

THE EFFECT OF MICROBIAL ACTION ON NUCLEAR WASTE MANAGEMENT: IS
THERE ENHANCED LEACHING FROM BITUMEN AND INCREASED
RADIONUCLIDE MOVEMENT THROUGH GEOLOGIC MEDIA?

by

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Abstract

Long-term management of nuclear wastes demands absolute physical isolation of noxious radionuclides from the biosphere until decay to safe levels has occurred. Due to the extremely long half-life of some isotopes, the required isolation may be on the order of millennia.

Past research on radioactive wastes has centered on the physicochemical mechanisms that may effect a premature return of radionuclides to the environment. However, biological action in a radwaste disposal site may have two major effects: 1) physical destruction of the solidifying matrix through solubilization or oxidation; and/or 2) enhanced movement of radionuclides through (adsorbent) geologic media by production of various chelating agents. The work presented here is focused on both these microbiological processes.

^{60}Co and ^{137}Cs encapsulated in bitumen was allowed to undergo microbial attack by a selected hydrocarbonoclastic culture under idealized environmental conditions. The radionuclides released by this process were then evaluated for their ability to bind with selected geologic media. In order to compare the effect of reduced adsorption due to microbial action, synthetic chelating agents were used as a standard. The same hydrocarbonoclastic culture used for these experiments was also tested for its sensitivity to γ -irradiation.

Subsequent analysis showed microbial attack of bitumen did not enhance the release of the ions. However, a decreased adsorption to the geologic media was observed but the effect was much less than that observed for the synthetic chelating agents. The level of γ -radiation expected in the final waste repository will not effect the viability of the organisms tested.

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I. INTRODUCTION

1. BACKGROUND

Full scale atomic energy research in Canada has culminated in a pressurized heavy water reactor known as CANDU*. This reactor, one of Canada's great technical achievements, is one of three reactor designs on world commercial markets. The uniqueness of the reactor design is characterized by the use of 1) natural uranium; 2) deuterium oxide (heavy water) as moderator and coolant; 3) a multiple pressure tube configuration instead of a single large pressure vessel of other reactors; and 4) on power fueling (fuel bundle replacement during reactor operation). The commercial power reactor uses uranium with greater efficiency than the LWR** types. Because of its neutron economy the CANDU reactor also has the advantage of being adaptable to more efficient fuel cycles, such as the thorium fuel cycle without major

* CANada Deuterium Uranium.

**Light Water Ractor

modification of the existing design. This means it is possible to operate at or near breeding, making the CANDU reactor comparable to the fast breeder reactor. Thorium is at least three times as abundant in the earth's crust as uranium thus the available nuclear fuel resource would be considerably increased.

If this cycle is embraced, Canada will require a reprocessing facility in order to recover fissile materials contained in the spent fuels.¹ The separated fission products will require safe permanent disposal. The Canadian waste management program was initiated to evaluate disposal options. Prior to 1971 all irradiated fuel was either processed abroad or sold.² Since then, the spent fuel has been stored in water filled bays at the nuclear power stations. The fate of the spent fuels has not yet been decided, but either it will be reprocessed and the residues immobilized and disposed, or it will be disposed intact, most likely in mined cavities within granitic mineral free structures abundantly located in the Canadian Shield.

2. THE CHALK RIVER NUCLEAR LABORATORIES' LOW AND INTERMEDIATE LEVEL WASTE PROGRAM

Along with the high level wastes are less active wastes generated from daily operation of the nuclear power reactors. The broad categories including all low and intermediate level

radwastes encompass only 0.1% of the total "waste" radioactivity.³ Unfortunately, these wastes are substantial in volume and are chemically and radiologically heterogeneous. Some of the various differences with Canada's low and intermediate level wastes are: 1) the production of ^{14}C is much greater than in a LWR due to the much smaller LWR core size; 2) a high degree of system integrity minimizes losses containing fission and neutron activation products; and 3) tritium appears to be more abundant than in LWR's.⁴ Furthermore, the major sources of CANDU low and intermediate level wastes are associated with:

- 1) Routine operation and maintenance.
- 2) Purification in heavy and light water circuits (the majority of the radioisotopes are contained on spent ion-exchange resins and filters containing ^{60}Co , ^{137}Cs and ^{14}C (present as a carbonate on ion-exchange resins)).
- 3) Equipment decontamination.⁵

The ultimate disposal of radwastes will incorporate a multiple barrier system in which a waste nuclide would have to breach a series of obstacles⁶ prior to recontact with the biosphere. These obstacles include a number of pretreatment and treatment steps discussed below.

A. TREATMENT AND DISPOSAL OF LOW AND INTERMEDIATE LEVEL WASTES

i. Volume Reduction

All wastes that occur as incinerable solids will be incinerated to a stable ash. Both Ontario Hydro (Bruce Nuclear Power Development) and Atomic Energy of Canada Limited (AECL) at the Chalk River Nuclear Laboratory (CRNL) have incinerators operating for this purpose.

Liquid wastes may undergo a two-step procedure for volume reduction:

- 1) Reverse osmosis; and
- 2) Evaporation.

Reverse osmosis concentrates solids by excluding water through a semipermeable membrane. A second step in volume reduction may utilize a vertical thin-film evaporator⁶ to further increase the percentage of total solids (up to 50% has been achieved with some wastes).⁷

ii. Immobilization

The ultimate aim of immobilization of radwastes is the production of a durable leach-resistant solid. Compounds that have been used as a solidifying matrix include cement, urea-formaldehyde, polyester and bitumen.⁸ The selection of bitumen by CRNL's low and intermediate level waste program was due to its versatility, volume savings and leach resistance (highest

of those materials mentioned).⁹

Bitumen is chemically very heterogenous but its components may be grouped into four broad categories: saturated hydrocarbons, resins, cyclic hydrocarbons and asphaltenes.¹⁰ Other elements that may be present are oxygen(1-17%), sulfur(1-9%) and/or nitrogen(1%).¹¹ At room temperature(20-25°) bitumen's physical state may be described as a complex colloidal system.¹²

Bitumen has been used to cement building materials (Babylon), caulk boats or as a water stop between brick walls in the the third millenium B.C.¹³ The establishment of bitumen as a solidifying matrix for nuclear wastes may be attributed to the Research Centre for Nuclear Energy at Mol, Belgium and the Plutonium Research Centre of Marcoule, France. The Belgium establishment was constructed and operated (on a small scale) from 1960-1964 while the Marcoule installation started operation in 1965. Canadian experience with bitumen has been limited to laboratory and pilot scale projects. The completion of CRNL's Waste Treatment Centre (WTC) has been slated for 1982. This complex will utilize volume reduction by incineration or reverse osmosis and evaporation, followed by bitumenization.

iii. Ultimate Disposal

In order to ensure physical isolation of the radwaste from contact with the biosphere (until the waste radionuclides have decayed to acceptable limits) terminal disposal will be subterranean. The final waste repository will probably be located on the Canadian Shield in a geologically stable hard rock formation known as a pluton. Some of the obvious advantages for the selection of a pluton are that they are: "relatively homogeneous structures of high integrity and long stability" and "have remained undisturbed since early geologic times ie, for 200 to 2000 million years".¹⁴

iv. Natural Barriers

Although the utmost consideration will be given to siting the repository in a hydrogeologically inactive zone, groundwater intrusion may occur. If intrusion does occur the deleterious effect of the groundwater flow may be twofold: leaching of the solidified waste with concomitant movement through the repository and the subsurface environment. In order to minimize the passage of leached radionuclides out of the repository, naturally occurring adsorbents may be used to backfill the repository environment. This method is in accordance with the multiple-barrier approach to nuclear waste management. If a strong adsorbent ie, bentonite is used, leached nuclide attenuation will be greatly enhanced.

3. MICROBIAL ANTAGONISATION OF RADWASTE ISOLATION

Microbial activity may enhance the movement of radionuclides from a repository by effecting physical destruction of the solidified matrix or production of complexing agents that may decrease the effectiveness of sorption reactions by backfill material.

A. MICROBIAL ACTIVITY AND LEACHING

The release of waste radionuclides encapsulated in bitumen may be envisioned as a process involving two mechanisms:

1) Matrix decay caused by direct microbial attack of the bitumen; and 2) Matrix solubilization effected through the production of alcohols, esters, ketones and other metabolic end-products.¹⁵

Although the result of each mechanism is assumed to be negligible over a few years, the effect may have significant consequences for a waste that must remain isolated for centuries.¹⁶ The effect of direct microbial attack on bitumen has been documented by previous investigators.¹⁷⁻²³ However, investigations on microbial attack of nuclear wastes encapsulated in bitumen is virtually non-existent.²⁴ The research that has been done was performed under the auspices of the Los Alamos Scientific Laboratory.²⁵ Unfortunately, many of their findings do not have general applicability. The

experimental design and rationale focused on experiments that would yield information that pertained directly to the U.S. Waste Isolation Pilot Plant (WIPP). These studies included CO₂ gas evolution from (inactive) asphalt, microbial methylation of Pu, enumeration of WIPP microflora and general radiobiological studies. The geological environment of plutons and that expected for the WIPP (hard rock-salt) is dissimilar and therefore cross comparisons should only be made with caution.

B. MICROBIAL ACTIVITY AND SUBSURFACE NUCLIDE MOVEMENT

The ability of strong synthetic chelating agents to mitigate against radionuclide sorption to backfill material has been established elsewhere.²⁶ Emery²⁷, has shown that hydroxamate and polyhydroxamate chelating agents may be produced through microbial metabolism of organic materials. It follows logically, therefore, that microbially produced chelating agents may serve to enhance the movement of radionuclides through backfill material and the surrounding subsurface environment.

The fate and migratory properties of nuclear waste elements in a natural geological environment has been evaluated by numerous methodologies. However, evaluation of the partitioning of the waste element between the solid media and liquid phase (groundwater) is the focal point of most studies. Various researchers have tried to illustrate the

destiny of escaped radionuclides under a myriad of conditions.²⁸⁻³⁰

4. SCOPE OF THIS WORK

As mentioned earlier, microbial populations may have an adverse effect on nuclear waste management by direct attack of a bituminized waste package and/or production of chelating agents. The scope of this thesis will span these two issues. Unlike the Los Alamos work, this research should find general applicability throughout the nuclear industry. However, sundry details (choice of radionuclides, organisms and techniques) were chosen for their relevance to the Canadian nuclear program.

II. MICROBIAL DEGRADATION OF BITUMEN

1. BACKGROUND

Compared with glucose degradation, microbial oxidation of hydrocarbons poses a unique set of problems:

they are insoluble in water and present problems of how they are solubilized or emulsified; they are chemically unreactive so require specialized enzymes for their initial oxidation, and, finally they reverse the metabolism of microorganisms from being glycolytic and lipogenic to being lypolytic and gluconeogenic.³¹

Since metabolism of hydrocarbons is not as "energy-efficient" as degradation of common sugars, use of hydrocarbons as a substrate will only occur as a secondary mechanism.* As a result, adaptation and natural selection have evolved microorganisms with the ability to overcome or circumvent some of the constraints listed above. These adaptations have endowed organisms with the ability to:

* Hydrocarbons will only be degraded in those cases where other more suitable substrates are lacking or absent.

produce surface-active agents for the emulsification of their hydrocarbon substrate; oxidize their reactant (usually to an alcohol or diol) thereby making them more prone to an enzymatic catabolism similar to fatty acid metabolism; produce simple sugars from fatty acids or other lipoidal precursors. Although the high substrate specificity of most microbial species limits the overall degradation of a heterogeneous mixture of hydrocarbons, individual components may be attacked preferentially by a mixed culture of hydrocarbonoclastic bacteria. With respect to the major organic groups that compose bitumen, microbial degradation may occur in the following ways:

A. ALIPHATICS

According to Ratledge,³² the following characteristics apply to the degradation of aliphatic hydrocarbons:

1. Aliphatic hydrocarbons are assimilated by a wide variety of microorganisms. Other classes of compound, including aromatics, may be oxidised but are assimilated by only a few bacteria.
2. n-Alkanes of chain length shorter than n-nonane are not usually assimilated but may be oxidised. Only some bacteria have the ability to grow on alkanes shorter than n-octane. As the chain length of the alkane increases beyond C₉, the yield factor increases but the rate of oxidation decreases.
3. Saturated compounds are degraded more readily than unsaturated ones.
4. Branched-chain compounds are degraded less readily than straight chain compounds.

Although hydrocarbons of chain length <C₉, are more

soluble and therefore more available to microorganisms they seem to illicit a toxic response.³³ This toxicity may be attributed to a disruption of the cytoplasmic membrane with a concomitant loss in functional integrity.³⁴ In addition to toxicity from short hydrocarbons, fatty acids may also be noxious. The deleterious effect of fatty acids is evident from their amphoteric nature by means of which they may act as an emulsifier.³⁵

The first step in the metabolism of aliphatic hydrocarbons is usually to a primary alcohol by one of two possible enzymes (cytochrome P-450 or rubredoxin). To convert the primary alcohol to its corresponding carboxylic acid, a second oxidation usually follows. After this conversion to a carboxylic acid is complete, final degradation can occur via normal biochemical catabolic pathways such as β -oxidation.

Although the initial mode of oxidation changes for alkenes and branched-chain substrates, the resultant product is usually similar, namely, a terminal carboxylic group.

B. AROMATICS

Not unlike the decomposition of an aliphatic compound - metabolism of an aromatic species requires oxidation of the initial substrate to a common product. In the case of most simple benzene-like compounds, the common product is usually a catechol. Chapman³⁶ illustrates that 3 major products of an

initial oxidation sequence (catechol or 1,2-dihydroxybenzene, protocatechuic acid or 3,4-dihydroxybenzoic acid and gentistic acid or 2,5-dihydroxybenzoic acid) are "at the focal points of pathways for a wide range of compounds" and that "other substituted catechols or substituted parahydric phenols may serve as ring-fission substrates".³⁷

The subsequent reaction step that follows conversion to a catechol is ring-fission. For catechols the ortho-fission (cleavage between the two carbons containing the hydroxy groups) pathway predominates, while substituted catechols may undergo meta-fission (cleavage of the bond between an hydroxy-bearing carbon and a carbon adjacent to it that is not hydroxy-substituted).³⁸ Thus, following conversion to a catechol or a substituted catechol and ring-fission, central metabolic pathways function to cause complete oxidation.

C. POLYCYCLIC AROMATIC HYDROCARBONS

In comparison to degradation of the simple aromatic compounds, polycyclic aromatic hydrocarbons (PAH) are also dependent on conversion to a dihydrodiol before further metabolism may proceed. Although some genera (Aeromonas) appear to vary from this scheme through production of a 1-hydroxy-2-napthoic acid from phenanthrene, the general microbial attack seems to start with production of the diol. Again, dihydrodiol production is usually followed by ring-

fission and then total degradation via various central metabolic pathways.

The degradation of PAH's larger than 3 rings has not yet been unequivocally demonstrated. Although this may represent a lack of perseverance by experimenters in the field, the asphaltene fraction of bitumen may be resistant to microbial attack.

In conclusion, microbial degradation of hydrocarbons is dependent on conversion of the substrate to a more reactive intermediate such as a carboxylic acid or an alcohol, this product in turn is completely degraded by intracellular systems that have a more general function.

2. REPRESENTATIVE HYDROCARBONOCLASTIC BACTERIA

As mentioned earlier, the rate of microbial oxidation of hydrocarbons is usually extremely slow. Thus, to maximize the possible degradation of bitumen (containing waste radionuclides) fresh cultures of unidentified hydrocarbonoclastic microbes were obtained from:

- 1) Gemni Biochemical Research Limited - 1 mixed (Culture A) and 2 pure (Cultures B and C) cultures.
- 2) University of Calgary - 1 mixed culture (Culture D).*

* Thanks to Dr. Ian Forrester and Mr. Cam Wyndham of Gemni Biochemical Research and the University of Calgary respectively for their kind donation of these cultures.

