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Low-Level Liquid Waste Disposal at the Savannah River Site:

A Large Scale Demonstration of Saltstone (U)

by

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ABSTRACT

Lysimeters are large-scale, field experiments used at the Savannah River Site (SRS) to measure the effect of percolating rainfall on the release of contaminants from wasteforms. The saltstone lysimeters described are demonstrations of a disposal concept for a low-level radioactive waste resulting from the processing of high-level defense waste for vitrification. Results from the lysimeters confirm the efficacy of the slag formulation in retaining chromium and technetium. Lysimeter results were also useful in validating mathematical models used in predicting environmental effects of saltstone disposal in engineered vaults.

INTRODUCTION

At the Savannah River Site (SRS), approximately one hundred million liters of high-level liquid radioactive waste is stored for future reprocessing. This waste has been generated from the production of defense nuclear materials over the past forty years. When reprocessing of the waste begins in 1992, greater than ninety-nine percent of the radioactivity will be precipitated and vitrified into borosilicate glass. The resulting low-level radioactive solution ("decontaminated" solution) will be disposed of on-site.(1,2)

The decontaminated solution will contain primarily sodium nitrate ($NaNO_3$) as well as other trace contaminants. The anticipated nominal composition of the solution is shown in Table 1. This liquid will be mixed with cementitious materials, and poured into engineered concrete vaults. With time, the mixture will solidify into the final product called saltstone.

During the development of the saltstone disposal concept, several cementitious formulations were studied, and two potential recipes were developed. The "reference formulation" consists of class H cement and class C flyash; however, saltstone produced according to this formulation is costly because these materials are not locally available. Additional studies have indicated that materials such as slag cement and class F flyash, which are produced locally, can be used to generate a "slag formulation" more economically.(3)

Since the development of these saltstone formulations, laboratory and field studies have been conducted to evaluate the effectiveness of the saltstone matrices in retaining contaminants of the salt solution(4). The field leaching studies, which continue to date, utilize large-scale lysimeters to study the performance of saltstone monoliths under actual humid weather conditions. The lysimeters are designed to allow the detection of contaminants released to the soil column by the saltstone. Rainwater that infiltrates the lysimeter and percolates past the wasteform emplaced in the apparatus is collected periodically and analyzed for contaminants known to be contained in the saltstone wasteform.

This paper describes the results of the field leaching experiments using wasteforms composed of the reference and slag formulations of saltstone. These experiments, along with laboratory leaching studies, demonstrate the increased effectiveness of the slag formulation for retaining metal contamination.

EXPERIMENTAL DESIGN

Construction of the lysimeter containing the reference formulation monolith was completed in December 1983. During construction, an earthen trench was excavated and lined with a liquid impermeable hypalon layer (45 mil) to form the closed lysimeter system, as shown in Figure 1.A. Pea-sized gravel was placed in the base of the lysimeter to form a sump for the collection of percolate water. A pump was installed in the sump to remove the percolate water periodically for analysis. Native soil placed above the gravel was compacted to the original conditions prior to excavation. Next, the reference formulation of cementitious materials and decontaminated salt solution was poured into the trapezoidal-shaped trench. After the monolith solidified into saltstone, the lysimeter was completed by covering the wasteform with additional native soil and compacting.

The lysimeter containing the slag formulation of saltstone was completed in February 1985. A fiberglass cylindrical tank was installed on end over a concrete pad. The base of the lysimeter was sealed, and a gravel sump and submersible pump were installed (Figure 1.B.) similar to the reference formulation lysimeter. Native soil was compacted over the sump before placing a pre-fabricated, unencapsulated slag wasteform in the lysimeter. Construction was completed by filling the lysimeter with native soil to within six inches of the top of the fiberglass cylinder.

The decontaminated salt solution used in both of the lysimeters was produced from actual high-level waste in a full-scale test of the decontamination process (6). The formulations used in the lysimeters are shown in Table 2.

As shown in Figures 1A and 1B, both lysimeters are equipped with soil moisture probes. These probes are placed in the soil column below the wasteform, but above the sump. These ceramic devices enable the collection of samples of the soil moisture, which allows detection of contaminants released from the monolith prior to washing to the sumps by infiltrating rainwater.

