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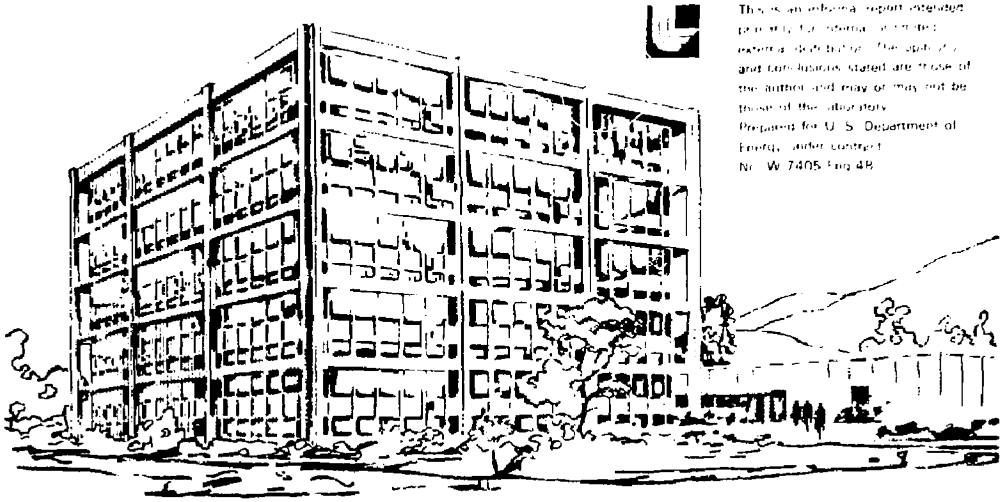

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LEACHING CHARACTERISTICS OF ACTINIDES FROM SIMULATED REACTOR WASTE GLASS

by

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INTRODUCTION

This investigation is conducted for the Office of Nuclear Waste Isolation through Task #2 of the PNL Waste Isolation Safety Assessment Program (WISAP). One of the important goals of the WISA program is to be able to calculate the rate of migration of radionuclides in geologic formations surrounding repositories.

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

- To provide information for the source term in the migration rate calculations. This includes the effect of leaching, solution composition, flow rate, temperature, and time on the leach rate of simulated high level reactor waste glass.

- To compare results obtained from the dynamic one-pass leaching method at LLL those from the static IAEA method at PNL.

This progress report includes results to 120 days. Additional reports will be issued as the results are analyzed statistically, and a final report will be issued with PNL after the end of the experiments.

EXPERIMENTAL WORK

As shown in Figure 1, LLL uses a statistically designed factorial experiment with unequal replication. Leach rate is the dependent variable; and solution composition, flow rate, temperature, and time are the independent variables. The study extends from 1 to 420 days; each of the 28 simultaneous experiments is sampled at 11 times during this interval. The PNL experimental design is a modification of the IAEA method in which monthly sampling continues indefinitely.(1) Also, PNL experiments are triply replicated. Both PNL and LLL use the same waste form, the same leaching solutions, and the same low temperature, 25°C. The PNL simulated reactor waste is a sodium zinc borosilicate glass (WFP76-68) 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 LLL results will be analyzed statistically by fitting the leach rate (R) as a function of time (t) to the model:

$$R = \alpha + \beta t^{-\gamma/2} + \delta \qquad (1)$$

where the parameters α, β, γ are functions of the experimental variables temperature, flow rate, and solution composition; and the error term is δ. The effect of the experimental variables on the model parameter will be explored using statistical analysis of variance methods.

RESULTS

Preliminary results are presented here without statistical analysis. For the first 120 days, log R vs. log t is shown in Figures 2-8. Figure 2 shows the coordinate system which applies to Figures 3-8 and also shows how the PNL data taken at various sampling intervals correspond with subsets of the LLL 25°C data at fast, medium, or slow flow rates. In Figures 3-8, the full lines correspond to 25°C, the dotted lines to 75°C, and F, M, S to fast, medium, and slow flow rates, respectively. In each figure, the blank is shown by a dashed line. The PNL results obtained by their modified IAEA method are shown as filled circles.