All four cultures had been located in the bitumen-rich Athabasca tarsands and were therefore well adapted to the task of utilizing a heterogenous mixture of hydrocarbons as a metabolic substrate.

3. CONDITIONS FOR GROWTH AND PRELIMINARY IDENTIFICATION

A. GROWTH CONDITIONS

All cultures were grown on a mineral salts solution that had been found to be satisfactory for microbial degradation of hydrocarbons. As described by Bushnell and Haas,³⁹ the contents of this mixture are listed in Table I below:

Distilled, deionized water	1000.0 mls
MgSO ₄	0.2 g
CaCl ₂	0.02 g
KH ₂ PO ₄	1.0 g
K ₂ HPO ₄	1.0 g
(NH ₄) ₂ SO ₄	1.0 g
FeCl ₃	2 drops conc. soln.

Table I - MICROBIAL MINERAL SALTS SOLUTION

To prevent precipitation of various inorganic species after sterilization (>20 minutes at 125°C), FeCl₃ and CaCl₂ were autoclaved separately and added after the main solution had cooled.

In addition to the salts listed above, non-selective nutrients including 0.3 wt.% malt extract, 0.3 wt.% yeast extract and 0.5 wt.% peptone were added to the mineral salts

solution on the recommendation of Forrester.⁴⁰ Temperature was maintained at close to ambient (20-25°C) and aeration was maximized through continuous agitation of all cultures.

B. IDENTIFICATION

Cultures A, C and D contained gram negative rod-shaped cells that showed some motility at 30°C. An Ontario Ministry of The Environment analysis⁴¹ (see Appendix A) report showed the 4 cultures may contain the following genera:

- | | | |
|-----------|---|---|
| Culture A | - | <u>Pseudomonas</u> |
| Culture B | - | <u>Bacillus</u> |
| Culture C | - | VE group* |
| Culture D | - | <u>Citrobacter</u> , <u>Pseudomonas</u> |

C. GROWTH CURVES

In conjunction with the Ontario Ministry of The Environment analysis (Appendix A), microbial growth profiles (Figures 1, 3, 5 and 7) were established by the following methods: 0.30 ml of fresh inoculum was added to 20 mls of the nutrient media salt solution (described earlier) and allowed to rotate (5-10 RPM) at 25°C. Samples were taken from the reaction vessel every hour for at least 11 hours. Serial

* Bacteria in the VE group share characteristics with the genera Pseudomonas, Xanthomonas and Chromobacterium and as yet are not well defined taxonomically.⁴²

dilutions followed by media plating was used to determine the number of viable cells per sample.⁴³ Standard curves (Figures 2,4,6 and 8) of absorbance vs cell concentration, were also obtained for each culture by taking absorbance readings of 0.5 to 1.0 ml aliquots of each hourly sample on a Gilford 240 spectrophotometer (wavelength=600nm).

These plots (Figures 1-8) illustrate that Culture D showed the most rapid initial growth. A second interesting observation of the culture D profile is its biphasic nature. This information coupled with that in Appendix A, confirms the multi-organism content of Culture D.

4. SELECTION OF A SUITABLE CULTURE AND GROUNDWATER

A. SELECTION OF A SUITABLE CULTURE

In order to isolate the mixed culture with the greatest bitumen degrading potential, the following procedure was followed: 1.0 g of bitumen(30-40 mesh of Sp-170)* was placed in 20.0 mls of deionized water (DIW) containing 0.1340 g Yeast Nitrogen Base (YNB)** and 0.1 wt.% peptone. This solution was then sterilized by γ -irradiation (500 Krads). Two identical

* "Sp-170" is the designation used by Husky Oil (the bitumen supplier) for this class of oxidized bitumen.

**YNB is a non-selective source of non-carbon nutrients for microbial growth, including the following: $(\text{NH}_4)_2\text{SO}_4$ (75 wt.%), KH_2PO_4 (15 wt.%), MgSO_4 (7 wt.%), NaCl (1 wt.%), CaCl_2 (1 wt.%) and selected vitamins and nutrients(<1 wt.%)

CULTURE A 11 HOUR GROWTH CURVE

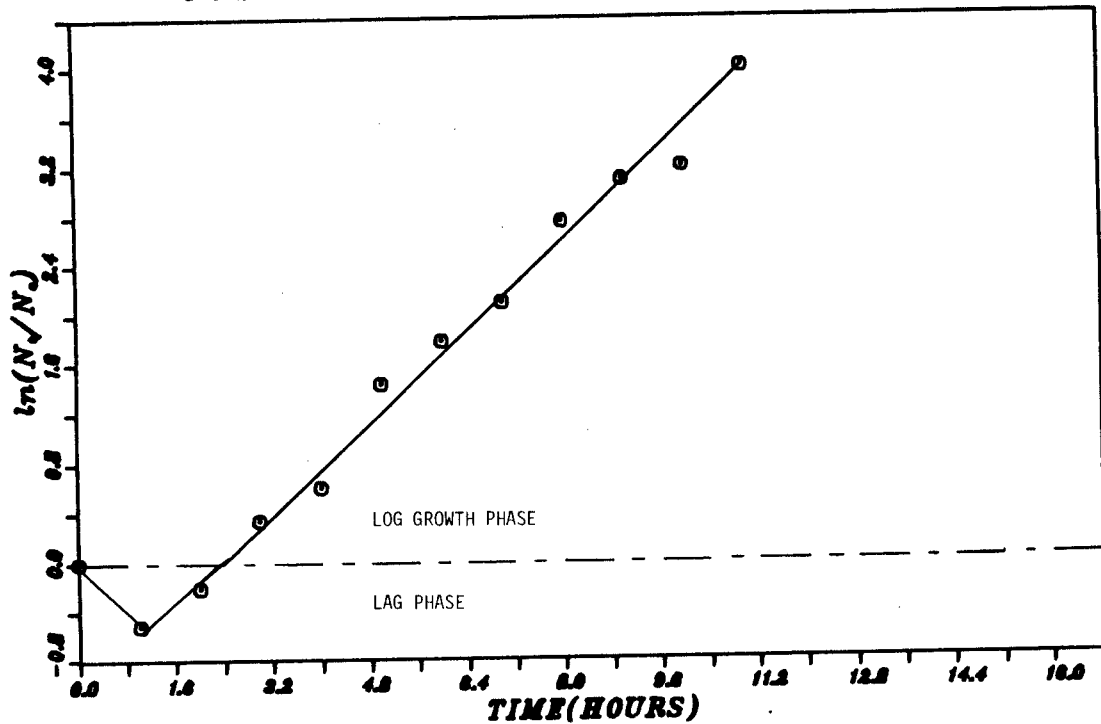


Figure 1 - Culture A - 11 Hour Growth Curve

CULTURE A STANDARD CURVE-(ABS. VS CELL CONC.)

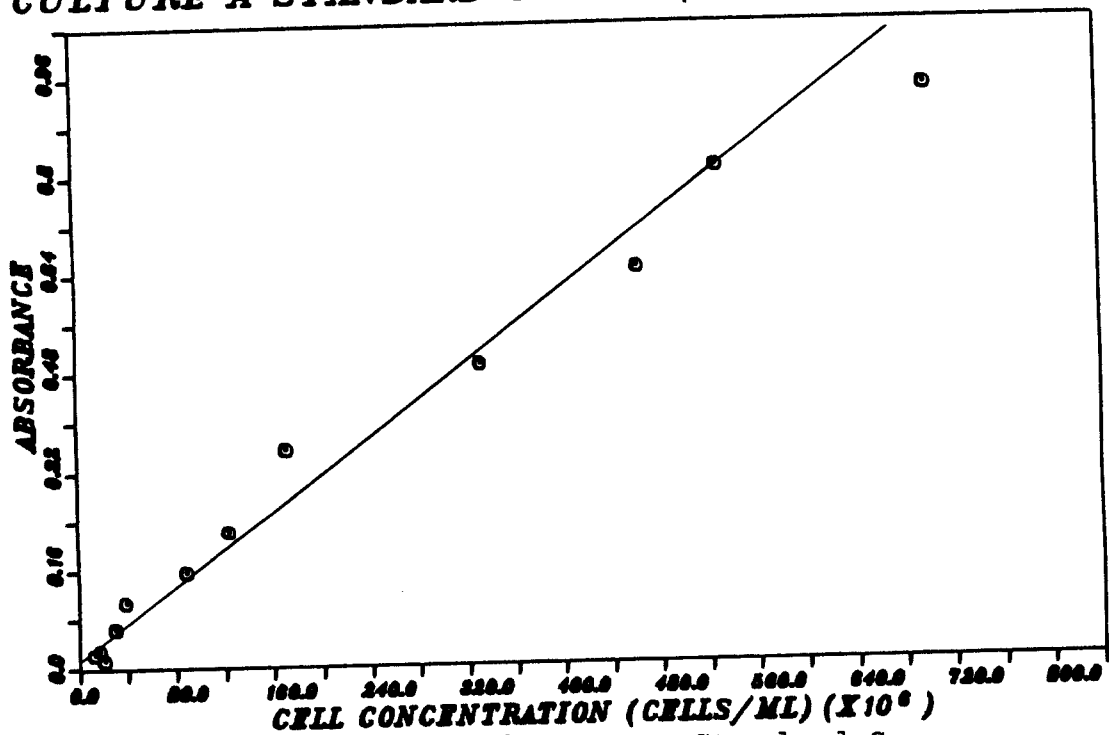


Figure 2 - Culture A - Standard Curve

CULTURE B 12 HOUR GROWTH CURVE

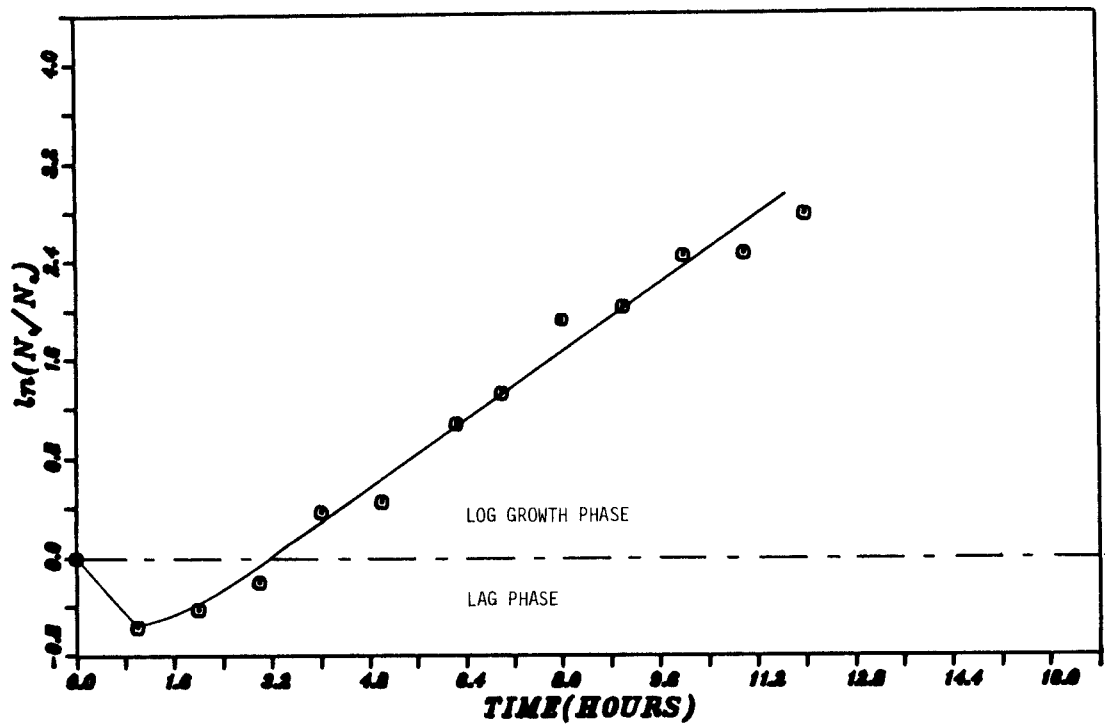


Figure 3 - Culture B - 12 Hour Growth Curve

CULTURE B STANDARD CURVE-(ABS. VS CELL CONC.)

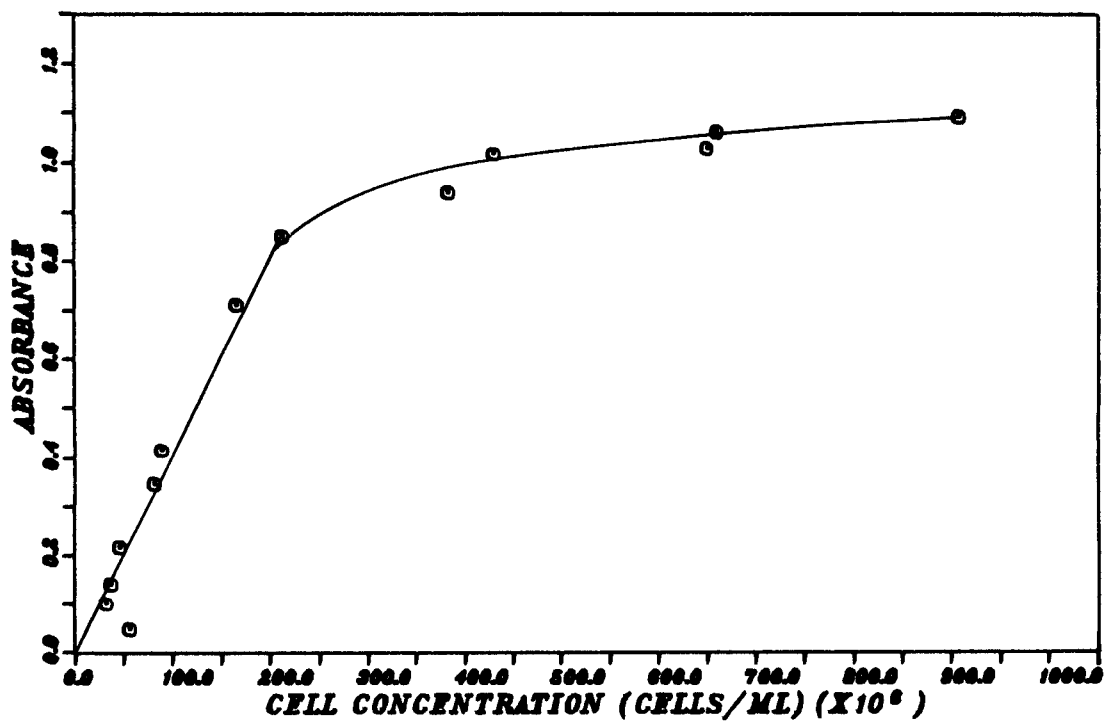


Figure 4 - Culture B - Standard Curve

CULTURE C 12 HOUR GROWTH CURVE

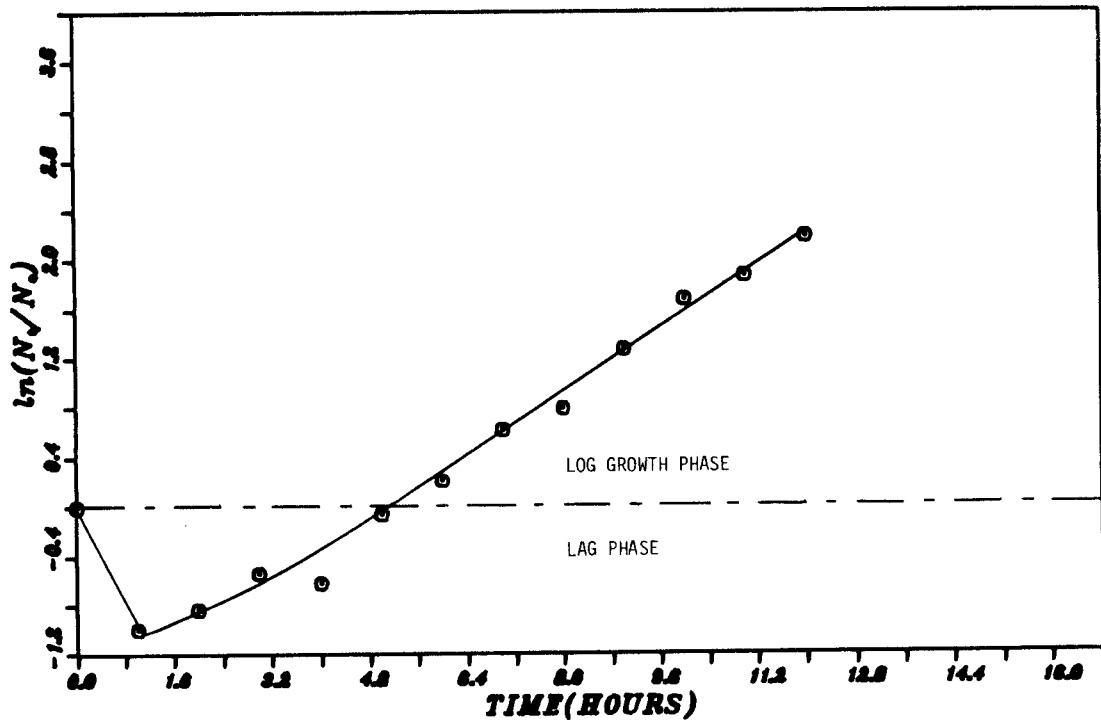


Figure 5 - Culture C - 12 Hour Growth Curve

CULTURE C STANDARD CURVE-(ABS. VS CELL CONC.)