The lysimeters were constructed in the SRS Solid Waste Disposal Facility, and are exposed to the weather. Rainfall infiltrates the lysimeters and percolates downward through the soil column, past the wasteform, and collects in the sumps. However, the two lysimeters and emplaced wasteforms differ in several ways. The lysimeter containing the slag formulation wasteform has a lip that extends above grade and forces any rainfall that occurs within the diameter of the lysimeter to infiltrate into the apparatus. Conversely, large amounts of rainfall on the reference formulation lysimeter may run off before infiltration occurs. The differences in the wasteforms are described in Table 3. These variations in experimental design are considered when interpreting the results.

Samples of the percolate water as well as rainwater are collected biweekly. A tipping bucket rain gauge is used to measure rainfall amounts, and the volume of percolate water pumped from the sump during each sampling period is determined using flow meters. Rainfall and percolate water samples are analyzed for major cations and anions, as well as organics, heavy metals, and radionuclides.

Nitrate, nitrite, and sulfate are determined by ion chromatography. Inductively coupled plasma-emission spectroscopy is used with acidified samples to measure concentrations of calcium, sodium, and chromium; mercury is determined with atomic absorption spectroscopy. If present, organic contaminants such as benzene are observed using gas chromatography with a detection limit of 1 ppb. The pH and conductivity of each sample are also determined. Radiochemical analyses include measurement of total alpha and total nonvolatile beta. The samples are prepared by successive evaporation and dissolution steps directly on the counting planchet. Radiochemical analysis has shown that the nonvolatile beta radioactivity is technetium-99 (4). Tritium is determined by distilling the sample and counting the distillate with liquid scintillation.

RESULTS

Nitrate, which is regulated by the Environmental Protection Agency (EPA), is the contaminant present in saltstone in the largest quantity. Laboratory leaching studies indicate that nitrate is one of the fastest leaching contaminants from saltstone matrices (1). Also, soil sorption studies have demonstrated that nitrate is not significantly sorbed onto soil particles. Tritium is present in the saltstone as tritiated water, which is also rapidly leached and migrates easily through the soil column. Therefore, tritium and nitrate serve as tracers to monitor contaminant release from saltstone.

Previous laboratory leaching studies indicate that nitrate release from the reference and slag matrices of saltstone are roughly equivalent. The reference formulation had an average leaching index of 8.3 compared to the slag formulation that gave a leaching index of 8.9 (3). Figure 2.a. illustrates that the total concentration of nitrate detected in the sumps of both lysimeters is similar.

While nitrate release and migration appear to be similar, the data from the slag lysimeter shows a period of discontinuity. This anomaly occurred during a period of extreme drought when no percolate water could be obtained from the slag lysimeter sump (days 1168-1293, July 1988-February 1989). Once the drought eased, infiltrating water washed a significant amount of nitrate from the soil column into the sump, as can be seen in the dramatic increase in nitrate on day 1462 (May 1989). Perhaps due to it's larger size, the lysimeter containing the reference formulation monolith was less affected by the drought, as indicated by the relatively smooth increase in nitrate.

The tritium detected in the sumps of the two lysimeters (Figure 2.b.) shows similar trends during the drought period. The slag formulation data is discontinuous while the data from the reference formulation is not. However, the tritium levels detected in the sump of the reference formulation lysimeter are significantly less than the levels found in the slag lysimeter sump. Although the source terms for the two monoliths are approximately the same, the cause of the differences in tritium release and migration is not clear.

Although sump water samples from both lysimeters are routinely monitored for chromium, the results are usually below the detection limit of 4 ppb. However, samples of the soil moisture obtained beneath the wasteforms by porous cup samplers yield detectable levels of chromium and technetium. As shown in Table 4, chromium in samples collected below the reference formulation monolith is ten times greater than chromium beneath the slag monolith, while technetium levels are more than 100 times larger. These results confirm that the slag monolith retains technetium and chromium more effectively that the reference formulation (3).