The range in leach rates is from 2×10^{-4} to $2 \times 10^{-[illegible]}$ $g/(cm^2 \cdot d)$ for ^{237}Np (Figures 3-5) and from 2×10^{-5} to 2×10^{-9} $g/(cm^2 \cdot d)$ for ^{239}Pu (Figures 6-8). Leach rate usually decreases with time, but at high temperature the time dependence is usually smaller than it is at room temperature. At high temperature, leach rate increases with flow rate for all solutions and both radionuclides. At room temperature, leach rate is approximately independent of flow rate; this will be checked for statistical significance during analysis. 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. As indicated in Figure 2, the room temperature results for LLL and PNL can be compared directly. Agreement is fair in WIPP brine (Figures 5-8) and good in distilled H_2O (Figures 3 and 6) and $NaHCO_3$ (Figures 4 and 7). Further work on this part of the bead leaching studies includes collection of the final (420-day) effluent sample in June, continuation of data reduction and analysis of the results, and post-run examination of the sample cells and beads.

CONCLUSIONS

Even without statistical analysis, some general trends can be seen in the results:

- Leach rate increases with flow rate at high temperature, but is approximately independent of it at room temperature.
- Agreement between the results from the one-pass method and those from the IAEA method are fair in the case of WIPP brine solution, and good in the case of the others.
- The ^{237}Np leach rate increases with temperaure, but the ^{239}Pu leach rate either decreases with temperature, or does not change. This low net transport rate is surprising, and we therefore plan to look for ^{239}Pu in various parts of the sample cells in order to determine where it is located in the system.

REFERENCES

1. E. D. Hespe, "Leach Testing of Immobilized Radioactive Waste Solids, A Proposal for a Standard Method", Atomic Energy Review 9:195 (1971).