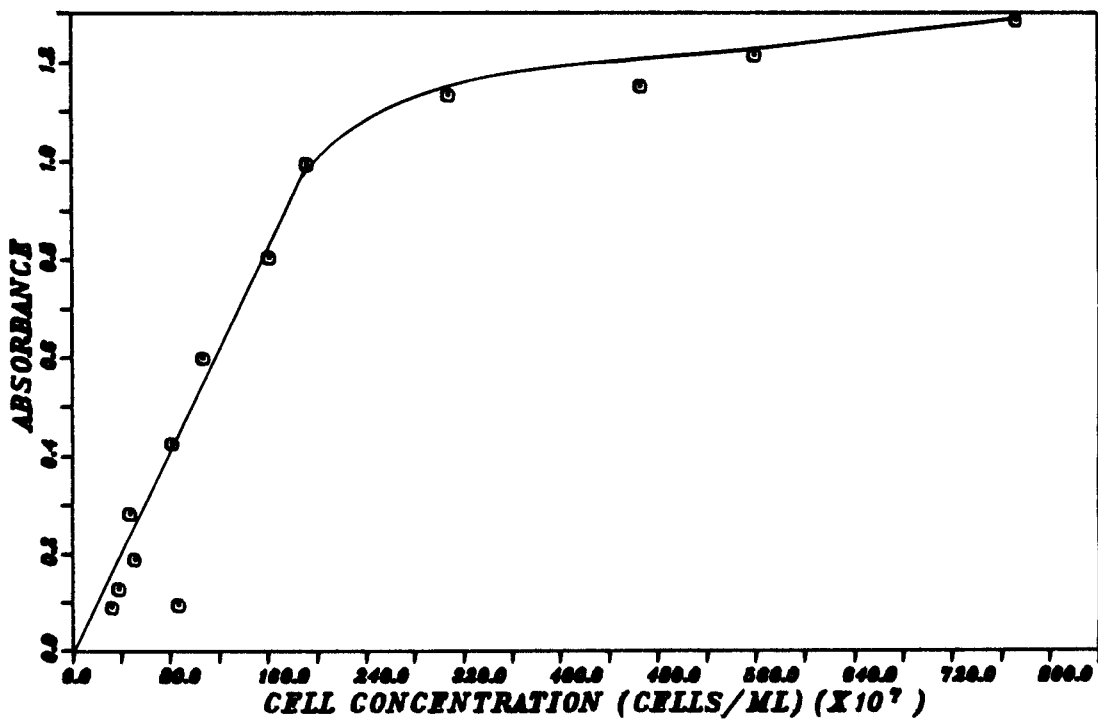


Figure 6 - Culture C - Standard Curve

CULTURE D 12 HOUR GROWTH CURVE

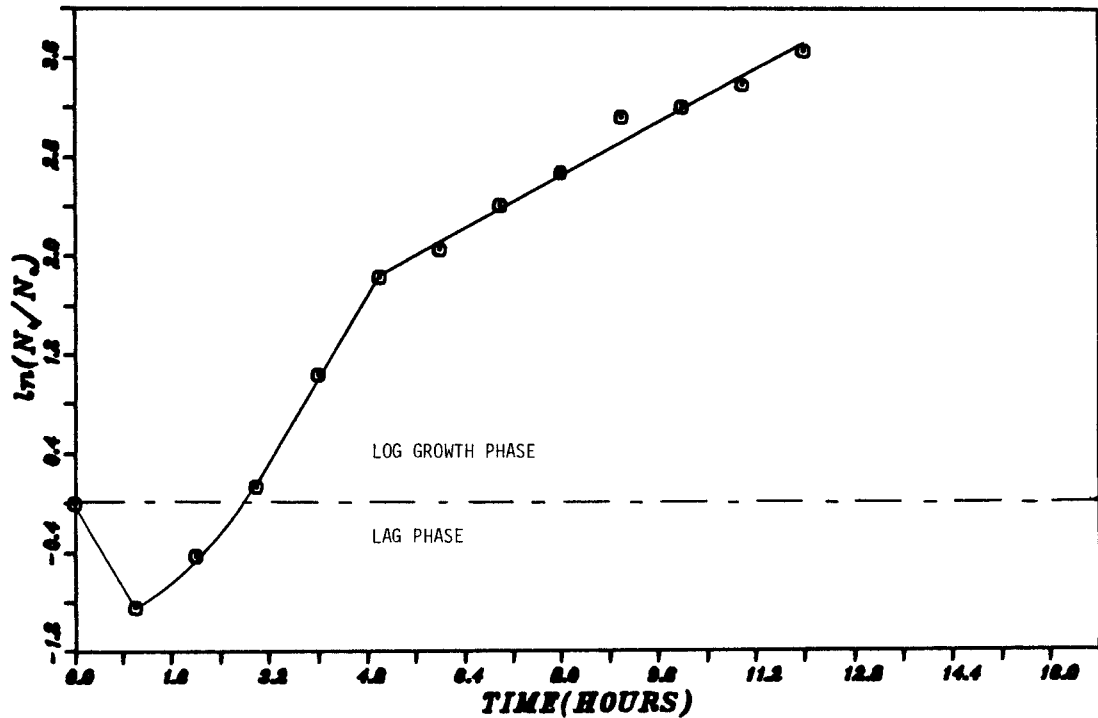


Figure 7 - Culture D - 12 Hour Growth Curve

CULTURE D STANDARD CURVE-(ABS. VS CELL CONC.)

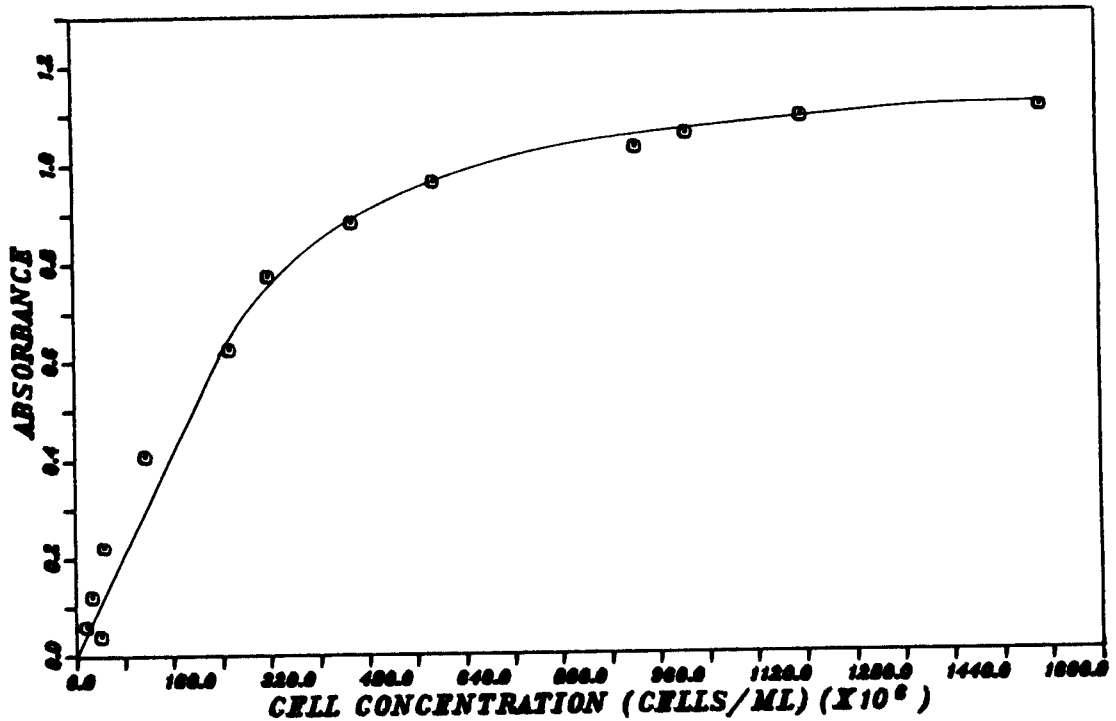


Figure 8 - Culture D - Standard Curve

solutions were prepared following this procedure. To confirm that bitumen was the sole carbon source present in the test vessels, appropriate controls (sets a and b) were run. Also, to directly compare rate of growth on bitumen to rate of growth on a common carbon source a second set of controls were run (e and f) consisting of sucrose substituted for bitumen.

Thus, six individual sets were run consisting of:

- a) YNB + Culture A
- b) YNB + Culture D
- c) YNB + Bitumen(1 g) + Culture A
- d) YNB + Bitumen(1 g) + Culture D
- e) YNB + Sucrose(1 g) + Culture A
- f) YNB + Sucrose(1 g) + Culture D

At T=46 hours, growth was found only for those sets containing Culture D and similarly, the largest population sizes were only found for sets d and f.

B. SELECTION OF A SUITABLE GROUNDWATER SOLUTION

Current work by the Applied Geoscience Branch of the Whiteshell Nuclear Research Establishment has established four synthetic groundwaters as appropriate for radionuclide adsorption studies.⁴⁴ In order to simulate "real" repository conditions for subsequent experiments (Sections IV and V), selection of a synthetic groundwater that would not suppress microbial growth was necessary.

The four synthetic groundwater solutions were prepared according to the methods outlined in Appendix B. Growth of Culture D was tested through simple inoculation of each

groundwater solution containing 0.3 wt.% malt extract, 0.3 wt.% yeast extract and 0.5 wt.% peptone but no additional mineral salts other than those contained by the groundwater. After incubation at 25°C the solution containing WN-1 Saline Solution supported the best growth of Culture D.

REMARKS

Since the degradation of bitumen is extremely slow, all foreseeable inhibitors of microbial activity were eliminated (Section IV). On the basis of the rudimentary evaluations of growth mentioned above, Culture D and WN-1 Saline Solution were selected as the most appropriate culture and groundwater solution respectively, to provide a "worst case" approach.

III. MICROBIAL RADIOSENSITIVITY

Since the final waste repository will be a continuous source of low-level radiation (<10 R/hr), the effect of radiation on Cultures A,B,C and D has been evaluated.

1. BACKGROUND

The majority of low-level waste radionuclides that may be encapsulated in bitumen emit γ -radiation (ie ^{137}Cs , ^{60}Co etc.). The absorbed dose from these radionuclides has been cited as a potential area of concern with respect to radiolytic degradation of asphalt with concomitant gas generation.^{4 5} However, these same fields could be bacteriocidal; the relative resistivity of Culture A,B,C and D to low LET* radiation was evaluated through irradiation of each culture with a high energy (1330.0 KeV) γ -source.

In order to graphically illustrate the lethal effects on

* LET or Linear Energy Transfer encompasses that fraction of the inherent energy associated with the radiation that is transferred to the target atoms - energy transmitted to the absorber per unit path length.

a population of each culture, percentage survival was determined over the range of 0-300 Krads. The results of this experiment will yield information that is important in the consideration of whether or not irradiation will effectively decrease or eliminate the rate of bitumen biodegradation.

2. METHODS

A. GROWTH MEDIA PREPARATION

Six litres of mineral salts solution were prepared as described earlier (Section II), with the exception of CaCl_2 . To prevent the precipitation of inorganic salts (during sterilization) a CaCl_2 solution of 0.02 g/ml was prepared separately. Peptone, yeast and malt extract were added according to Forrester, accompanied by 2 wt.% agar. These two solutions (medium and CaCl_2) were then sterilized separately and mixed upon termination of sterilization. Approximately 300 plates were prepared from this mixture and allowed to cool under intense UV irradiation.

B. CELL PREPARATION

A fresh solution of cells was prepared from each culture and allowed to incubate for approximately 72 hours. Due to this long incubation period all cell solutions were assumed to be in a stationary growth phase.

Cell washing was done three times by adding 1 ml of each fresh culture to Nalgene centrifuge tubes followed by 9-10 mls of PBS.* Each culture was then centrifuged at 9000 RPM for 5-10 minutes at 2°C. The supernatant was discarded and the pellet resuspended in 9-10 mls of PBS.

C. IRRADIATION AND ENUMERATION

Following washing, the pellets were vigorously agitated in PBS to yield a homogeneous cell solution. Immediately prior to and following each irradiation interval each culture was placed in ice to decrease the effect of enzymatically catalyzed repair. Irradiation was performed with an AECL Gammacell-220 with a dead time** and dose rate of 2.4024 Krad and 0.4640 Krad/sec*** respectively.⁴⁶ On the basis of this dead time and dose rate the cultures were irradiated directly in the centrifuge tubes according to Table II.

* PBS- Phosphate Buffer Solution is a mixture of K_2HPO_4 and KH_2PO_4 present in an appropriate molar ratio to yield a buffered pH of 7.50.

** Dead time is the residual radiation received by the sample prior to and upon termination of the dosing period.

***Dose rate and dead time are determined from the measured source (^{60}Co) activity at some reference time, $t=0$. Thus, doses and dead time at time t can be calculated from the original activity, reference time and source half-life.

<u>Dose(Krad)</u>	<u>Sequential Time(sec)</u>	<u>Cumulative Time(sec)</u>
0	0.0	0.0
5	5.6	5.6
10	5.6	11.2
15	5.6	16.8
25	16.4	33.2
50	48.7	81.9
75	48.7	130.6
100	48.7	179.3
200	210.3	389.6
300	210.3	600.2

Table II - γ -IRRADIATION TIMES USED FOR CULTURES A,B,C AND D

Upon receipt of the appropriate dose, 0.5 mls of the culture was removed and serial dilutions of 10^{-1} , 10^{-2} , 10^{-3} , 10^{-4} and 10^{-5} were made. Replicate plates were prepared for each of the first four dilutions and at each dose listed in Table II. A fifth set of replicate plates was prepared for the 0 dose control. Since irradiation of the cultures was performed sequentially, 0.5 ml aliquots of the sample were diluted and plated after receipt of the appropriate dose. All plates were then allowed to incubate for 48 hours at 20°C prior to colony enumeration.