The levels of technetium detected in the sumps of the lysimeters also support this observation. As shown in Figure 3, total technetium detected in the sumps can be plotted as a function of total nitrate detected for both the slag and reference formulations. The data obtained from the reference formulation monolith is linear with a slope of approximately one. This indicates that technetium is released and migrates from the monolith at the same rate as nitrate. The data from the slag matrix, however, has a slope less than one, indicating that technetium release and migration is slower than nitrate.

MATHEMATICAL MODELING

Results from the lysimeters have been used to validate mathematical models used to predict environmental effects from saltstone disposal in engineered vaults (7). Model predictions demonstrate that saltstone will meet or exceed all performance objectives, including the protection of the groundwater according to the Environmental Protection Agency (EPA) drinking water standards for chemicals and radionuclides.

CONCLUSIONS

Field and laboratory leaching studies have been used to support the adoption of the more economical slag formulation of saltstone due to its enhanced performance as described in this paper. The SRS saltstone processing and disposal facilities and the use of the slag formulation have been permitted by the state of South Carolina, and the facility began operation using actual decontaminated salt solution in July 1990.

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Table 1

Approximate Composition of Decontaminated Solution

Chemicals

Component	Weight Percent
H_2O	71.6
$NaNO_3$	14.3
$NaNO_2$	3.6
NaOH	4.2
Na_2CO_3	1.4
$NaAl(OH)_4$	2.6
Na_2SO_4	1.4
NaF	0.05
NaCl	0.1
Na_2SiO_3	0.06
Na_2CrO_4	0.04 (110 µg/g Cr)
NaHgO(OH)	0.00006 (0.5 µg/g Hg)
$NaAg(OH)_2$	1.2×10^{-7} (8×10^{-4} µg/g Ag)
Na_2MoO_4	0.006
KNO_3	0.002
$CaSO_4$	0.03
$Na_2C_2O_4$	0.2
Na_3PO_4	0.1
NH_4NO_3	0.07
$NaB(C_6H_5)_4$	0.06
Other Salts*	0.08
Total Organics	0.09

Radionuclides

Radionuclide	Bq/g
^{3}H	580.
^{14}C	0.3
^{59}Ni	0.007
^{60}Co	7.
^{63}Ni	0.7
^{79}Se	7
^{90}Sr	15.
^{90}Y	15.
^{99}Tc	2,000.
^{106}Ru	1,000.
^{106}Rh	1,000.
^{125}Sb	300.
^{125m}Te	7.
^{126}Sn	7.
^{126}Sb	0.7
^{126m}Sb	7.
^{129}I	1.
^{137}Cs	350.
^{137m}Ba	320.
^{147}Pm	130.
^{151}Sm	70.
^{154}Eu	30.
^{155}Eu	10.
^{238}Pu	20.
^{239}Pu	0.2
Other Beta, Gamma†	4.
Total Alpha	30.

* They include:

	wt %	µg/g
As	$3. \times 10^{-8}$	0.0003
Ba	0.00003	0.3
Cd	0.00001	0.12
Se	0.00007	0.7
Pb	0.00013	1.3

† Miscellaneous short-lived radionuclides

Table 2

Saltstone Formulations

	Weight Percent	
	Slag Lysimeter	Reference Lysimeter
Slag	26	–
Flyash (Class F)	27	–
Cement (Portland Class H)	–	12
Hydrated Lime	1	–
Flyash (Class C)	–	48
Waste Solution	46	40

Table 3

Comparison of Saltstone Lysimeters

	Slag Lysimeter	Reference Lysimeter
Date Started	Feb. 1985	Dec. 1983
Wasteform Shape	Cylindrical	Trapezoidal
Wasteform Weight	240 kg	27,200 kg
Cement	Slag	Portland Class H
Flyash	Class F	Class C
Distance from Bottom of Waste to sump	0.9 meters	2.7 meters
Lysimeter area available for infiltration	2.5 square meters	256. square meters

Table 4

Soil Moisture Results

Lysimeter	NO_3 mg/L	Tc-99 Bq/L	Cr mg/L	Tc/NO_3	Cr/NO_3
Reference	16,000	41	0.043	4	300 x10^7
Slag	5,000	0.4	0.004	0.1	8 x10^7

FIGURE 1.A.