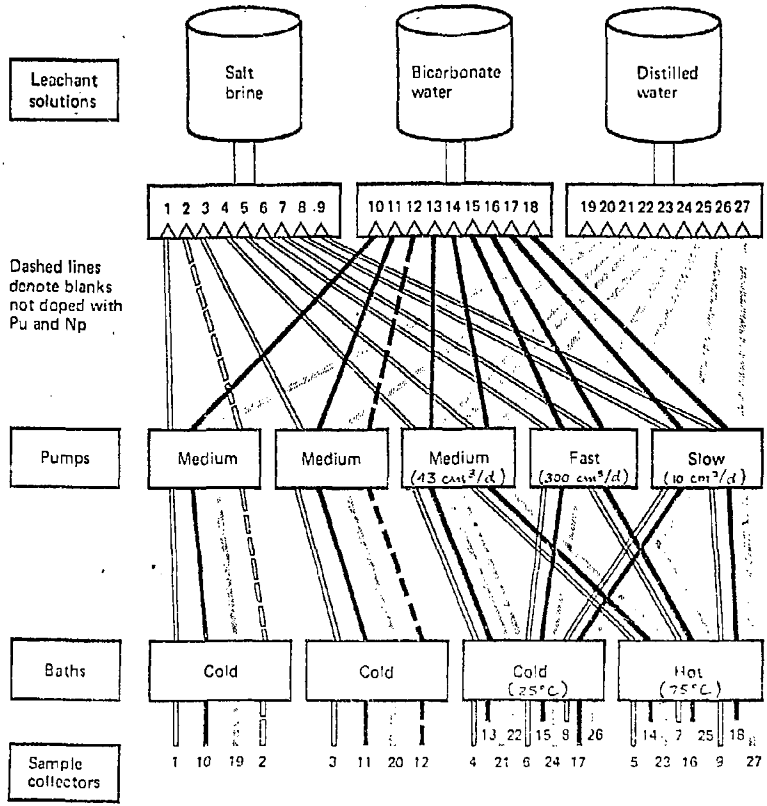
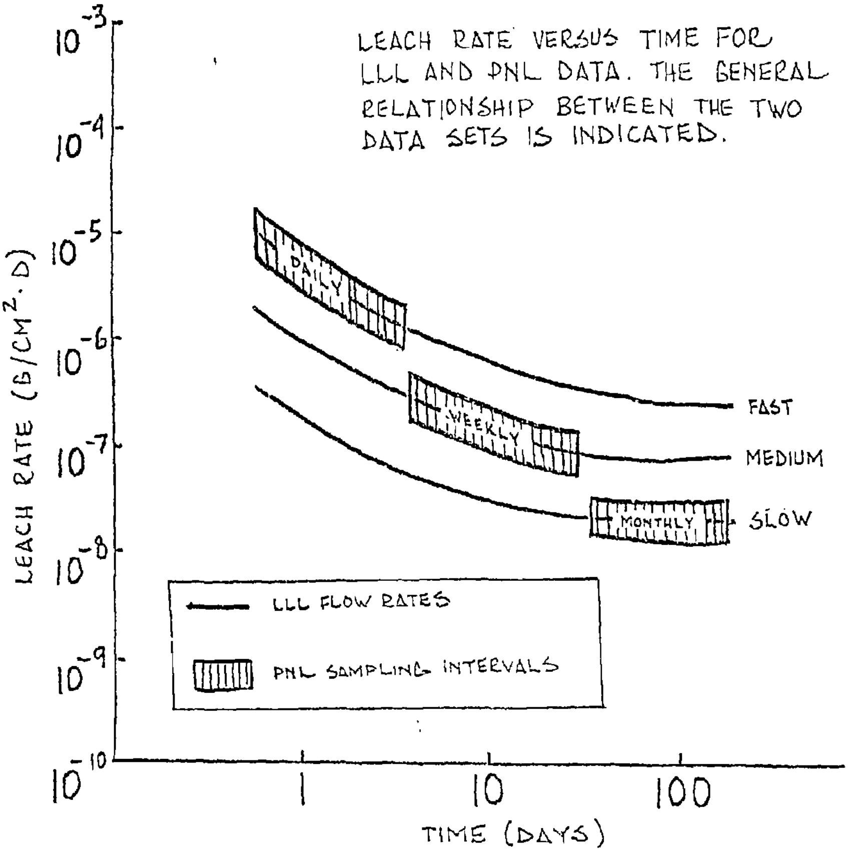
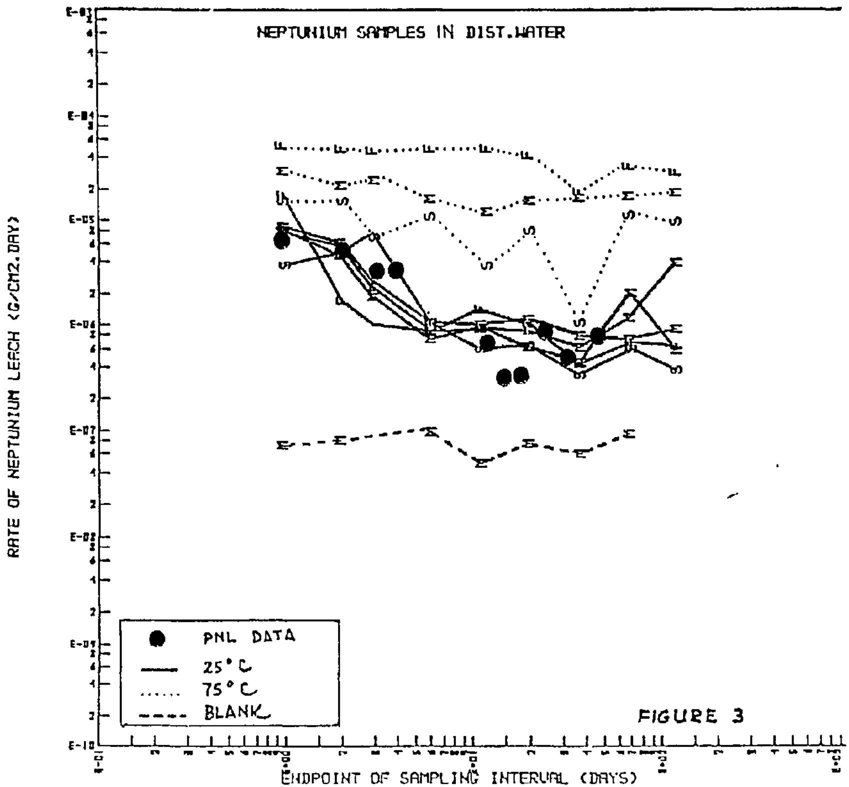


FIGURE 1. EXPERIMENTAL DESIGN, SINGLE-PASS LEACH SYSTEM



LEACH RATE VERSUS TIME FOR LLL AND PNL DATA. THE GENERAL RELATIONSHIP BETWEEN THE TWO DATA SETS IS INDICATED.