3. RESULTS

Dose-response curves (Figures 9-12) were generated for cultures A,B,C and D. These curves illustrate that none of the cultures are capable of growth at a dose >75 Krads. At least half the population will die (LD_{50}) after exposure to a dose exceeding 6 Krad. These results indicate that none of

the cultures tested are radiosensitive (substantial cell death is only realized at >1000 rads) however, Culture A,C and D all showed survival curves characteristic of a Multitarget* mechanism for cellular inactivation.⁴⁷ This result may be expected since many bacterial species show this response to γ -irradiation.

In order to minimize the exposure to personnel and prevent severe radiolytic damage to the bitumen, the initial surface dose rate will be kept below 10 R/hr.⁴⁸ Except for ^{14}C , ^{137}Cs and ^{60}Co will be the most predominant radionuclides contained in the low level waste repository.⁴⁹ These waste nuclides have a half-life of 5730, 30 and 5.3 years respectively. Thus, with an exponential decay, activity contributed by ^{137}Cs and ^{60}Co will have decreased to negligible levels within 300 years.

Since each culture tested is relatively radio-insensitive and the bitumen waste block will contain predominately short-lived radionuclides with a low initial activity - the radiological effect of the waste on these microbial species would be negligible. However, long-term exposure may increase the rate of mutation and/or increase the relative radiosensitivity of exposed organisms.⁵⁰

* Multitarget theory assumes an organism death will result only after various intracellular "targets" are inactivated by the radiation source.

RADIATION DOSE RESPONSE PROFILE OF CULTURE A

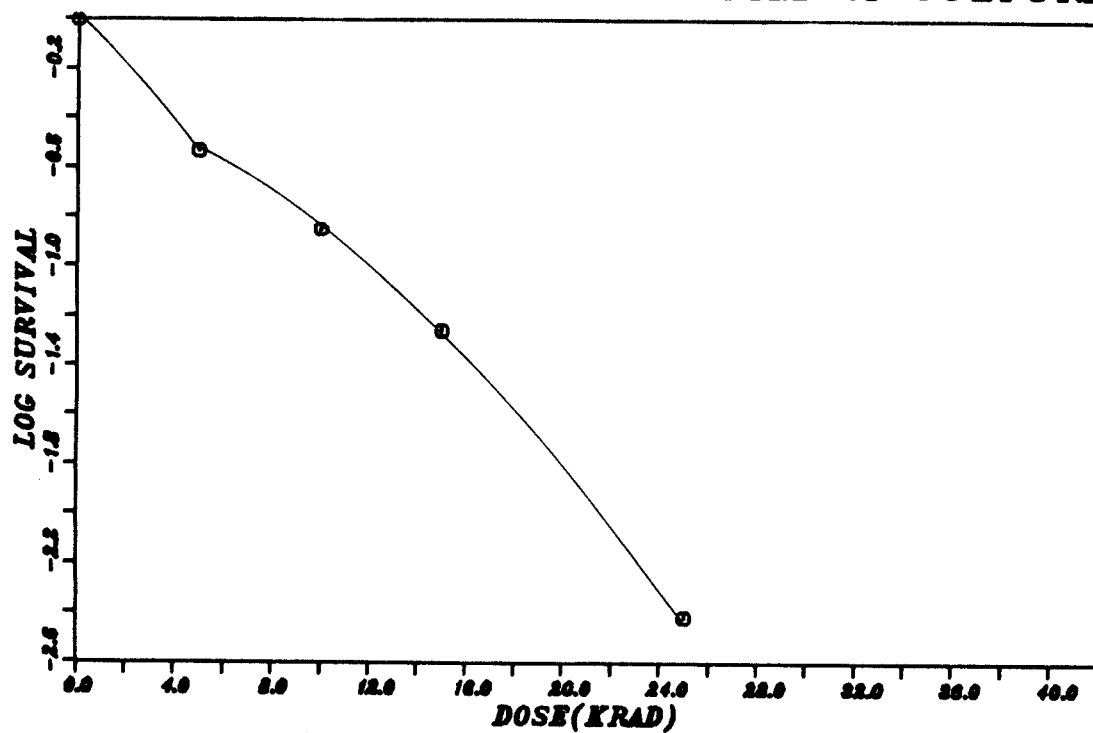


Figure 9 - Dose-Response of Culture A

RADIATION DOSE RESPONSE PROFILE OF CULTURE B

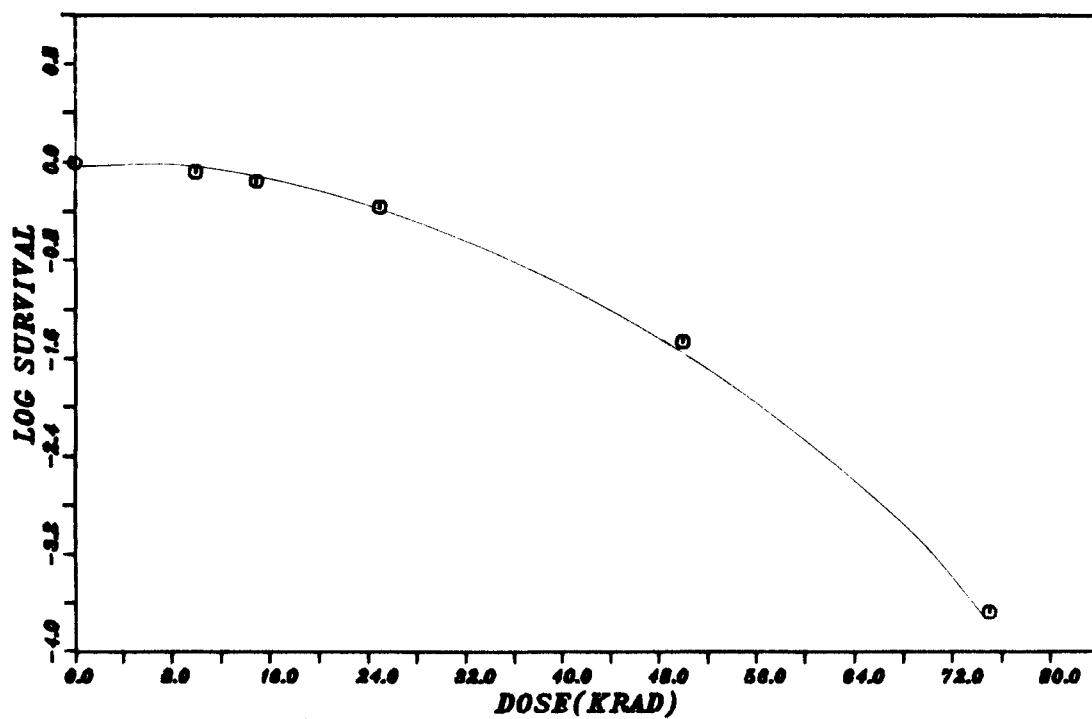


Figure 10 - Dose-Response of Culture B

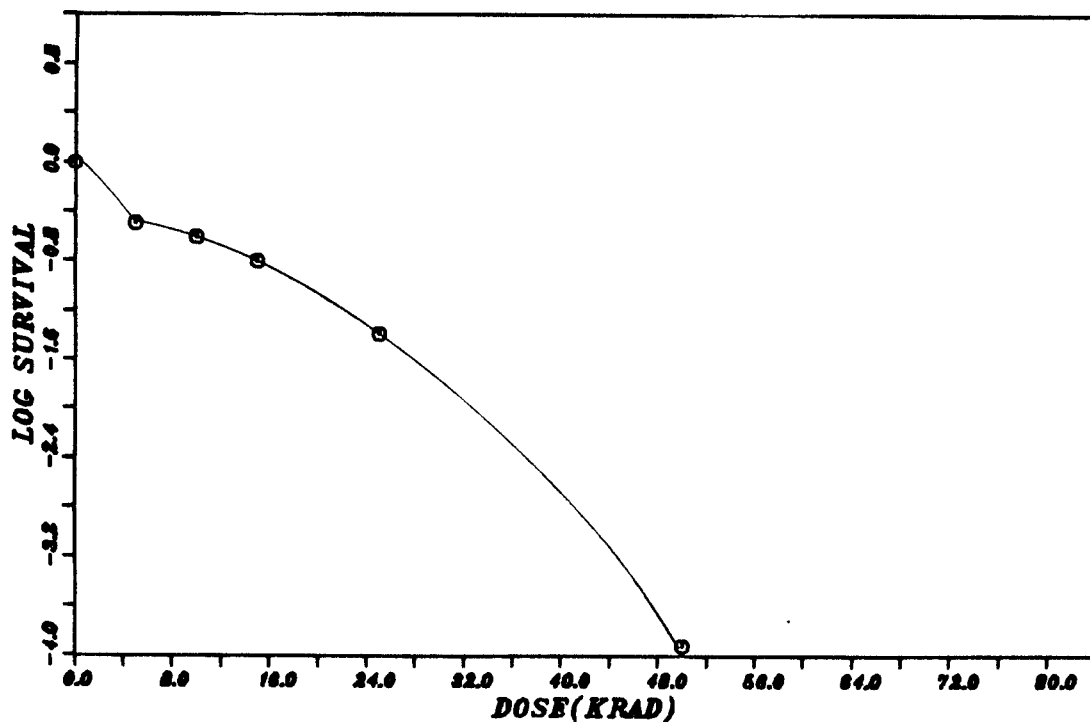
RADIATION DOSE RESPONSE PROFILE OF CULTURE C

Figure 11 - Dose-Response of Culture C

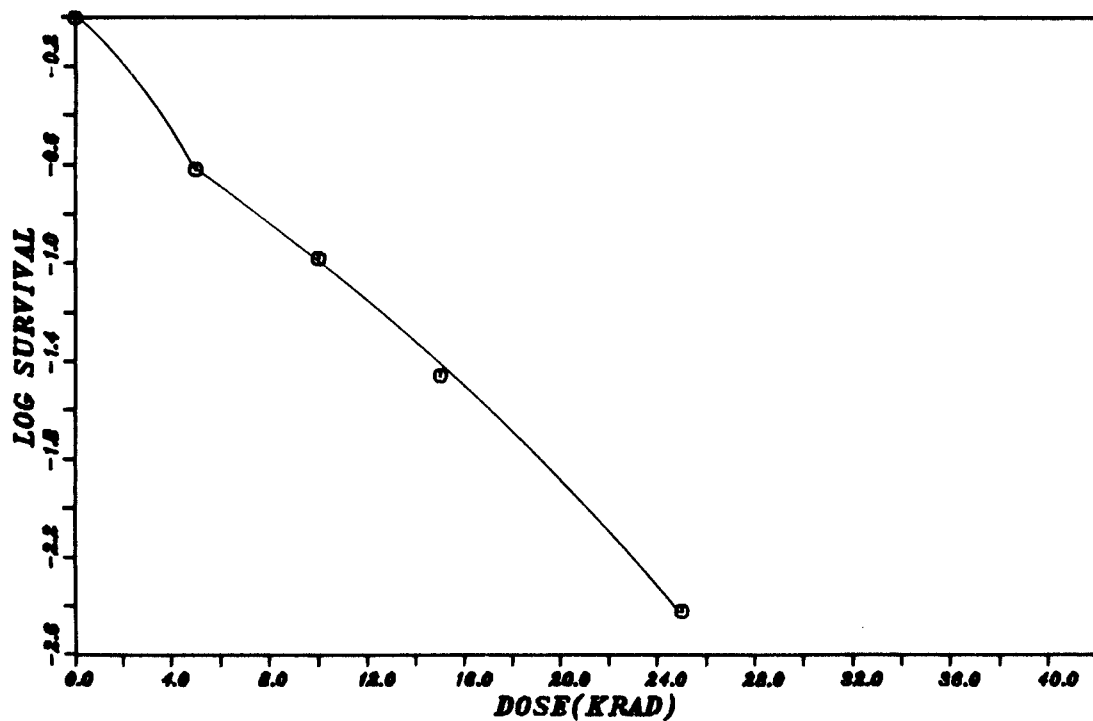
RADIATION DOSE RESPONSE PROFILE OF CULTURE D

Figure 12 - Dose-Response of Culture D

IV. EVALUATION OF MICROBially ENHANCED LEACHING

Leaching, the process by which a relatively insoluble species such as an inorganic salt is solubilized, is the primary mechanism for release of encapsulated waste radionuclides. The process of leaching immobilized wastes has been shown^{51, 52} to be approximated by Fick's diffusion equations,^{53, 54} and a plot of:

$$\frac{\sum a_n}{A_0} \frac{V}{F} \quad \text{vs} \quad (t_n)^{0.5} \quad \text{Eq. IV.1}$$

will show linearity if the process is governed by diffusion.* If the cumulative fraction of activity leached is plotted against time the resultant curve (for a diffusion-mediated response) may be described as parabolic initially then tapering off to a straight line with slope ~ 0 . This type of plot was used to illustrate the data presented in this Section. However, diffusion (and therefore leaching) will not

* Where a_n =radioactivity leached during the leachant renewal period, n ; A_0 =radioactivity initially present in specimen; F =exposed surface area of specimen (cm^2); V =specimen volume (cm^3) and t_n =duration (days) of leachant renewal period.⁵⁴

occur unless the solidified wastes are in contact with the leachant. An increase in any of the mechanisms that will allow greater availability of the encapsulated species with the leachant will therefore result in an increase in leaching.

1. ENHANCED LEACHING

The pertinent factors with respect to bitumen that may result in enhanced leaching are:

- a) Mechanical abrasion,
- b) Temperature,
- c) Diffusion,
- d) Biodegradation.

Since the final waste repository will be located in a geologically stable location, the effect of mechanical abrasion will elicit a negligible increase in contact of the immobilized waste radionuclides with native groundwaters. Unfortunately, the temperature characteristics of the repository are unknown. Initially the repository temperature is expected to be approximately 10°C however, waste heat from high-level radioactive decay may increase the overall repository temperature. If the increase is large enough, leaching may be enhanced as a result of the increased surface contact.⁵⁵ The remaining two factors may result in the greatest increase in leaching under "repository conditions" and therefore deserve greater elaboration.

Diffusion

The enhanced movement of ions from bitumen as the concentration gradient is decreased between the bitumen and solvent can be rationalized by a diffusion mediated phenomenon.^{5 1} The radionuclide - bitumen mixture will be essentially homogeneous thus, the waste solid may be considered an isotropic medium. In this case, a random uniform movement of ions between the bitumen surface and the surrounding media will occur. If the surrounding media has a low initial ionic concentration (ie DIW), a net increase in the ion content will be observed after contact with the bitumen block. The driving force for this steady-state movement is the difference in concentration between the bitumen and water, ΔC . As ΔC approaches 0, the net exchange between the isotropic solid and leachant will also become 0.

Mathematically, diffusion may be described as:

the rate of transfer of diffusing substance through unit area of a section is proportional to the concentration gradient measured normal to the section, ie

$$F = -D \frac{\Delta C}{\Delta x}$$

Eqt.IV.2

where F is the rate of transfer per unit area of section, C the concentration of diffusing substance, x the space coordinate measured normal to the section, and D is called the diffusion coefficient.^{5 6}

The resultant effect of this process will be an equilibrium state in which the concentration of ions in the

leachant approximates the concentration of ions in the bitumen. Unfortunately, equilibrium will only be attained in those cases in which the leachant is static. Groundwater flow in a "real" repository would represent a dynamic state that may afford infinite dilution provided a concentration gradient exists between the solid matrix and groundwater. This behaviour is clearly illustrated by Figures 13 to 26 in which the cumulative fraction leached (as a function of time) is still increasing for all cases in which an ionic counterbalance was not present (Set-A). Sets B,C, and D* however, all illustrate the case in which an equilibrium is being approached and slope $\rightarrow 0$, due to the mitigating effects of ions present in solution prior to the initiation of the test.