REFERENCE FORMULATION LYSIMETER

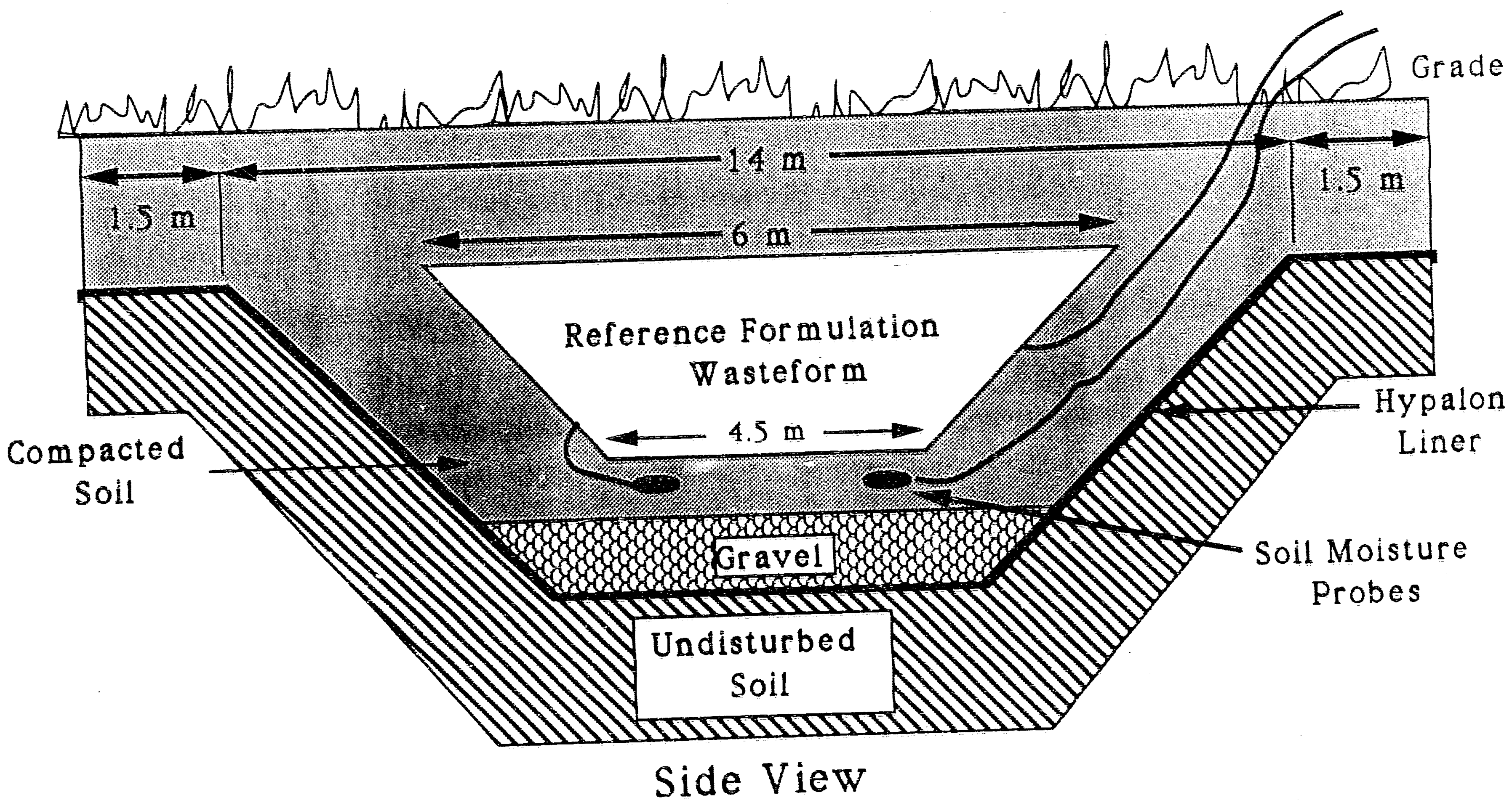

FIGURE 1.A.: Schematic diagram of the lysimeter apparatus containing the reference formulation monolith of saltstone.