FIGURE 2



NEPTUNIUM SAMPLES IN DIST.WATER
RATE OF NEPTUNIUM LEACH (G/CM2.DAY)
ENDPOINT OF SAMPLING INTERVAL (DAYS)
PNL DATA
25° C
75° C
BLANK

FIGURE 3

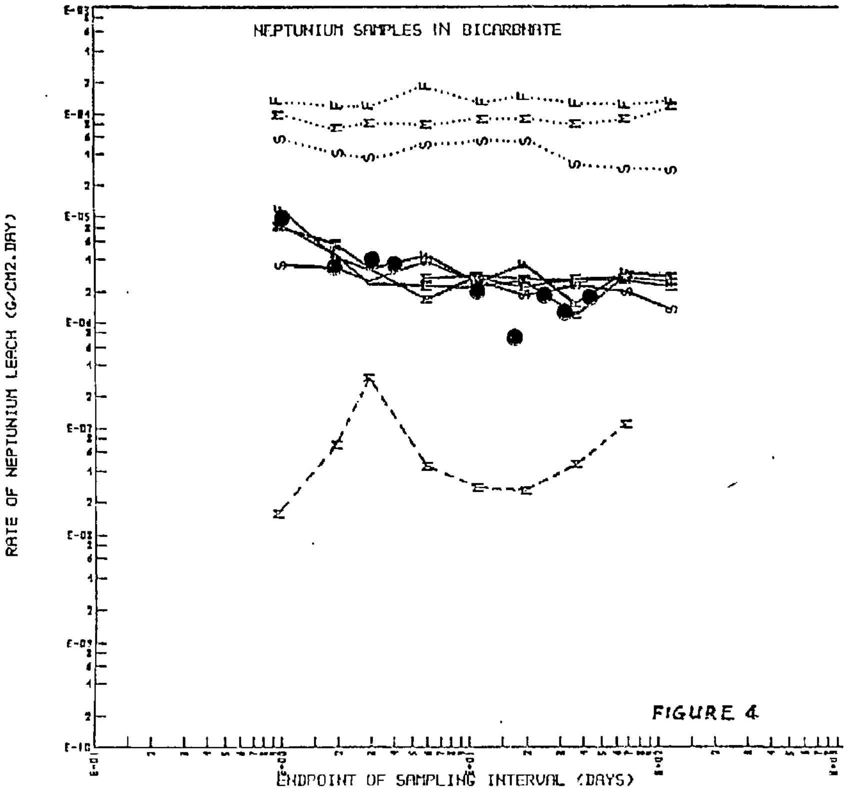
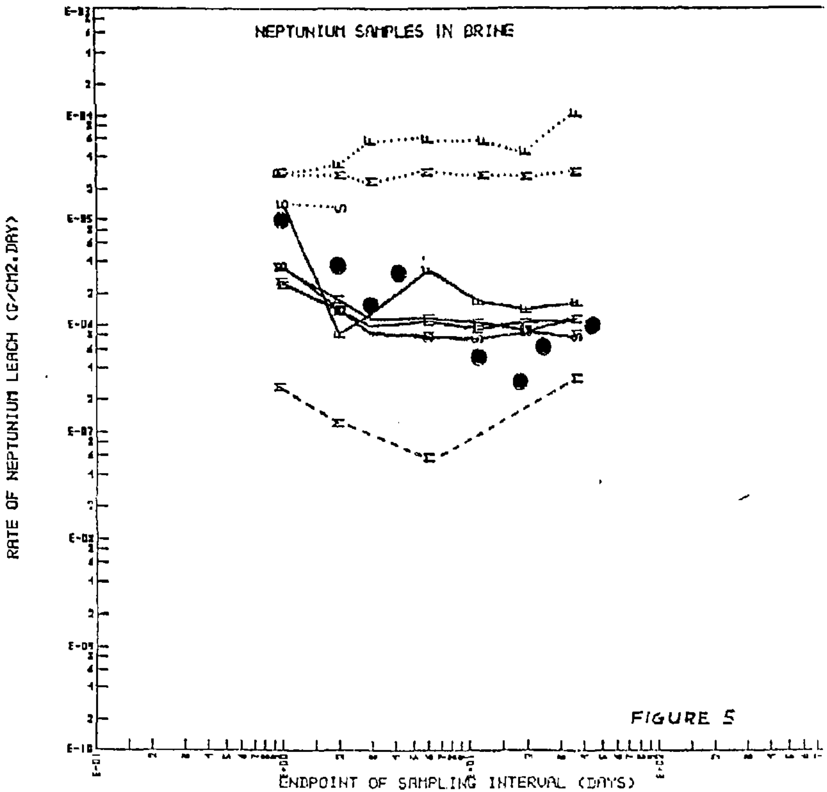