Biodegradation

As previously mentioned both laboratory¹⁷⁻¹⁹ and in situ^{21-23, 57-60} attack has been adequately demonstrated. The effect of microbial attack of a bituminized waste may enhance leaching via: physical removal of the exposed layer of the bitumen as it is used as a microbial substrate; solubilization by metabolic intermediates, end-products¹⁵ or co-oxidative products⁶¹; or emulsification of the bitumen by microbially

* Set A contained DIW only. However, B,C and D were composed primarily of the nutrient solution discussed in Section II.

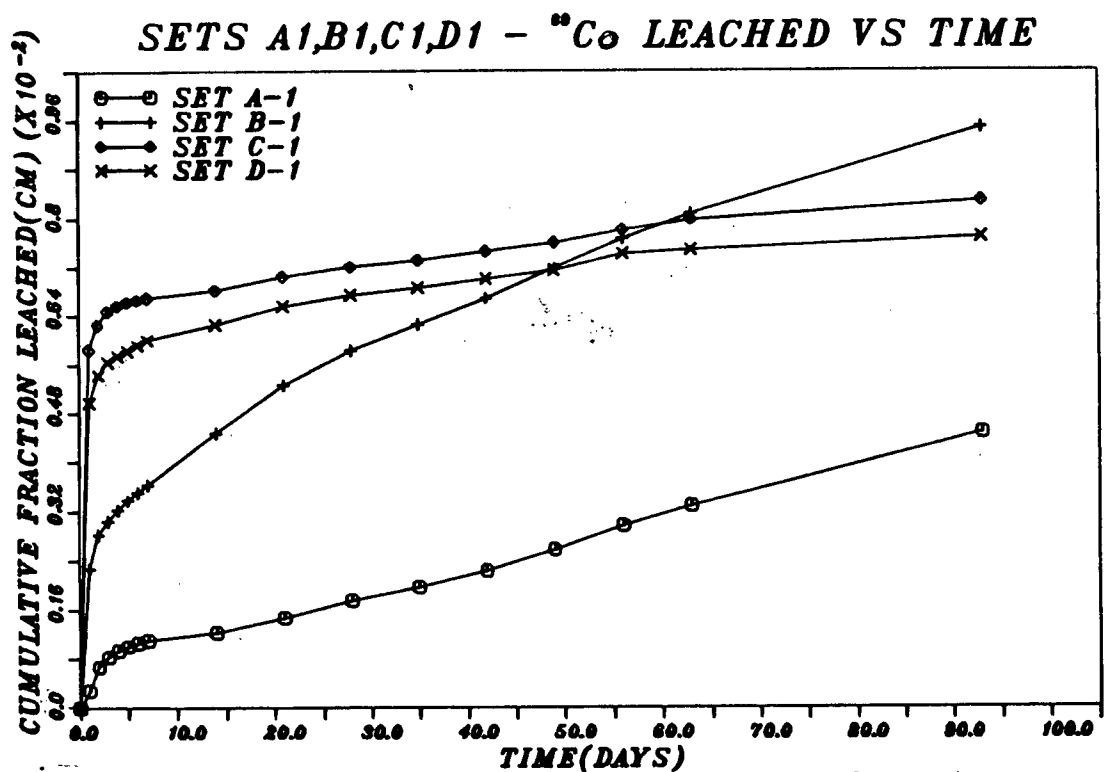


Figure 13 - Leaching profiles for sets A1,B1,C1 and D1 - ^{60}Co

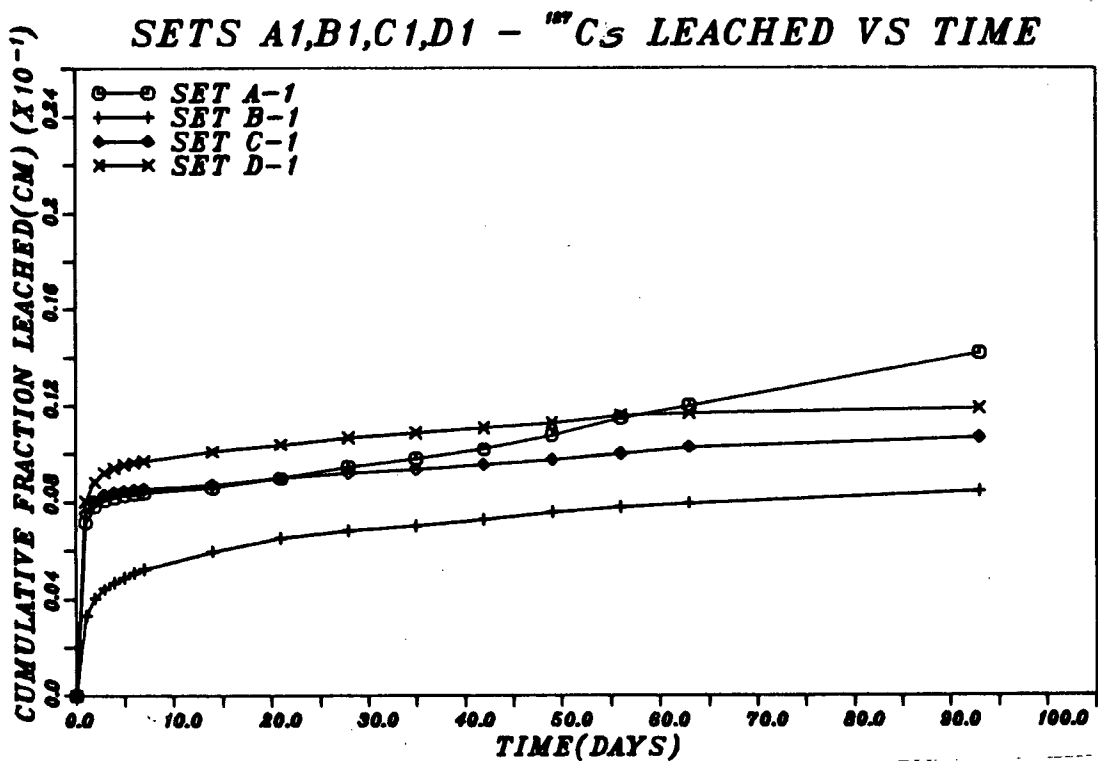


Figure 14 - Leaching profiles for sets A1,B1,C1 and D1 - ^{137}Cs

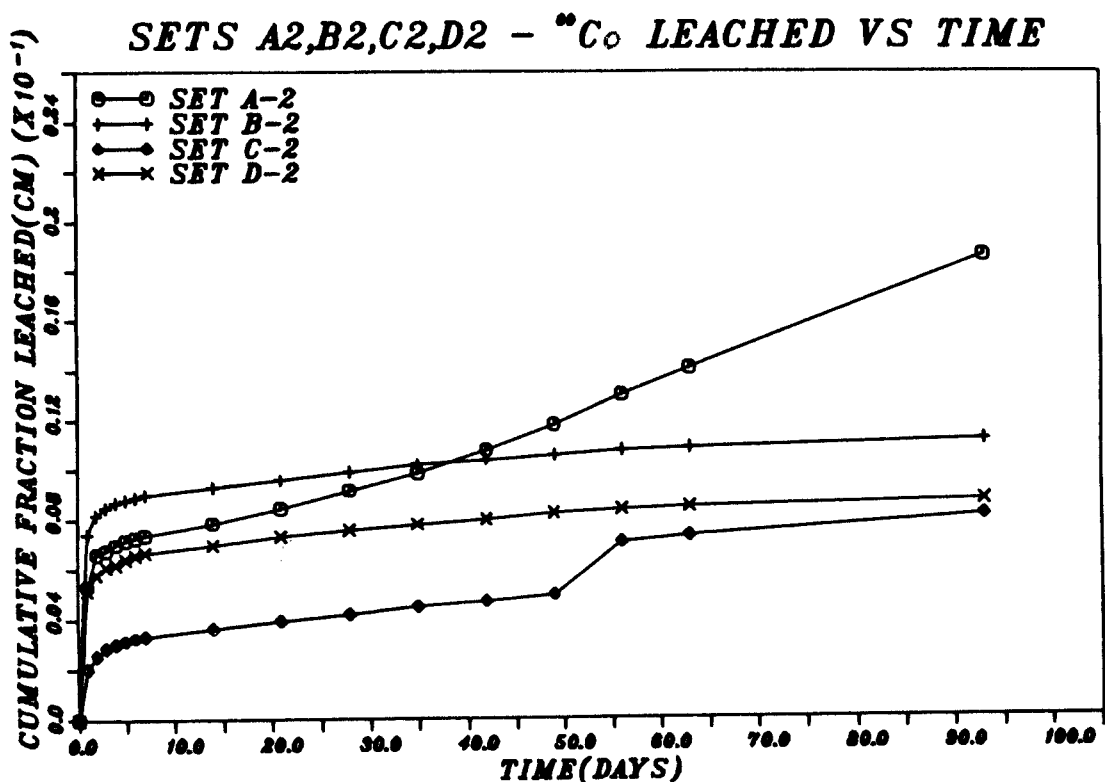


Figure 15 - Leaching profiles for sets A2,B2,C2 and D2 - ^{60}Co

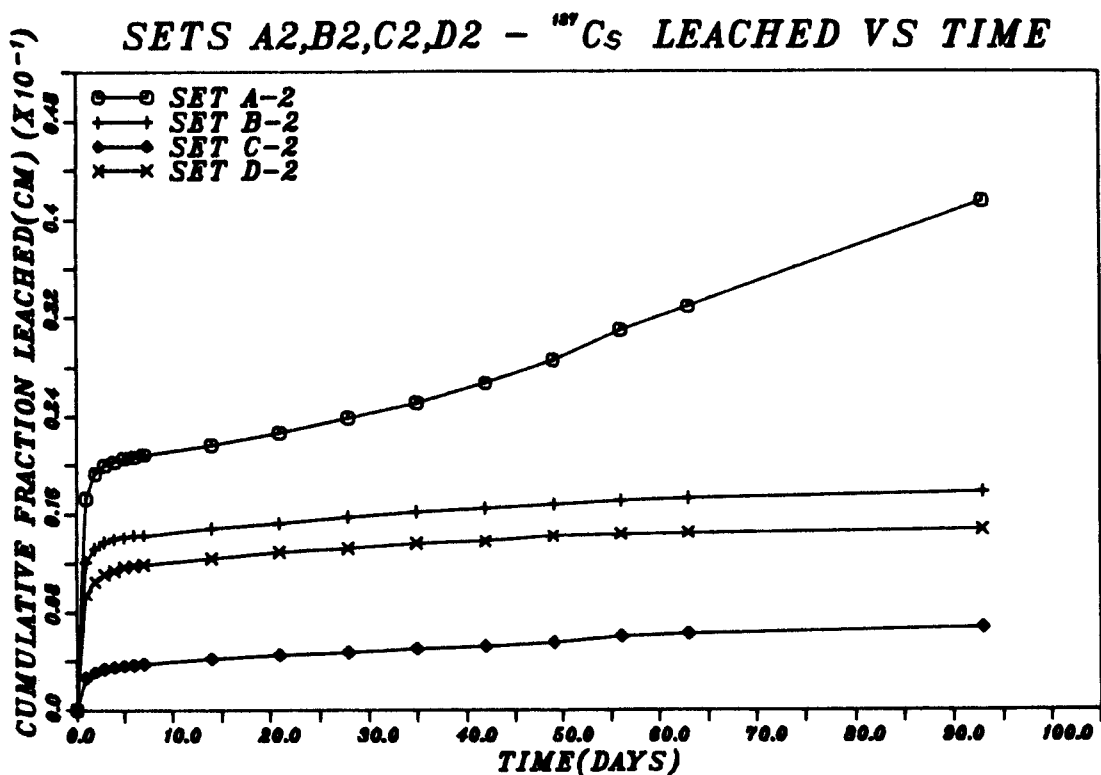


Figure 16 - Leaching profiles for sets A2,B2,C2 and D2 - ^{137}Cs

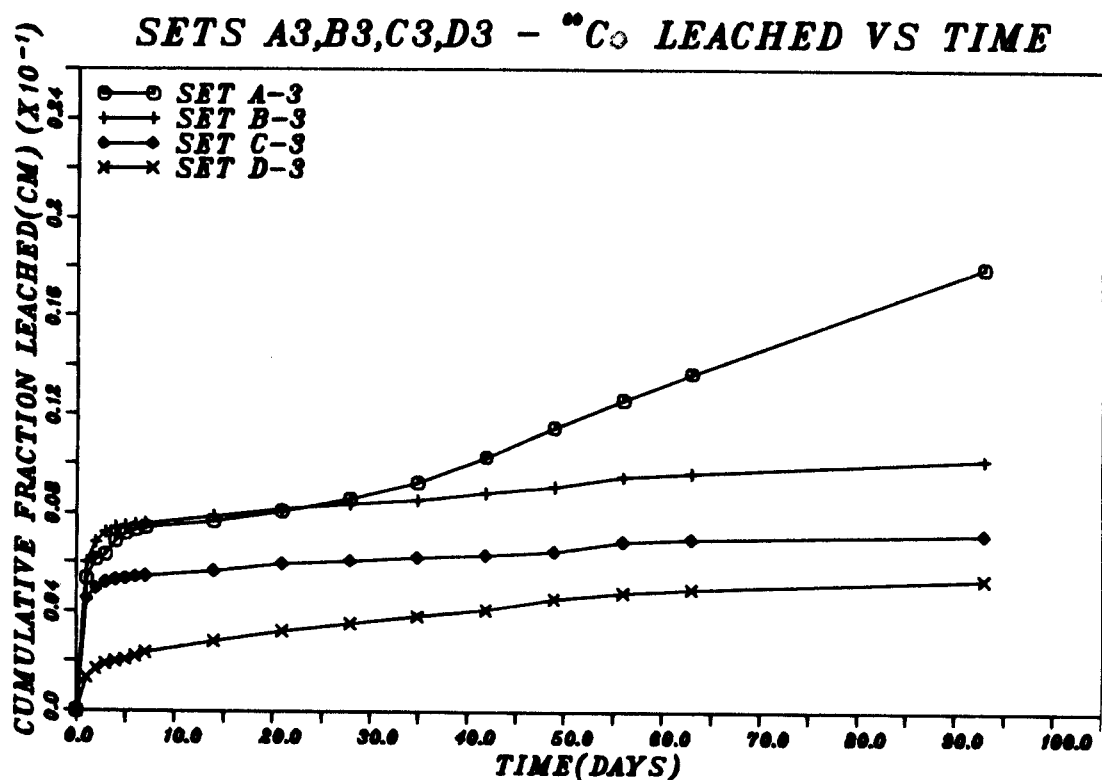


Figure 17 - Leaching profiles for sets A3,B3,C3 and D3 - ^{60}Co

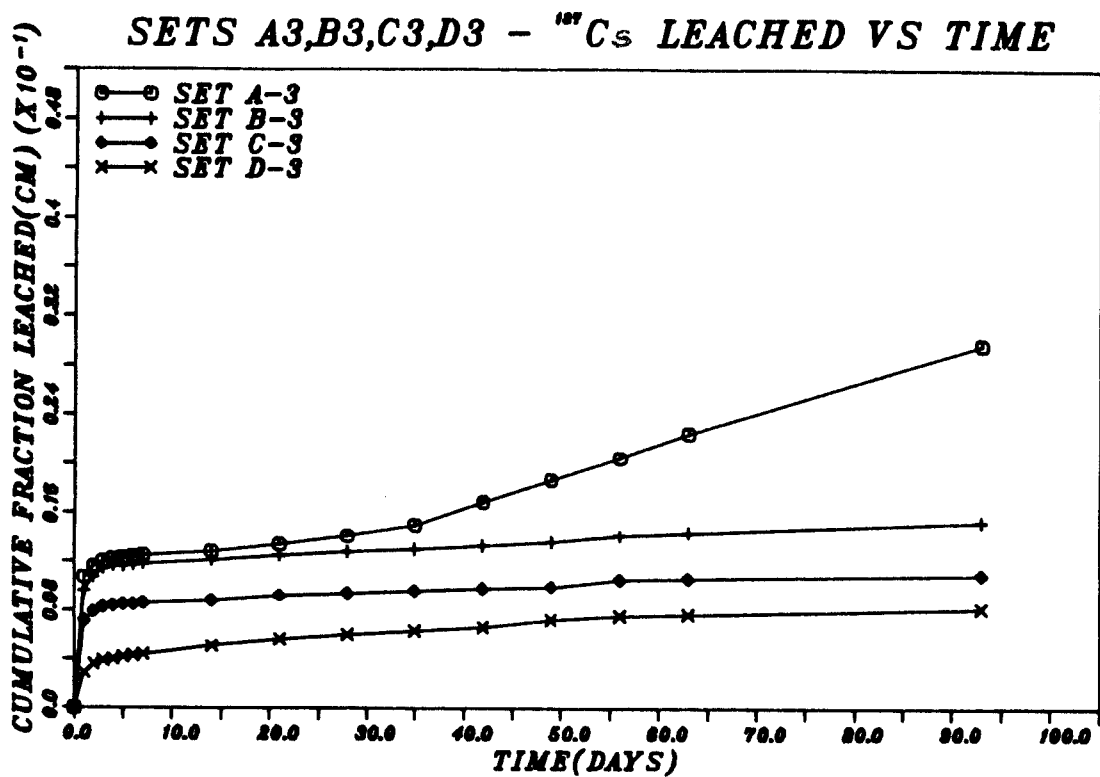


Figure 18 - Leaching profiles for sets A3,B3,C3 and D3 - ^{137}Cs

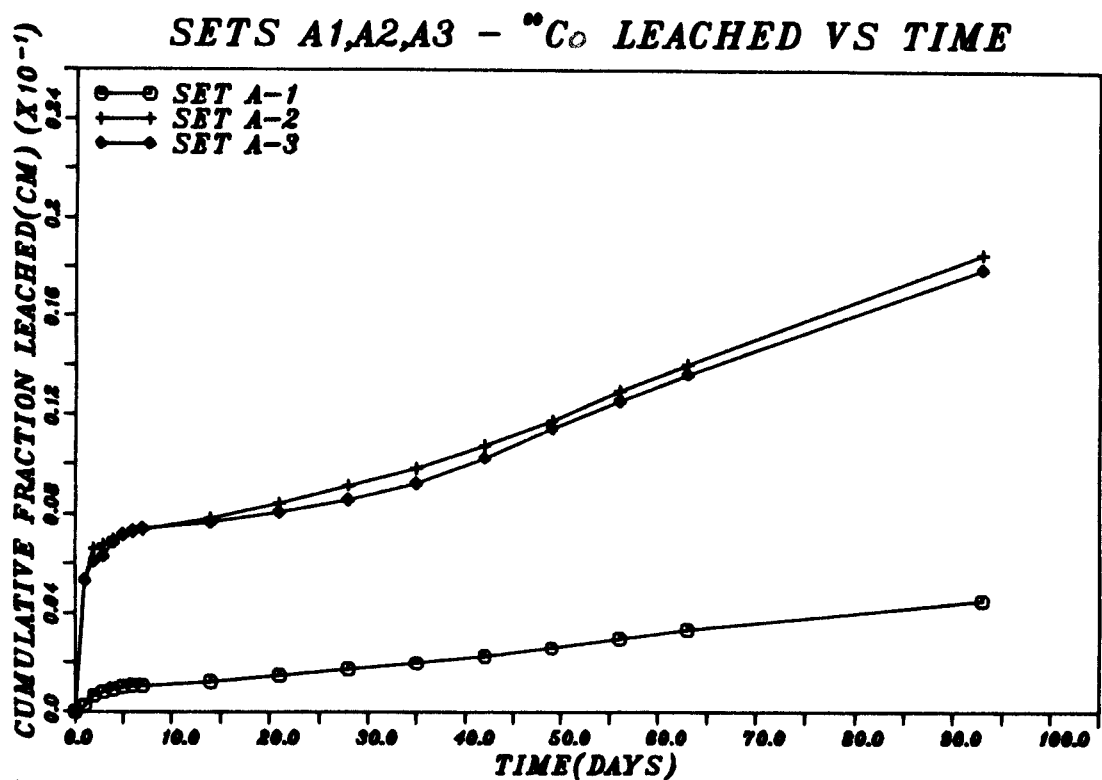


Figure 19 - Leaching profiles for sets A1,A2 and A3 - ^{60}Co

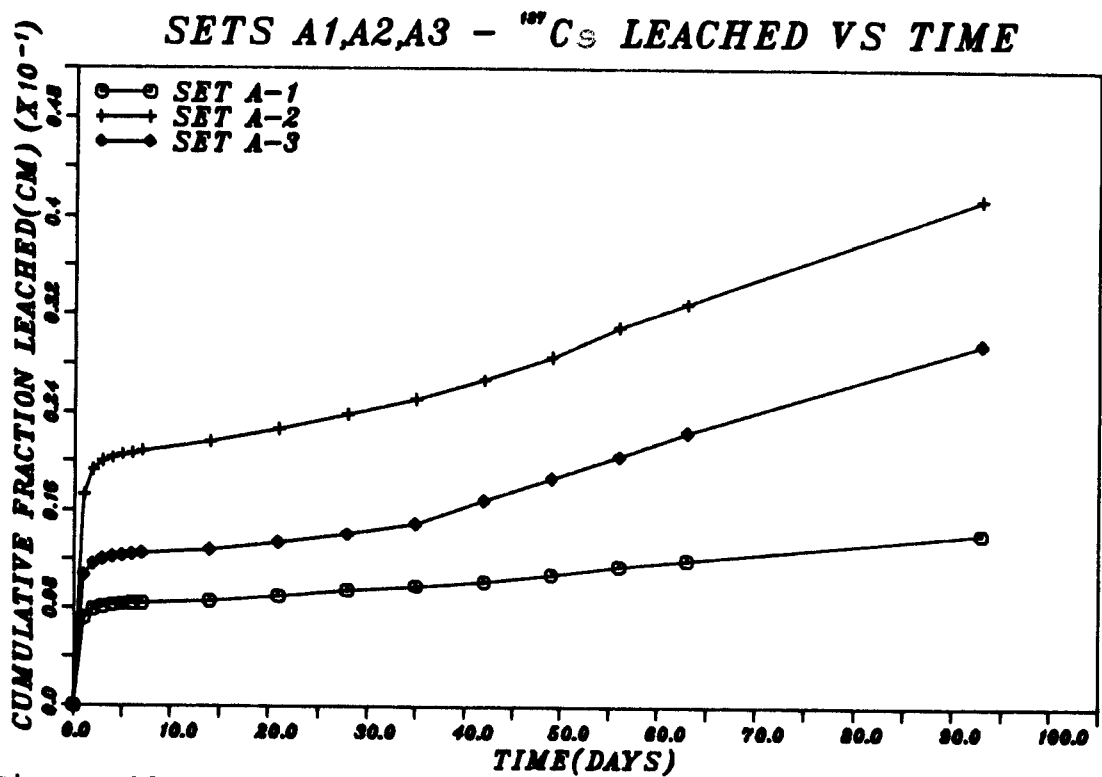


Figure 20 - Leaching profiles for sets A1,A2 and A3 - ^{137}Cs

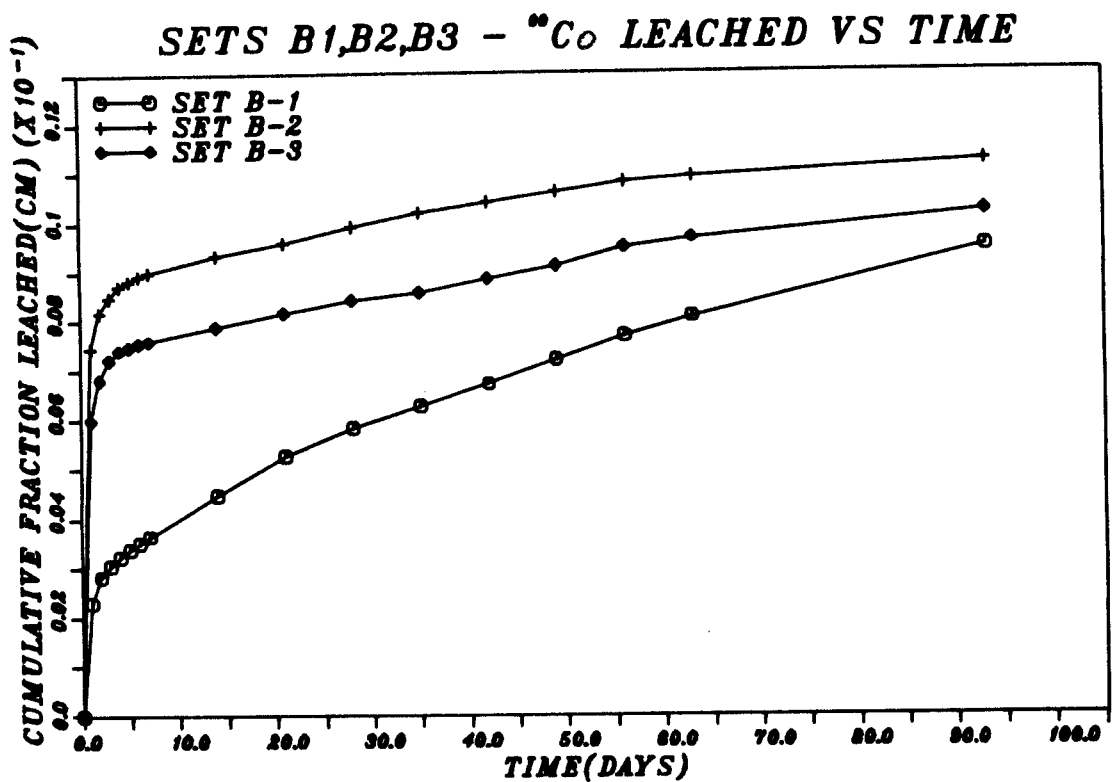


Figure 21 - Leaching profiles for sets B1,B2 and B3 - ^{60}Co

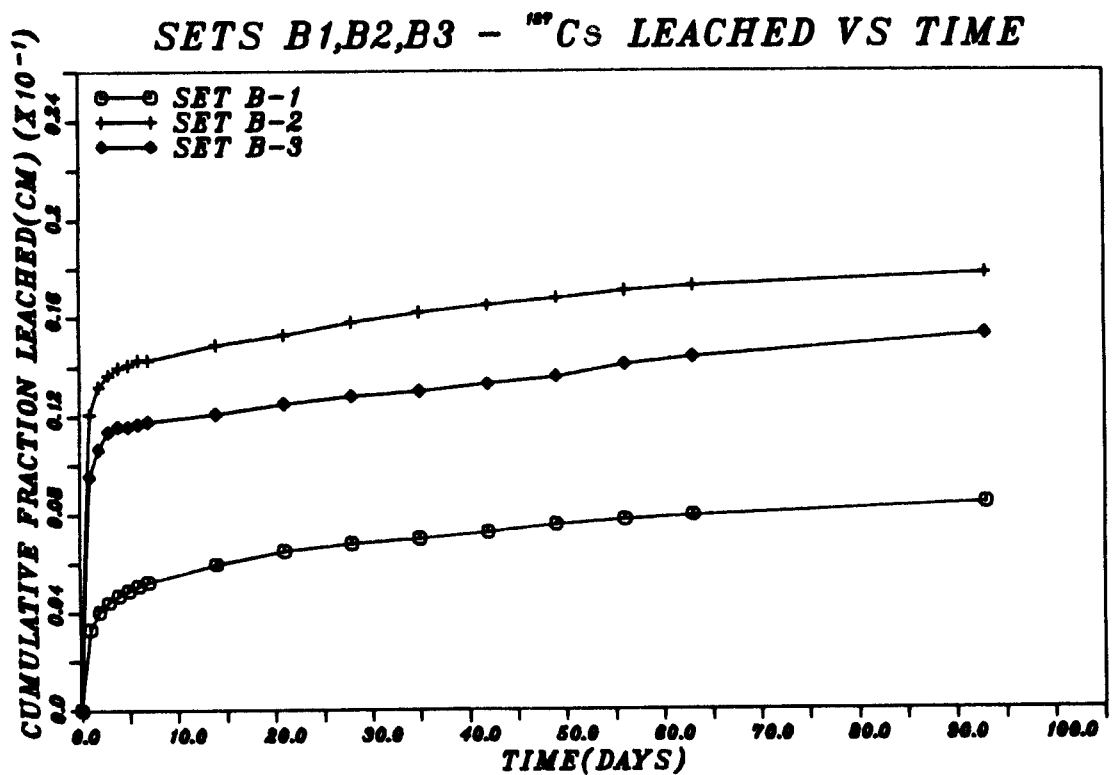


Figure 22 - Leaching profiles for sets B1,B2 and B3 - ^{137}Cs

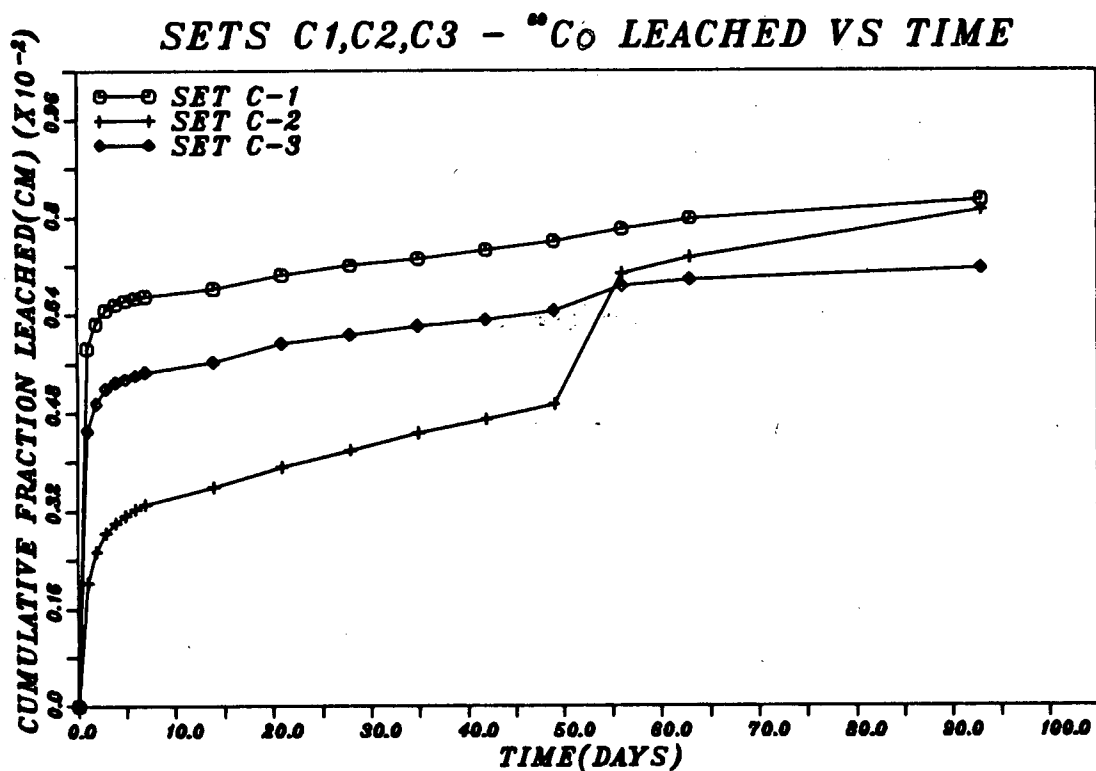


Figure 23 - Leaching profiles for sets C1,C2 and C3 - ^{60}Co

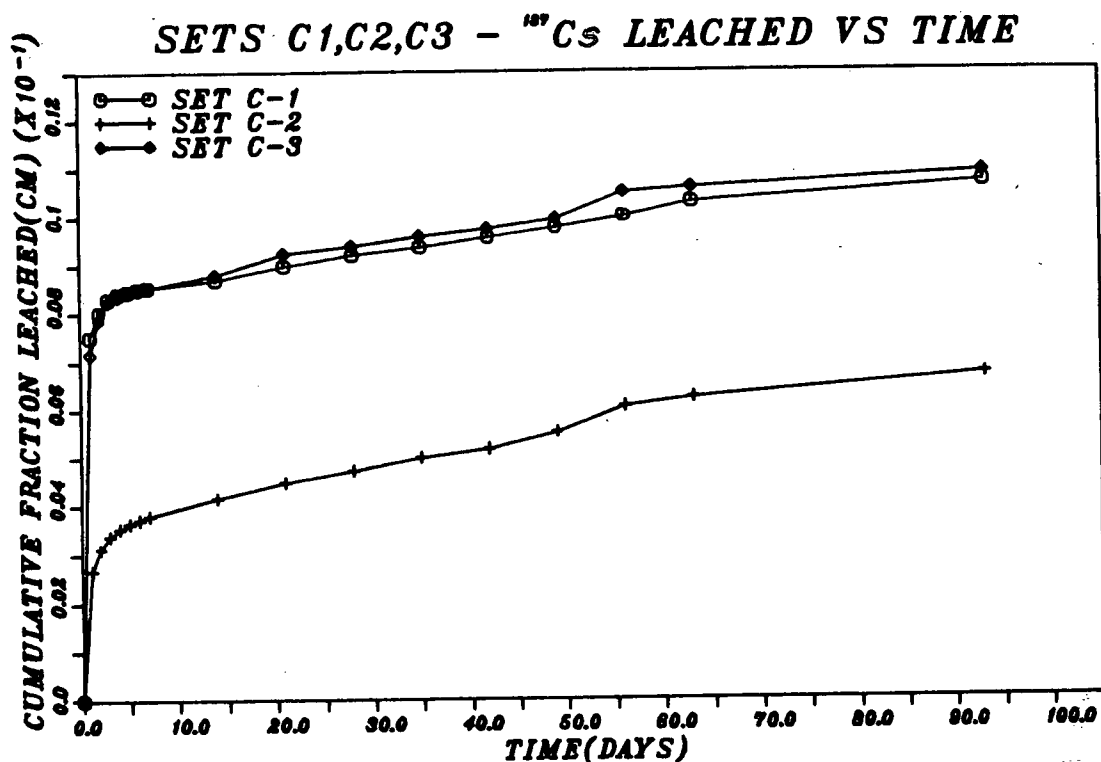


Figure 24 - Leaching profiles for sets C1,C2 and C3 - ^{137}Cs

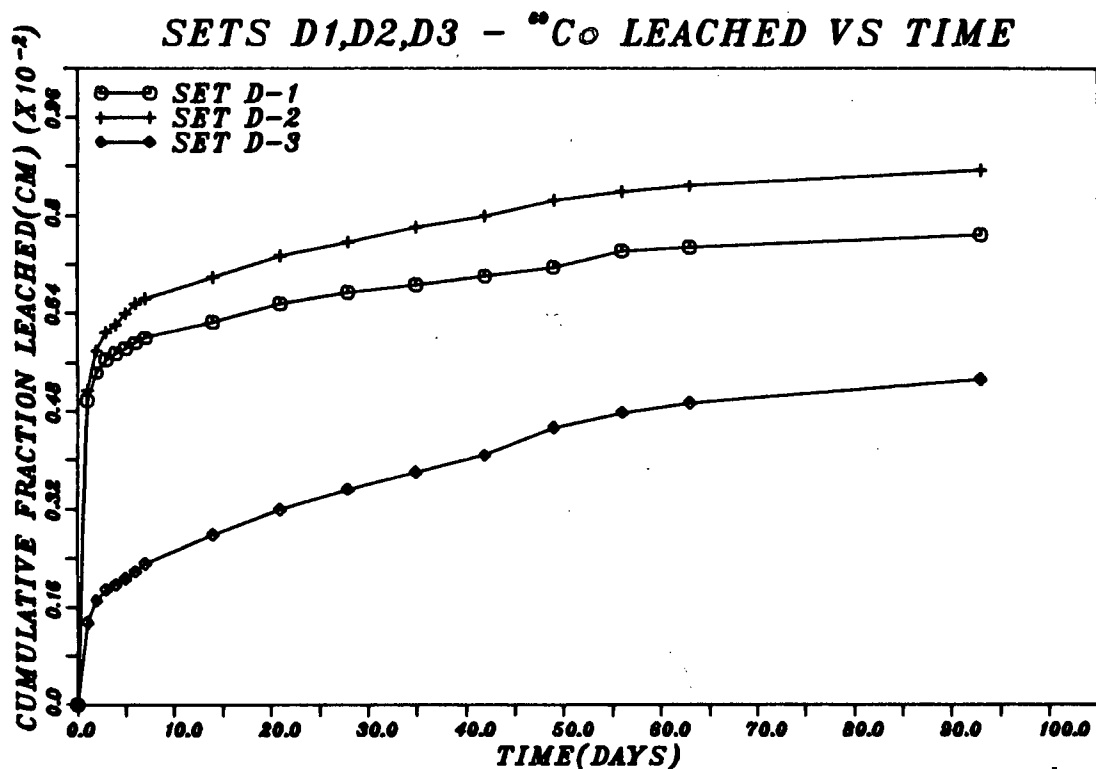


Figure 25 - Leaching profiles for sets D1,D2 and D3 - ^{60}Co

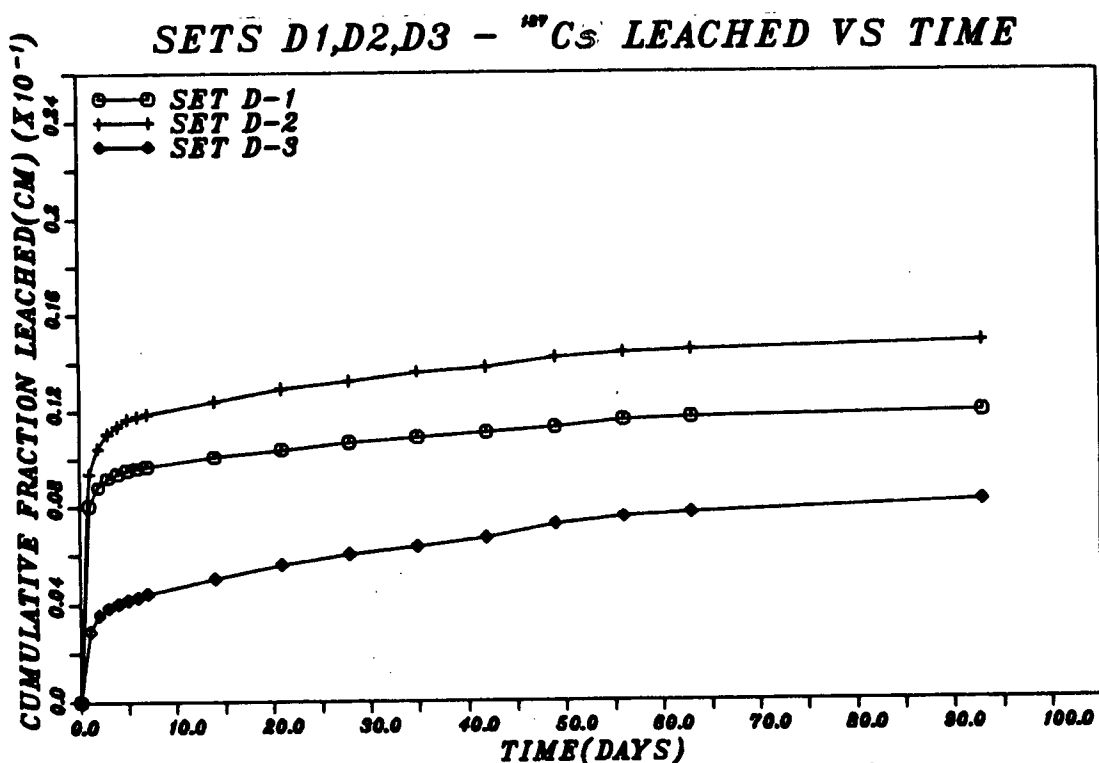


Figure 26 - Leaching profiles for sets D1,D2 and D3 - ^{137}Cs

produced surface-active agents such as lipids, glycolipids and lipoproteins.⁶²⁻⁶⁵ Obviously, the latter two modes of microbially initiated radioactive waste release are dependent on the presence of the first if bitumen is the sole carbon source (a likely scenario in a deep waste repository). Furthermore, the presence of exogenous agents that may solubilize or emulsify the bitumen is not a relevant problem unless the repository containing the waste is in or adjacent to a geological formation containing petroleum.

Until now, no investigators have undertaken to show increased release of radionuclides encapsulated in bitumen due to microbial action and only one study has reviewed microbial attack of asphalt containing inactive salts.⁶⁶ In the past, investigators have utilized any available method to maximize the contact of the microbial biomass with the substrate in order to increase the rate of attack. Unfortunately, these techniques have usually disrupted (or destroyed) the structural integrity of the bitumen to such an extent as to preclude the possibility of their use in any satisfactory experiment designed to show enhanced leaching.