FIGURE 1 B.

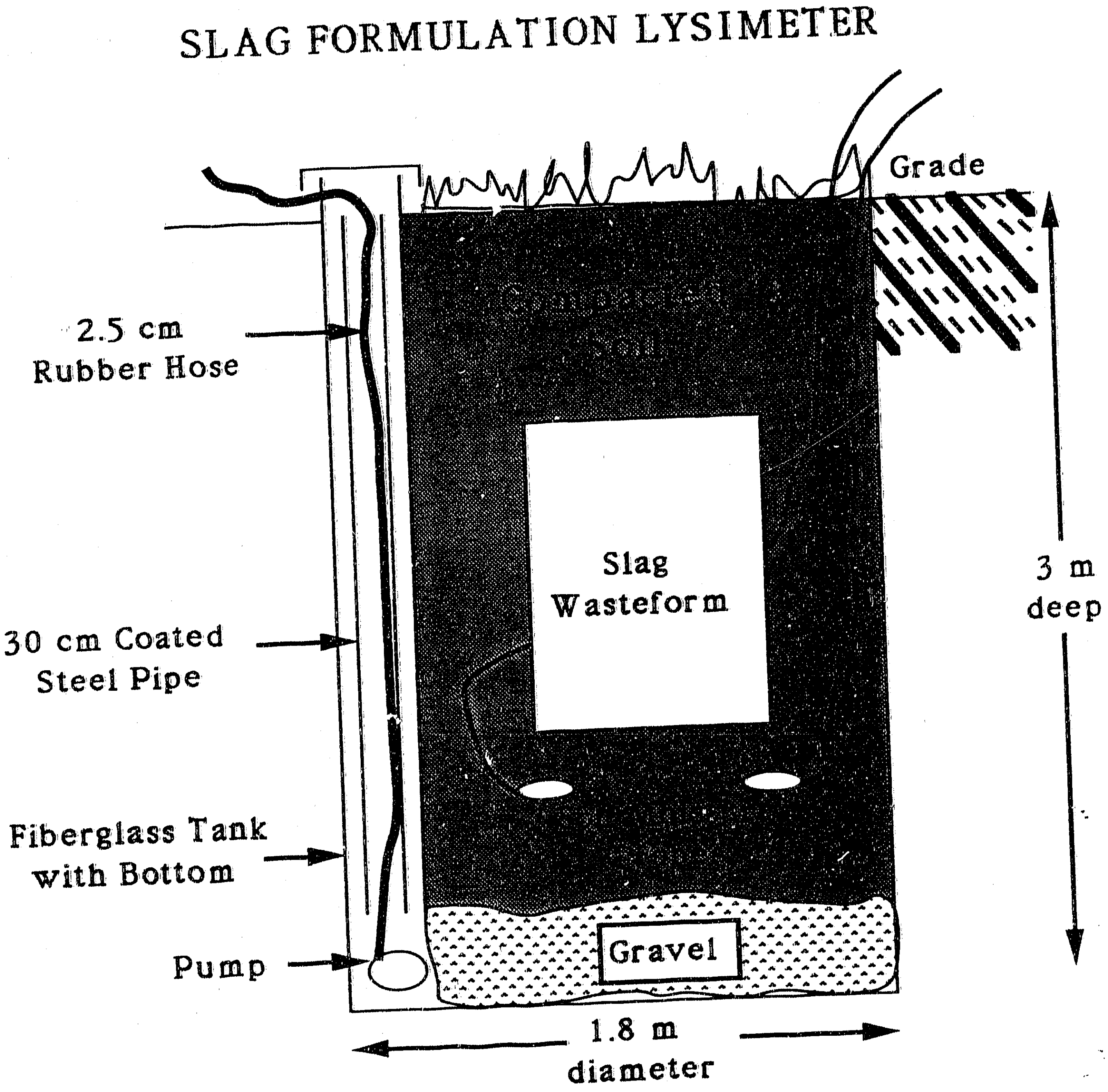

FIGURE 1.B.: Schematic diagram of the lysimeter apparatus containing the slag formulation monolith of saltstone.

