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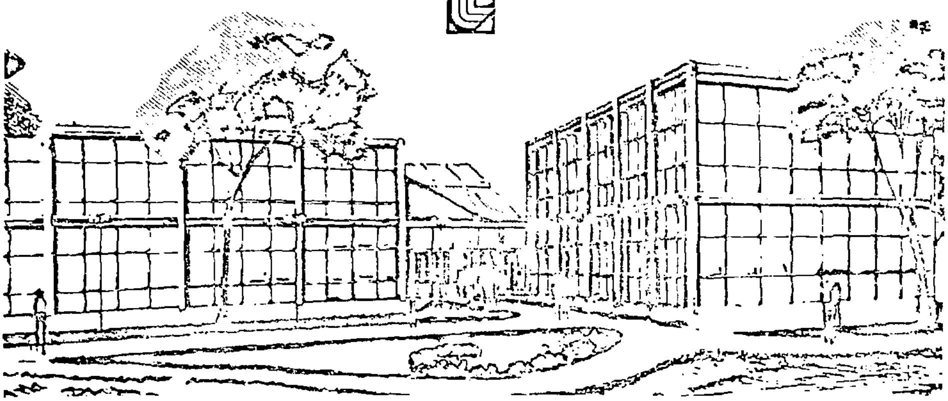

LEACHING CHARACTERISTICS OF ACTINIDES FROM SIMULATED REACTOR WASTE, PART 2*

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ABSTRACT

Leach rates for ^{237}Np and ^{239}Pu are investigated with a single-pass leaching system. The factorial experimental design uses several combinations of solution composition and flow rate; and two temperatures, 25C and 75C. The 25C results are compared with those from a modified IAEA procedure. At 25C, leach rates decrease with time. Agreement between results from the single-pass and modified IAEA methods is fair with WIPP brine leachant, good with $NaHCO_3$, and good with distilled H_2O. Leach rates are approximately independent of flow rates at room temperature, but increase with flow rates at high temperature. Rates for ^{237}Np increase with temperature, but those for ^{239}Pu either decrease or do not change with temperature.

INTRODUCTION

This investigation is part of the Waste Isolation Safety Assessment Program (WISAP) conducted by PNL for the Department of Energy. One of the important goals of the WISA program is to be able to calculate the rate of release and subsequent migration of radionuclides in geologic formations surrounding repositories.

The present study has the following objectives in support of that goal:

- To provide information on the source term to be used in radionuclide migration and safety assessment calculations.

*"Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under contract number W-7405-ENG-48."

This includes measuring the effect of solution composition, flow rate, temperature, and time on the leach rate of simulated high level reactor waste glass.

- To compare results obtained from a dynamic one-pass leaching method developed at LLL with those from the modified IAEA method used at PNL.

This interim report includes results to 420 days; a final report will be issued with PNL after the conclusion of the study.

EXPERIMENTAL WORK

As shown in Figure 1, LLL uses a statistically designed factorial experiment with unequal replication.[1] Leach rate is the dependent variable; and solution composition, flow rate, temperature, and time are the independent variables. The duration of the experiment is 420 days; each channel is sampled at 11 times during this interval. The PNL experimental design is a modification of the IAEA method[2] in which monthly sampling continues indefinitely and experiments are triply replicated. Both PNL and LLL use the same waste form, leaching solutions, and low temperature (25C) so that the low temperature results can be compared directly. The PNL simulated reactor waste is a sodium zinc borosilicate glass (76-68) in the form of hemispherical beads about 8 mm in diameter. The composition is like that of fully radioactive waste, except for the substitution of non-radioactive fission products. The radionuclides are:

$^{238}U_3O_8$ (4.2 w/o), $^{237}NpO_2$ (0.46 w/o), and $^{239}PuO_2$ (0.046 w/o).

The leachant solutions are distilled H_2O, 0.03M $NaHCO_3$, and synthetic WIPP brine. The final (420-day) effluent sample was collected in June. We are continuing with radiochemical and chemical analysis of the samples, data reduction and interpretation, and post-run examination of the sample cells and beads.

For each combination of temperature, flow rate and solution, the leach rate (R) is given as a function of time (t) by the model:

$$R = \alpha + \beta t^{-\gamma} \qquad (1)$$

The use of least-squares procedures determines estimates of the parameters α, β, and γ, which do not depend on the time but may depend on the temperature, flow rate, and solution composition. Analysis of variance methods will then be used to determine the nature of this dependence.