In the experiment described in the following pages a standardized procedure⁶⁷ was followed that should allow good intercomparison of results. The procedure was modified slightly for test Sets C and D (described in Section II) to allow for maximum growth of hydrocarbonoclastic bacteria.

Optimal nutrient, oxygen and temperature conditions were maintained to create an experimental system that could resolve any difference in mass leaching of radionuclides from bitumen that was or was not undergoing microbial attack. Although microbial growth was not dependent on the use of bitumen as a sole carbon source, after approximately 12 hours (see Section II) an endogenous growth phase had been reached for the mixed population and further growth was dependent on the use of bitumen as a carbon source.

The objective of this investigation was simply to determine if a statistically significant difference existed between inoculated samples and their respective controls, irrespective of the mechanism (solubilization, emulsification or biodegradation) under conditions that were optimized for microbial growth.

2. METHODS

Twelve identical samples of Sp-170 (oxidized) bitumen containing 38 wt.% sodium nitrate, 4.914 $\mu\text{Ci/gm}$ ^{60}Co and 8.229 $\mu\text{Ci/gm}$ ^{137}Cs (New England Nuclear Ltd.) were prepared in a twin-screw extruder at a product flow rate of 2.163 kg/hr and a peak temperature of $<170^{\circ}\text{C}$. The final homogeneous cylindrical product had a mean mass, volume and area of 21.2 g($\sigma=1.08$), 15.7 cm^3 ($\sigma=7.90 \times 10^{-1}$) and 35.1 cm^2 ($\sigma=1.05$) respectively. While the leach samples cooled for four days at ambient temperature, four separate leachant solutions were prepared and consisted of the following:

Leachant Solution A

Distilled, demineralized water (conductivity $<1.00 \times 10^{-6}\text{Mho/cm}$).

Leachant Solution B

- 1) Distilled, demineralized water (conductivity - as above).
- 2) Mineral salts solution as described earlier (Section II).
- 3) Microbial nutrient media consisting of: 0.3% malt extract, 0.3% yeast extract and 0.5% peptone.

Leachant solution C

- 1) Distilled, demineralized water (conductivity - as above).
- 2) Mineral salts solution as described earlier (Section II).
- 3) Microbial nutrient media consisting of: 0.3% malt extract, 0.3% yeast extract and 0.5% peptone.
- 4) Hydrocarbonoclastic bacteria (Culture-D).

Leachant Solution D

- 1) Distilled, demineralized water (conductivity - as above).
- 2) WN-1 Synthetic groundwater solution as described

earlier in Section II.*

3) Microbial nutrient media consisting of: 0.3% malt extract, 0.3% yeast extract and 0.5% peptone.

4) Hydrocarbonoclastic bacteria (Culture-D).