FIGURE 2.A.

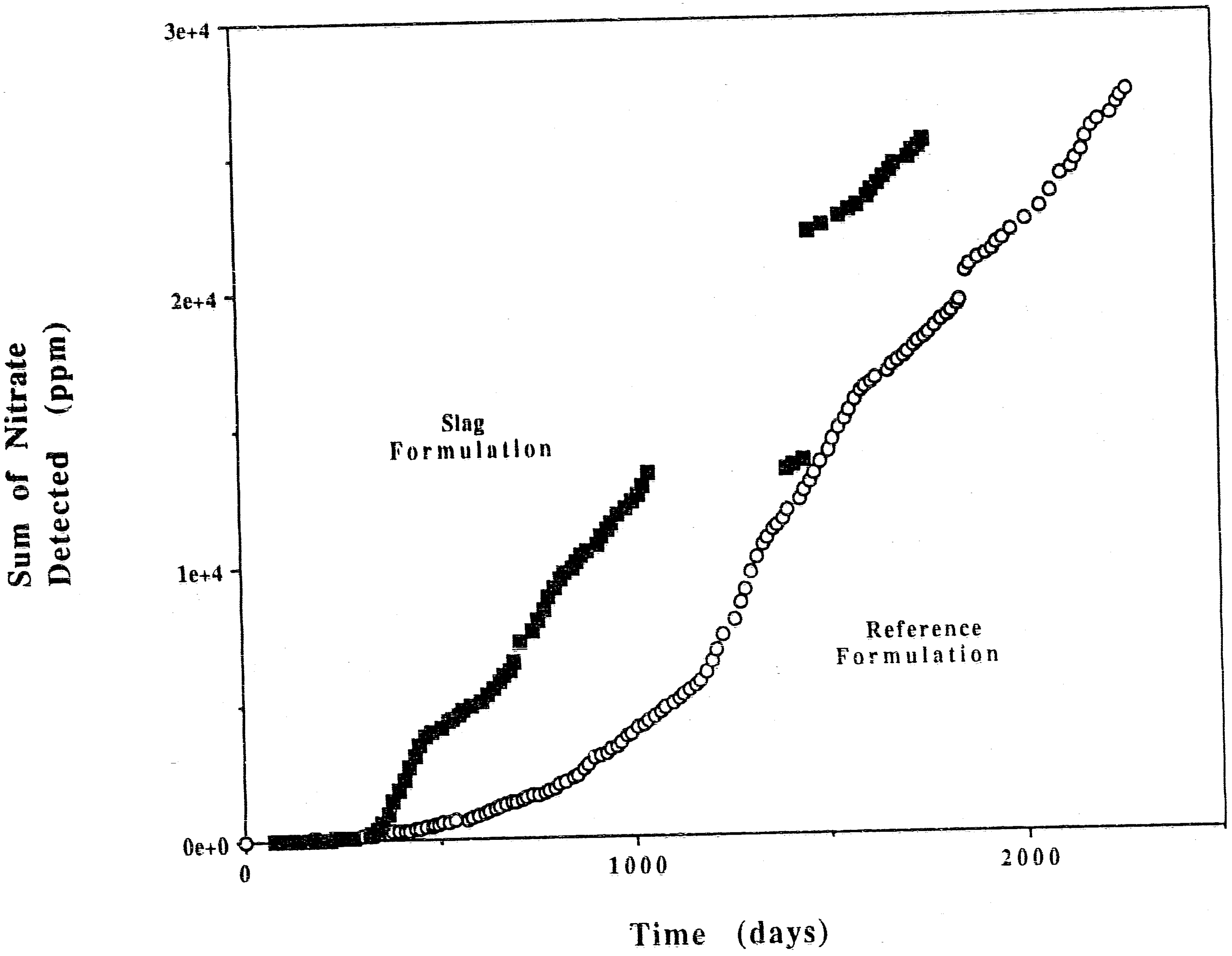

FIGURE 2.A.: The sum of nitrate concentration detected in the sumps of the lysimeters with time.