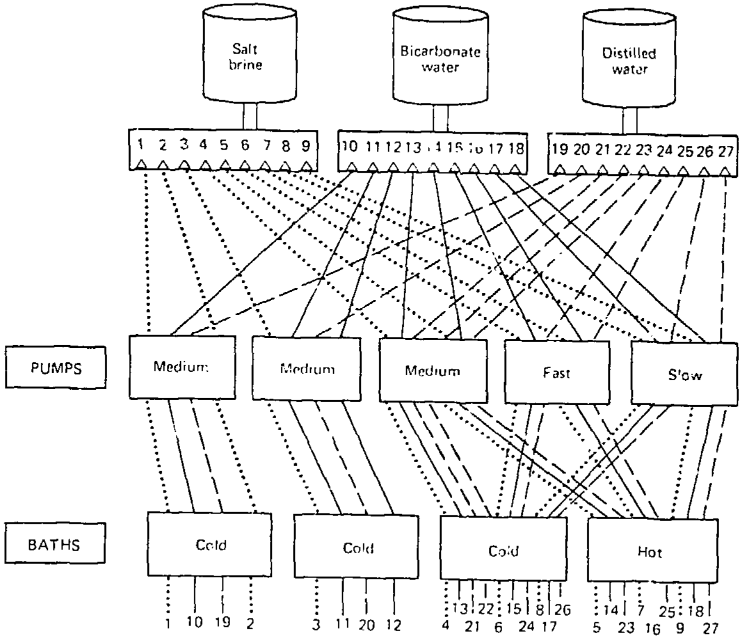

Fig. 1. Experimental Design, One-pass Leaching Study

Preliminary results are presented here without detailed statistical analysis. The rate R vs. t is shown in Figs. 2-7 for the complete experiment to 420 days. In the figures, the full lines correspond to 25C, the dashed lines to 75C, and F, M, S to fast ($300 cm^3/d$), medium ($43 cm^3/d$), and slow ($10 cm^3/d$) flow rates, respectively. Blank results are labeled B; the horizontal dot-dashed lines are mean values. PNL results are shown as filled circles.

Leach rates vary from $2X\ 10^{-4}$ to $2X\ 10^{-7} g/(cm^2.d)$ for ^{237}Np (Figs. 2-4) and from 2 X 10^{-5} to 2 X $10^{-9} g/(cm^2.d)$ for ^{239}Pu (Figs. 5-7). At high temperature the leach rates for ^{237}Np show little dependence on time. The ^{239}Pu leach rates decrease with time, but in a very irregular fashion. Some of the low flow, high temperature ^{239}Pu leach rates appear smaller than those of the blanks (Fig. 5). The reason for this is that the radionuclide concentrations for both leach samples and blanks are near the lower detection limit of the counting system, so that differences between them are not significant. Leach rates for both nuclides increase with flow rate for all solutions. High temperature leach rates are greater than those at room temperature for ^{237}Np, but less than or equal to those at room temperature for ^{239}Pu. We plan to look for ^{239}Pu in various parts of the sample cells and effluent tubing in order to determine whether or not it has been leached from the beads.

At room temperature, leach rates are approximately independent of flow rate; this will be checked for statistical significance during variance analysis. Direct comparison of the results from PNL and LLL at 25C shows that agreement is fair in WIPP brine (Figs. 4 and 7), good in distilled H_2O (Figs. 2 and 5), and good in $NaHCO_3$ (Figs. 3 and 6). The PNL leach rates are consistently lower than those of LLL for ^{239}Pu in WIPP brine (Fig. 7).

CONCLUSIONS

The following general trends can be seen in the results:

- Leach rates increase with flow rate at high temperature, but are approximately independent of it at room temperature.

- Agreement between the results from the one-pass method and those from the IAEA method is fair in the case of WIPP brine solution, and good in the case of the others.