NEPTUNIUM SAMPLES IN BICARBONATE
RATE OF NEPTUNIUM LEACH (G/CM2.DAY)
ENDPOINT OF SAMPLING INTERVAL (DAYS)

FIGURE 4


NEPTUNIUM SAMPLES IN BRINE
RATE OF NEPTUNIUM LEACH (G/CM2.DAY)
ENDPOINT OF SAMPLING INTERVAL (DAYS)
FIGURE 5

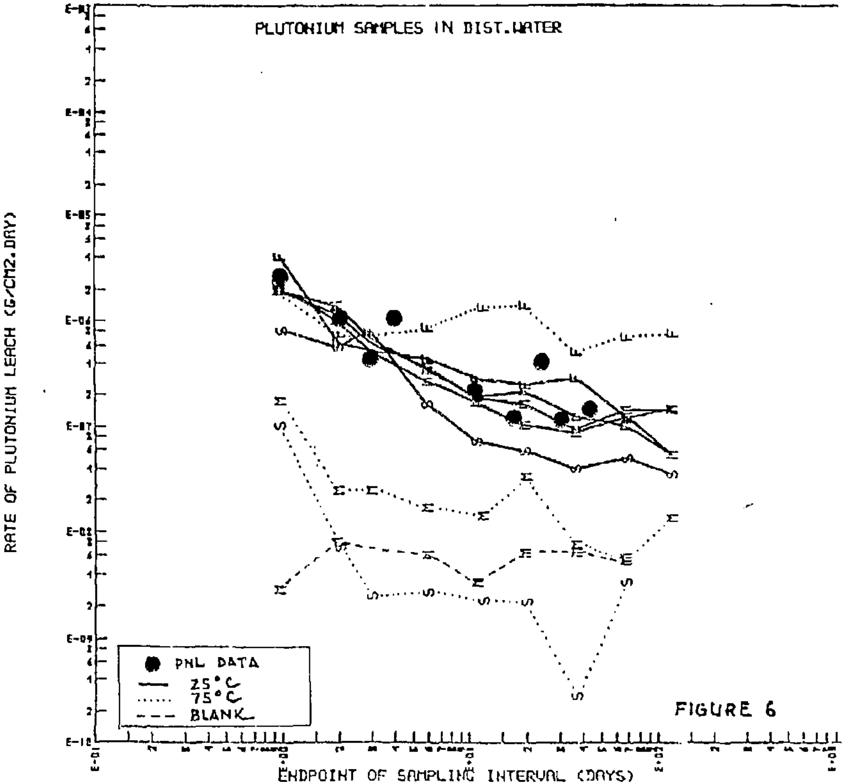

PLUTONIUM SAMPLES IN DIST.WATER
RATE OF PLUTONIUM LEACH (G/CM2.DAY)
ENDPOINT OF SAMPLING INTERVAL (DAYS)
PNL DATA
25°C
75°C
BLANK

