution through the column will be employed. A flow rate of 1 mℓ/cm^2/hr will be used for the first set of columns. A minimum flow rate of 0.1 mℓ/cm^2/hr and a maximum flow rate of 2 mℓ/cm^2/hr will be used in studies to determine the influence of flow rate on leaching.

Soil Preparation

It will be necessary to begin with approximately 3 kg of moist sediment to fill 16 columns 2.54 cm in diameter by 20 cm in height at a density of ∿1.6 g/cm^3. The "as received" sediment will be placed in a glove box where it can be stored and handled without the danger of airborne contamination. Before handling, the sediment will be air dried overnight by spreading it in a thin layer on a sheet of smooth paper. During drying, a porous sheet of tissue paper will be placed over the sediment to prevent any possible contamination in the glove box from settling on it.

The following procedure for sediment preparation will be employed and be carried out inside a glove box:

1. Weigh out approximately 3 kg of sediment and mix the sediment thoroughly by tabling to homogenize the sediment.

2. Pass all of the "as received" sediment sample (except the stones and gravel) through a 2-mm sieve to remove large particles which may cause channelling. Use a mortar and pestle or rolling pin if necessary to break up aggregates. Discard the stones and gravel.*

3. Place the sieved sediment in a polyproplylene bottle, seal tightly, and place on a roller overnight to further homogenize the sediment.

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*Gravel retained on a 2-mm sieve is removed because of its interference and lack of exchange capacity.

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4. Remove the sediment from the polyproplylene bottle and form it into a cone-shaped pile on a large sheet (Kraft paper, rubberized cloth, canvas, etc.) by lifting each corner of the sheet as necessary.

5. Flatten the cone and divide it into four equal parts as though cutting a pie.

6. Remove three of these parts and place each part in a separate container labeled 1, 2, and 3, respectively.

7. Form a cone from the remaining part, flatten the cone, and quarter as before.

8. Pack the columns as described in the section on column preparation using 162 g of sediment from one of these four equal parts to pack each column at a density of 1.6 g/cm^3 and a sediment column height of 20 cm.

9. Label the columns 4A, 4B, 4C, and 4D.

10. Pack the remaining columns using the same procedure, labeling each of the columns prepared from the sediment in container 1 as 1A, 1B, 1C, and 1D. Columns prepared from sediment in containers 2 and 3 shall be labeled 2A, 2B, 2C, and 2D and 3A, 3B, 3C, and 3D, respectively.

The column leaching tests should then be performed in quadruplicate using one column prepared from each container, e.g., 1A, 2A, 3A, and 4A for each different leaching solution.

Column Preparation

The sediment columns will be constructed inside a glovebox as follows:

1. Place the pyrex glass column in a clamp and cement the influent stopper (a one-holed rubber stopper with a small glass tube protruding) into the bottom of the tube using RTV rubber cement. (For unsaturated columns, a fritted glass disk will be substituted for the one-holed rubber stopper in the bottom of the column).

2. Place a glass wool plug in the bottom of the tube.

3. Weigh the tube.

4. Pour a predetermined amount of sediment continuously into the column while rapping the column with a rubber tube or vibrating the column with a hand vibrator.

5. Measure the length of the sediment in the column and weigh the column.

6. Place a glass wool plug on top of the sediment.

7. Cement the effluent stopper (same as influent stopper) into the top of the column using RTV rubber cement.

The bulk density (B_d) of the sediment can then be calculated as:

$$B_d = \frac{\text{weight of sediment}}{\text{volume of sediment}}$$

The pore volume of the column can be calculated from the bulk density and the particle density (P_d) using the following equation:

$$V_p = 1 - B_d/P_d$$

where the particle density is assumed to be constant and approximately equal to 2.65 g/cm^3 for Hanford sediments. The column volume (total volume occupied by the sediment) can also be calculated from these data.

Sample Collection and Analysis

A complete sediment analysis should be performed before and after leaching. This includes the determination of the cation exchange capacity, exchangeable cations such as Ca^{2+}, H^+, Mg^{2+}, K^+, and Na^+, anions such as NO_3^-, $CO_3^=$, nitrite and F^-, pH, E_h, moisture content, volatiles, mineralogy, surface area, B_d, grain density, grain size distribution, and the concentration of ^{239}Pu, ^{240}Pu, and ^{241}Am.

The effluent leach solution will be collected by an automatic fraction collector. At various intervals, the effluent will be analyzed for plutonium and americium using an intrinsic germanium diode or a scintillation counter. The plutonium and americium content in the effluent

solution will be determined both before and after filtration through a 0.01-μm millipore filter. This procedure will allow differentiation between species which were actually leached by the leaching solution and those which were transported as particulates by the leaching solution.

The total quantity of plutonium or americium removed by the leaching solutions will be recorded as well as the number of column or pore volumes of leaching solution used. If the original quantity of plutonium or americium present in the sediment is known, a percentage of the plutonium or americium removed can also be obtained. The original quantity of plutonium or americium in the sediments can be determined using an intrinsic germanium diode or a scintillation counter. If a plot of the total quantity of plutonium or americium leached versus time or some function of time is linear, it may indicate a possible mechanism for the leaching of these actinides.

AREAS TO BE STUDIED

To assess the stabilities of the actinides in the contaminated sediments, simulated groundwaters, rainwater, and simulated waste solutions will be passed through the sediment columns. This will give an indication of what could happen to the actinides in the trenches under natural conditions or in the event that waste disposal to a particular trench was restored. Plutonium and americium leach rates will be recorded as a function of time and the number of column volumes (pore volumes) passing through the columns.

Another area of interest which can be addressed using this technique is the chemical form of plutonium in the sediments. Plutonium can exist in the sediments in four chemical forms:

1. soluble
2. exchangeable
3. coprecipitated or complexed
4. discrete solid phases.

Of these forms, the soluble and exchangeable forms of plutonium are considered to be readily mobile while the coprecipitated or complexed forms of plutonium are considered to be tightly bound. It should be possible, therefore, to determine which of these forms of plutonium exist in a particular sediment by leaching sediment columns with various solutions. Batch equilibrium tests may also be of some value in determining the chemical forms of plutonium. Exchangeable plutonium will be determined by leaching the sediment column with a 1.0 N $MgCl_2$ solution at the soil pH. Problems may be encountered with this method if a carbonate source is present in the sediment sample; therefore, a check will be made for any changes in the alkalinity which might occur during leaching. It will also be possible to decompose any metal hydroxides (reducible plutonium) present in the sediments by the use of a 0.1M oxalic acid with 0.2M ammonium oxalate. Some competition for this reaction could occur if appreciable amounts of phosphates are present. Complexable plutonium will be leached by either 0.05M DTPA or ethylenediaminetetraacetic acid in 0.1 N $MgCl_2$ at pH 7. The data obtained with this leaching solution should be viewed critically due to the uncertainty about the competition of other metals (nonactinides) during this reaction. A 30% H_2O_2 solution or a sodium hypochlorite solution will be used as a leach solution to determine whether or how much plutonium is complexed with the organic matter in the sediments. A number of other reactions may also occur when this solution is employed. Therefore, it will be noted if oxygen bubbles are evolved during leaching, and a check will be made for any changes in alkalinity which may occur during leaching. A final leach with 8 N HNO_3 can be used to remove plutonium which is present in highly insoluble forms.

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