FIGURE 2 B.

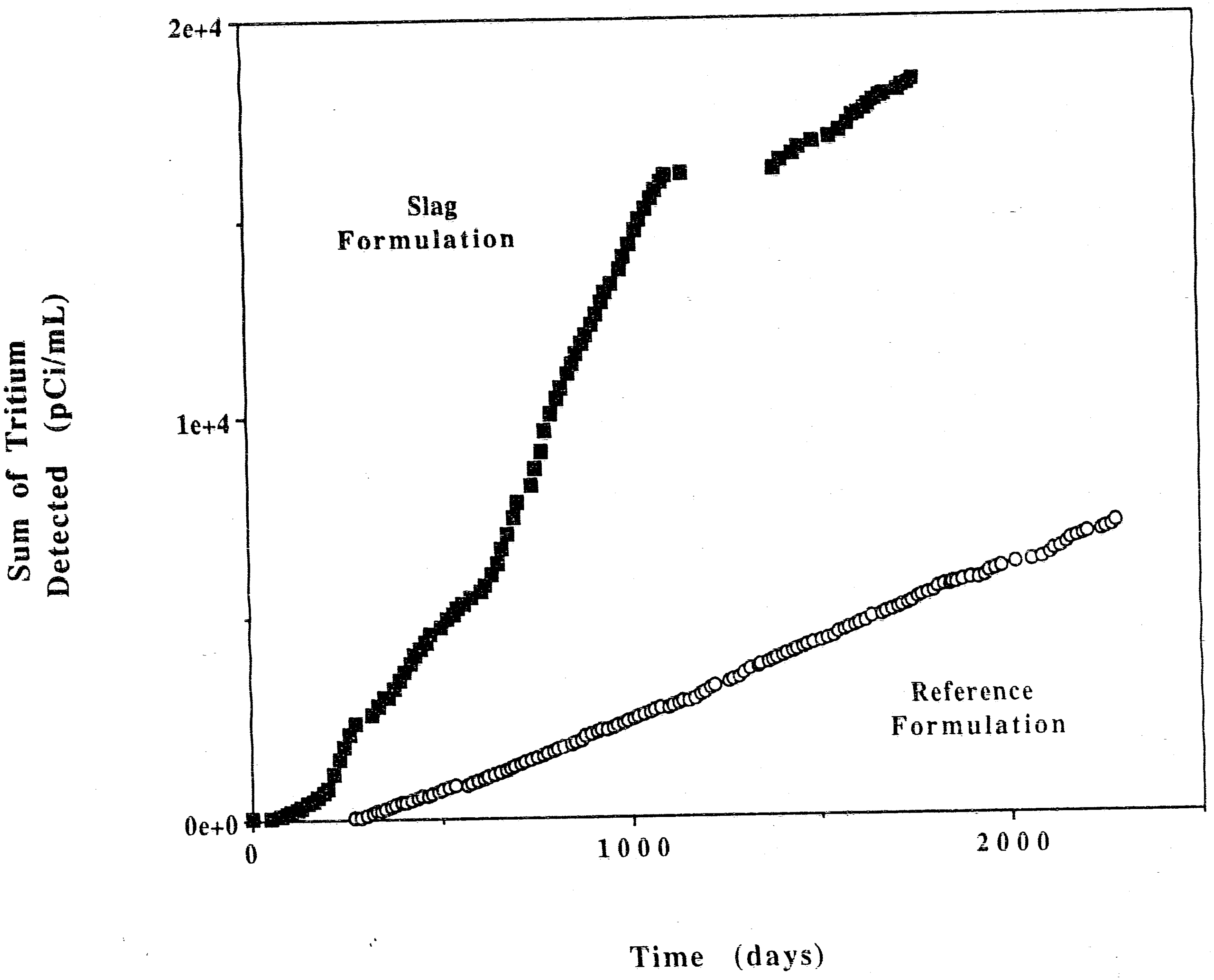

FIGURE 2.B.: The sum of tritium concentration detected in the sumps of the lysimeters with time.