The bitumen samples were placed in 250 ml Sybron/Nalge wide-mouth, straightside polymethylpentene jars with polypropylene screw closures. Three samples for each leachant solution were then immersed in 100 mls of leachant, in such a manner as to maximize the exposure between the leachant and sample surface (with not less than 5.0 mls covering the sample). To maintain good aeration of the microbial cultures but to minimize release of radionuclides as a result of mechanical agitation, all samples were agitated horizontally by a Lab-Line Junior Orbit Shaker at 100 RPM. The leachant solutions were changed according to the guidelines of Hespe^{6,7} - every 24 hours for the first 7 days and then once per week for the following 8 weeks. Prior to termination of the experiment a final sample was collected 30 days after the end of the 8 week sampling period. For the duration of the test all samples were maintained at an average temperature of 23.6°C ($\sigma=1.64$). In order to ensure microbial activity was at a maximum rate (exponential growth phase), all sterile leachant solutions (C and D) were inoculated 3-4 hours before

* In keeping with a "worst-possible-case" philosophy, WN-1 synthetic groundwater was selected as a suitable example since this groundwater was shown earlier (Section II) to be the most conducive to growth of Culture-D.

the time in which they were to be changed (see Section II).

A. ANALYSIS

Immediately prior to each leachant change, 25.0 mls of spent leachant were withdrawn from the sample containers and transferred to polypropylene vials. The subsequent analysis employed a multi-channel γ -spectrometer equipped with a Ge(Li)* detector. Counting took an average of 3 hours for each sample. Most samples averaged between 2-3 orders of magnitude above background (1×10^{-12} Ci/ml) therefore background was not considered a significant factor in the resulting statistical analysis.

Conductivity and pH measurements were made with the use of Radiometer (Copenhagen) and Fisher Instruments (see Appendix D).

* Ge(Li) - Germanium, Lithium.

3. RESULTS AND DISCUSSION

Visual interpretation of Figures 13 to 18 show only one consistent trend is evident in the profiles of:

$$\frac{\sum a_n}{A_0} \frac{V}{F} \quad \text{vs} \quad (t_n)^{0.5} \quad \text{Eq. IV.1}$$

(cumulative fraction leached vs time). Set A's slope is greatest in most cases at $t > 21$ days in comparison to Sets B, C and D. These latter test sets all appear to be approaching a slope=0 (Figure 16). As mentioned earlier, justification for the leaching behaviour of set A can be explained by a high ΔC . In this case, diffusion will cause a net movement of radionuclides between the bitumen and DIW.

Although all samples were virtually identical in terms of mass, volume and area, small surface imperfections would explain the initial differences in leaching behaviour of the samples. However, long term behaviour would be a function, ultimately, of the leachant rather than the sample.

A. STATISTICAL EVALUATION

Since only qualitative evaluations may be made by inspection of Figures 13 to 18, an Analysis of Variance (with one-independent variable) coupled with the Student-Newman-Keuls procedure⁶⁸ was employed as a quantitative approach to

determine differences in sample means. Since slope ~ 0 at $t \geq 14$ days (see Figure 18), sample means were evaluated from the cumulative fraction leached from $t=14$ to $t=93$ days. All statistical tests were performed independently for the two isotopes used, at an $\alpha=0.05$ significance level (see Appendix E for statistical results).