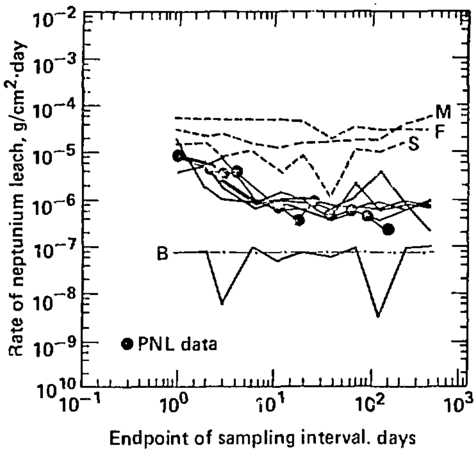

Fig. 2. ^{237}Np Leach Rate in Distilled Water

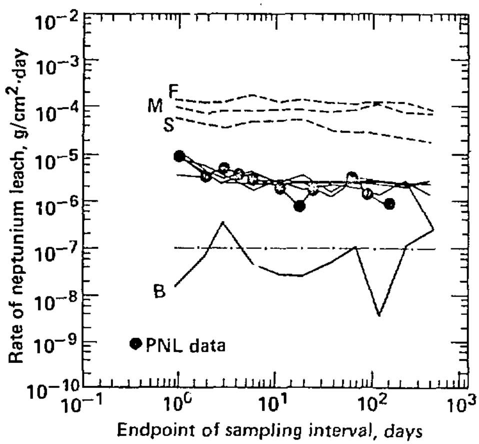

Fig. 3. ^{237}Np Leach Rate in .03M $NaHCO_3$

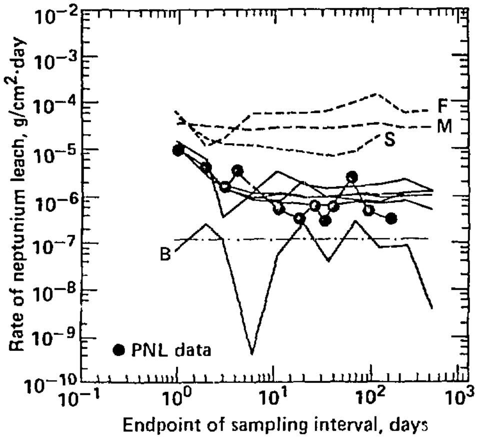

Fig. 4. ^{237}Np Leach Rate in WIPP Brine

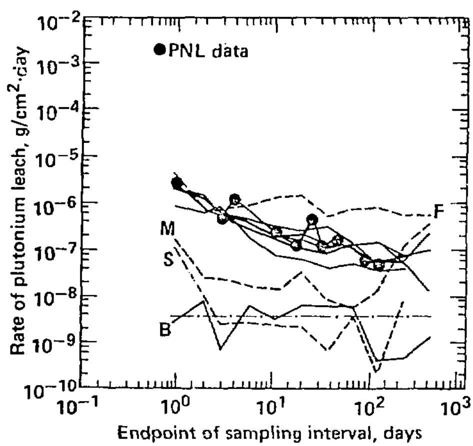

Fig. 5. ^{239}Pu Leach Rate in Distilled Water

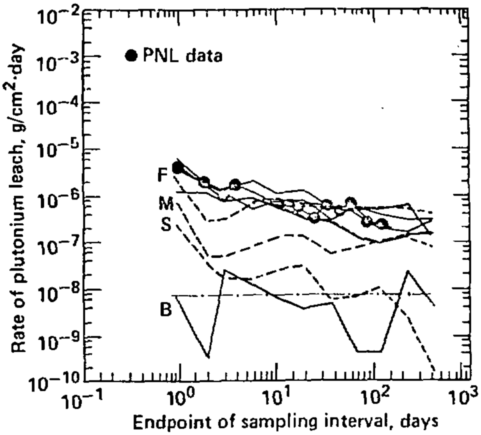

Fig. 6. ^{239}Pu Leach Rate in .03M $NaHCO_3$

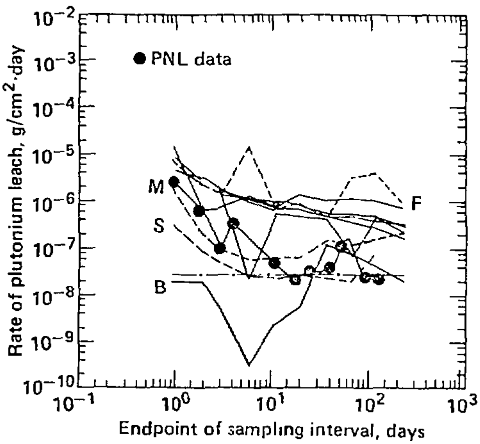

Fig. 7. ^{239}Pu Leach Rate in WIPP Brine

- The ^{237}Np leach rates increase with temperature, but the ^{239}Pu leach rates either decrease with temperature, or do not change.

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