FIGURE 3

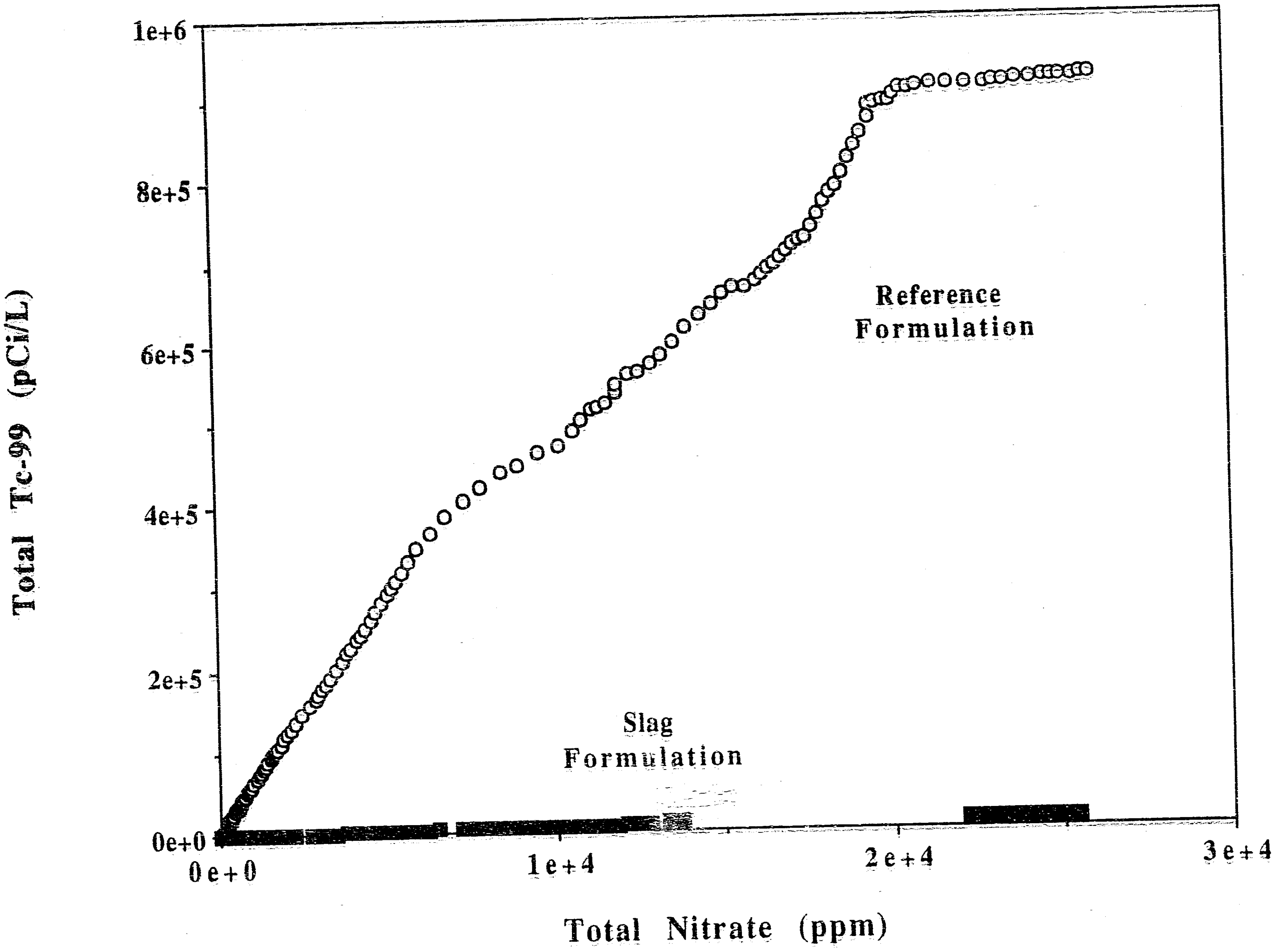

FIGURE 3: The sum of technetium-99 concentration detected in the sumps plotted as a function of the sum of nitrate detected in the sumps.

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