FIGURE 6

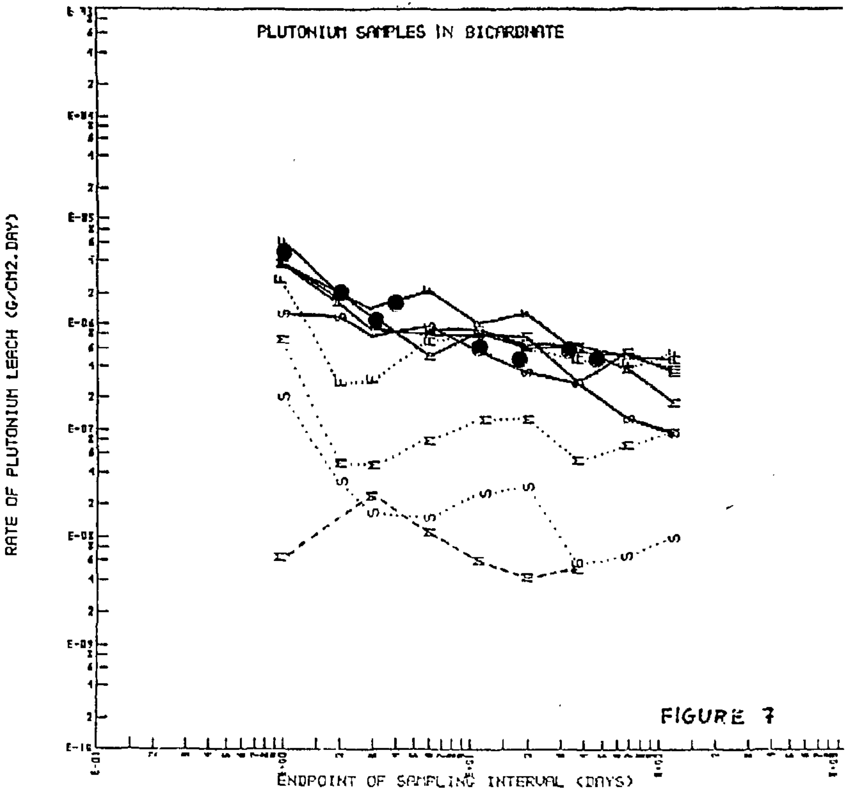

PLUTONIUM SAMPLES IN BICARBONATE
RATE OF PLUTONIUM LEACH (G/CM2.DAY)
ENDPOINT OF SAMPLING INTERVAL (DAYS)

FIGURE 7

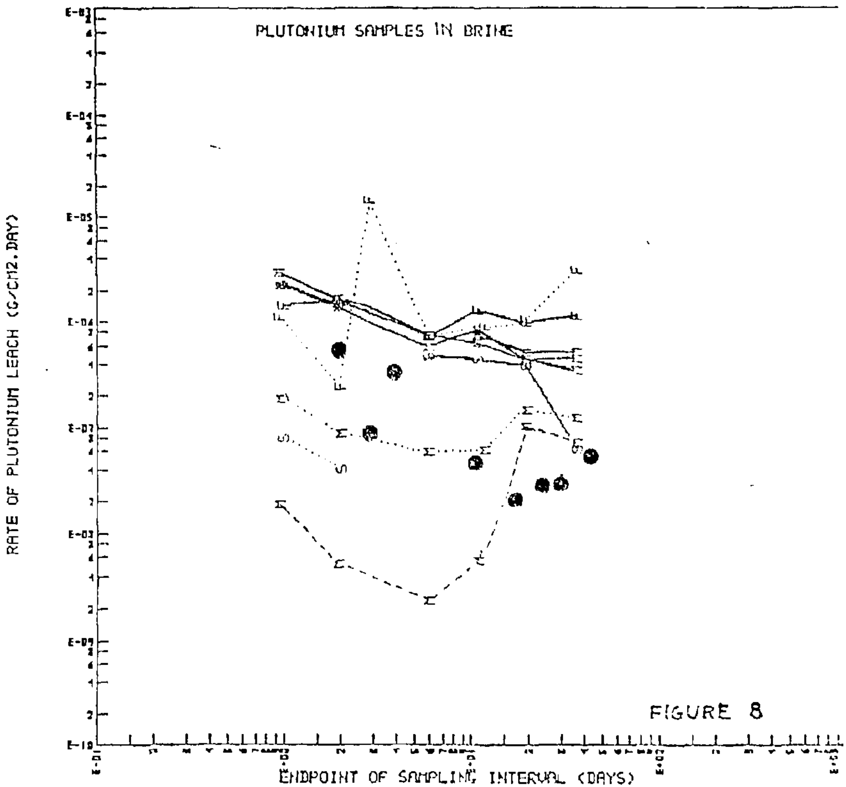

PLUTONIUM SAMPLES IN BRINE
RATE OF PLUTONIUM LEACH (G/CM2.DAY)
ENDPOINT OF SAMPLING INTERVAL (DAYS)

FIGURE 8