Inspection of Appendix E shows that for ^{60}Co the average means of Sets A and B were significantly different from the average means of Sets C and D at $\alpha=0.05$, and for ^{137}Cs Group A was significantly different from B, C and D. However, there was no significant difference between C and D and between B and D. Thus, the combined statistical analysis for ^{60}Co and ^{137}Cs illustrates only that the Set A average was significantly higher than sets C and D's average at $\alpha=0.05$.

Set B's mean was statistically homogeneous to Set A for ^{60}Co as a result of the abnormally low leaching behaviour of ^{60}Co for sample A1 (Figure 13). This is illustrated by the profiles of all other samples but especially Figure 21 in which A1 shows abnormally low leaching with respect to A2 and A3. Thus, it appears sample A1 could represent an anomaly in which the initial leaching (at Day 1, 2 and 3) was unusually suppressed. This abnormality, in turn, affects the statistical analysis for the entire test.

B. CONCLUSION

After 93 days of leaching under optimal conditions, microbial attack did not enhance the release of ^{137}Cs and ^{60}Co from bitmen. Extrapolation of the data and/or plots presented here, would not provide a viable case for enhanced leaching of these representative radionuclides for the long-term ($t > 93$ days). However, the cumulative fraction leached is a function of leachant conductivity (see Appendix C and D). This is certainly a consideration for real repository conditions in which the conductivity of native groundwaters are expected to be high.

V. EFFECT OF CHELATING AGENTS ON RADIONUCLIDE MIGRATION

As a result of a "defense in depth"* philosophy adopted by many of the organizations contemplating land-burial of radwastes, naturally occurring geologic media is being considered as a potential radionuclide adsorbent. The media that may comprise this barrier will occur naturally in situ, or various materials with a high adsorption capacity (bentonite) may be used to augment the burial site's natural ability to attenuate radionuclide migration through its subsurface environment.

1. BACKGROUND

Although numerous studies^{26-30, 69-75} have been performed to date on the adsorption of radionuclides to soils, sands and gravels, few experiments have considered adsorption to these materials under non-idealized conditions. The prognosis for a

* A "defence in depth" philosophy utilizes multiple barriers to arrest the return of escaped radionuclides to the biosphere.

radionuclide that has completely breached its solidifying matrix but abuts a well packed adsorbent is quite good. This adsorbent will tend to reduce the movement of the released radionuclide via two independent mechanisms: a) a well-packed, highly dense adsorbent of small particle size will drastically reduce the groundwater flow rate and therefore decrease the active particle's linear velocity; and b) the adsorbent will "tie-up" the escaped nuclides through various physicochemical mechanisms such as ion-exchange, van der Waals attraction, covalent bonding, etc.

Unfortunately, numerous mitigating factors exist that may reduce the ability of the backfill material to bind released radionuclides. These factors have only received cursory attention. For instance, the ability of some dissolved compounds to have strong ionic, secondary or complex interactions with common fission products may serve to impair or neutralize the geologic media's ability to attenuate these radionuclides. Of potential concern to the Nuclear Industry are multidentate chelating agents such as ethylenediamine-tetraacetic acid (EDTA), diethylenetriaminepentaacetic acid (DTPA) or cyclohexanediaminetetraacetic acid (CDTA). Since many of these compounds are used in conjunction with detergents, their use for radioactive decontamination is essentially ubiquitous.

A recent study²⁶ has shown a 20,000 fold decrease in the

adsorptive capacity of Conasauga shale for ^{60}Co due to the presence of extremely low concentrations of EDTA. As a result, many ^{60}Co contaminant plumes showing abnormally high migration rates may be attributed to the decreased ability of the soil to bind complexed ^{60}Co .²⁶

In addition to the synthetic multidentate ligands mentioned above - many naturally occurring organics may act as complexing agents. These organics include humic and fulvic acids, many dicarboxylic acids, and various microbially-generated biochemicals such as hydroxamate and polyhydroxamate.²⁷ Even glycine, the simplest amino acid, may serve to complex waste radionuclides.

Due to this potential for enhanced migration of radionuclides through geologic media due to microbial action, the following set of experiments was designed to illustrate if there was an observable effect and if so, to determine its magnitude. Accompanying those runs that directly employed microbial populations were other experiments that would show the adsorption of ^{60}Co , ^{137}Cs and ^{85}Sr to bentonite, gabbro and granite in the presence of EDTA, Turco (a common radioactive decontamination compound), SCSSS, Granite Groundwater* and deionized, demineralized water. In order to simulate real conditions, all solutions that contained

* The composition of SCSSS (Standard Canadian Shield Saline Solution) and Granite Groundwater is discussed in Appendix B.

microbial populations were taken from the leachant solution (after 1 week's contact) mentioned in Section IV. From the combined results of these experiments the effect of EDTA, Turco, microbes and the antagonistic (or enhanced) effect of competing cations (^{60}Co - ^{137}Cs , ^{85}Sr - ^{137}Cs) could be compared and interrelated.

2. METHODS

Eight independent experimental runs were performed and are listed in Tables III-X following. For each determination, 1.0 g of adsorbent (Fisher-bentonite, 40-50 mesh gabbro or granite) was placed in an acid-washed (10 minutes in 6N HNO_3) polypropylene test tube and mixed with 14.8 mls of adsorbate consisting of "super Q"* water, SCSSS, Granite Groundwater, 10^{-5}M EDTA, Turco** or the leachant material (1 week's contact) of set A or D described earlier (see Section IV). Standard solutions of ^{60}Co , ^{137}Cs , ^{85}Sr or a combination of ^{60}Co and ^{137}Cs or ^{85}Sr and ^{137}Cs were then added (0.2 mls) to all samples not containing leachant material to yield an initial total activity not exceeding $1.05 \times 10^{-1} \mu\text{Ci}$. Each tube was then sealed with parafilm and allowed to rotate on a

* "Super Q" water is water that has been distilled, deionized, millipore filtered and passed through activated carbon.

** Turco is a common decontamination solution containing 6.0 wt.% diammonium hydrogen citrate, 2.5 wt.% oxalic acid and 1.0 wt.% phenylthiourea. This solution was diluted to a diammonium hydrogen citrate concentration of 10^{-5}M .

Scientific Industries Incorporated Model 151 rotater. Agitation was continued for 48 hours at 5 RPM to minimize grain abrasion.³⁰ Temperature ranged from 20-25°C. Upon termination of each run adsorbate and adsorbent were separated via centrifugation at 15,000 RPM for 60 minutes. Appropriate controls (test radionuclide + liquid phase only) were run with each experimental group. To evaluate the effect nutrient media (composition reported in Section II) had on adsorptivity, an entire experimental run was set-up consisting of nutrient media spiked with ⁶⁰Co and ¹³⁷Cs. The results of this control group are shown in Table IX.

3. ANALYSIS

Upon termination of centrifugation 6.0 mls of supernatant were collected, placed in polypropylene vials and counted on a multi-channel γ -spectrometer as per Section IV. The γ -activity background and count time remained unchanged (from that described in Section IV) for all analyses.

According to convention,^{30, 76} the distribution coefficient, K_D , was evaluated according to the following formula:

$$K_D \text{ (ml/g)} = \frac{(C_0 - C_e)(V)}{(C_e)(M)} \quad \text{Eqt.V.1}$$

Where C_0 is the radionuclide concentration initially, C_e is the radionuclide concentration at equilibrium, V is the volume of liquid and M is the mass of adsorbent material.

Evaluation of this equation for every run yields a numerical expression for the distribution of each radionuclide between the liquid and solid phase. This method allows for good intercomparison of results as adsorbent or other experimental conditions change.

The initial concentration of each radionuclide was evaluated through controls that contained components identical to the experimental sets except for an adsorbent. Unfortunately, it became evident from these runs that some radionuclide adsorption to the reaction vessel walls occurred. To minimize this effect, 10^{-5}M EDTA was added to ^{60}Co and ^{137}Cs controls. The presence of EDTA in the control tubes decreased the adsorption of cobalt to the vessel walls. Since the net adsorption (as expressed by K_D) of ^{60}Co is greatly effected by C_0 all K_D 's for ^{60}Co were calculated from a C_0 that contained EDTA.

Since a microbial biomass may adsorb various species, all K_D 's of solutions containing active cultures were evaluated from a C_0 of net available (unadsorbed) activity. Thus, the available activity of an active solution of leachant (from Section IV) was evaluated as that activity remaining free in solution after the treatment described earlier in Methods but with no adsorbent. This alteration will resolve the difference between radionuclides adsorbed to the microbial population and the specific adsorbent.

4. RESULTS AND DISCUSSION

Tables III-V indicate that adsorption of ^{60}Co was higher than that of any other nuclide. However, the effect of EDTA was much more pronounced with cobalt and with a granite adsorbent, it elicits a 1900 fold decrease in K_D . This result, although not as dramatic, is in accordance with that reported by Means and Crerar.²⁶ As expected, EDTA had only a negligible effect on ^{137}Cs adsorption but ^{85}Sr had slightly decreased K_D 's due to EDTA. However, since EDTA does not form strong complexes with anything other than rare earths, transition metals or transuranics, this effect may be expected.

The relative effect of Turco on decreasing K_D was greatest for ^{85}Sr in which an 11 fold decrease in adsorption to bentonite was observed.

Sample	$^{60}\text{Co}(\mu\text{Ci/ml})$	K_D
Bentonite	2.051×10^{-4}	7.41×10^3
Gabbro	4.508×10^{-3}	3.23×10^2
Granite	1.493×10^{-3}	1.01×10^3
Bentonite+EDTA	3.068×10^{-3}	4.82×10^2
Gabbro+EDTA	1.050×10^{-1}	0.00
Granite+EDTA	9.813×10^{-2}	5.25×10^{-1}
Bentonite+Turco	2.908×10^{-4}	5.22×10^3
Control	1.016×10^{-1}	

Table III - ^{60}Co ADSORPTION DATA

Sample	$^{137}\text{Cs}(\mu\text{Ci/ml})$	K_D
Bentonite	1.643×10^{-3}	7.54×10^2
Gabbro	7.325×10^{-3}	1.57×10^2
Granite	4.615×10^{-3}	2.59×10^2
Bentonite+EDTA	1.695×10^{-3}	7.30×10^2
Gabbro+EDTA	7.513×10^{-3}	1.53×10^2
Granite+EDTA	3.154×10^{-3}	3.85×10^2
Bentonite+Turco	1.899×10^{-3}	6.50×10^2
Control	8.419×10^{-2}	

Table IV - ^{137}Cs ADSORPTION DATA

Sample	$^{85}\text{Sr}(\mu\text{Ci/ml})$	K_D
Bentonite	6.975×10^{-4}	2.23×10^3
Gabbro	3.164×10^{-2}	3.44×10^1
Granite	4.619×10^{-2}	1.88×10^1
Bentonite+EDTA	7.931×10^{-4}	1.96×10^3
Gabbro+EDTA	1.028×10^{-1}	2.02×10^1
Granite+EDTA	1.036×10^{-1}	8.51×10^2
Bentonite+Turco	7.625×10^{-3}	1.90×10^2
Control	1.042×10^{-1}	

Table V - ^{85}Sr ADSORPTION DATA

Sample	$^{60}\text{Co}(\mu\text{Ci/ml})$	$^{137}\text{Cs}(\mu\text{Ci/ml})$	$^{85}\text{Sr}(\mu\text{Ci/ml})$
Co,Cs+Bentonite	1.757×10^{-4}	8.650×10^{-4}	
Sr,Cs+Bentonite		8.906×10^{-4}	4.199×10^{-4}
Co,Cs+Gabbro	1.766×10^{-3}	2.066×10^{-3}	
Sr,Cs+Gabbro		2.481×10^{-3}	1.403×10^{-2}
Co,Cs+Granite	7.181×10^{-4}	1.824×10^{-3}	
Sr,Cs+Granite		2.024×10^{-3}	2.141×10^{-2}
Co,Cs(Control)	1.224×10^{-4}	2.508×10^{-3}	
Sr,Cs(Control)		3.658×10^{-2}	1.743×10^{-3}

Sample	^{60}Co K_D	^{137}Cs K_D	^{85}Sr K_D
Co,Cs+Bentonite	3.50×10^3	8.71×10^2	
Sr,Cs+Bentonite		8.45×10^2	1.83×10^3
Co,Cs+Gabbro	3.35×10^2	3.56×10^2	
Sr,Cs+Gabbro		2.94×10^2	4.03×10^1
Co,Cs+Granite	8.46×10^2	4.05×10^2	
Sr,Cs+Granite		3.64×10^2	2.13×10^1

Table VI - COMPETING ION ADSORPTION DATA

<u>Sample</u>	<u>$^{60}\text{Co}(\mu\text{Ci/ml})$</u>	<u>$^{137}\text{Cs}(\mu\text{Ci/ml})$</u>	<u>$^{85}\text{Sr}(\mu\text{Ci/ml})$</u>
Co,Cs+Bentonite			
+ Granite G/W	1.774×10^{-4}	1.086×10^{-3}	
Sr,Cs+Bentonite			
+ Granite G/W		1.033×10^{-3}	4.911×10^{-4}
Co,Cs+Granite			
+ Granite G/W	7.550×10^{-4}	3.408×10^{-3}	
Sr,Cs+Granite			
+ Granite G/W		3.859×10^{-3}	3.870×10^{-2}
Co,Cs+Bentonite			
+ SCSSS G/W	8.181×10^{-4}	1.084×10^{-2}	
Sr,Cs+Bentonite			
+ SCSSS G/W		1.091×10^{-2}	5.022×10^{-2}
Co,Cs+Granite			
+ SCSSS G/W	2.236×10^{-2}	2.084×10^{-2}	
Sr,Cs+Granite			
+ Granite G/W		1.969×10^{-2}	5.279×10^{-2}
<u>Sample</u>	<u>$^{60}\text{Co } K_0$</u>	<u>$^{137}\text{Cs } K_0$</u>	<u>$^{85}\text{Sr } K_0$</u>
Co,Cs+Bentonite			
+ Granite G/W	3.47×10^3	6.91×10^2	
Sr,Cs+Bentonite			
+ Granite		7.27×10^2	1.57×10^3
Co,Cs+Granite			
+ Granite G/W	8.04×10^2	2.10×10^2	
Sr,Cs+Granite			
+ Granite		1.84×10^2	5.06×10^0
Co,Cs+Bentonite			
+ SCSSS G/W	7.40×10^2	5.57×10^1	
Sr,Cs+Bentonite			
+ SCSSS G/W		5.52×10^1	4.55×10^{-1}
Co,Cs+Granite			
+ SCSSS G/W	1.26×10^2	2.18×10^1	
Sr,Cs+Granite			
+ Granite G/W		2.39×10^1	0.00

Table VII - ADSORPTION DATA FOR COMPETING IONS IN SELECTED GROUNDWATERS

Sample	$^{60}\text{Co}(\mu\text{Ci/ml})$	$^{137}\text{Cs}(\mu\text{Ci/ml})$
Set A+Bentonite	3.955×10^{-4}	7.763×10^{-4}
Set A+Gabbro	1.490×10^{-3}	1.902×10^{-3}
Set A+Granite	4.411×10^{-4}	9.788×10^{-4}
Set A (Control)	8.838×10^{-3}	2.359×10^{-2}

Sample	$^{60}\text{Co } K_d$	$^{137}\text{Cs } K_d$
Set A+Bentonite	3.20×10^2	4.41×10^2
Set A+Gabbro	7.40×10^1	1.71×10^2
Set A+Granite	2.86×10^2	3.47×10^2

Table VIII - LEACHANT A ADSORPTION DATA

Sample	$^{60}\text{Co}(\mu\text{Ci/ml})$	$^{137}\text{Cs}(\mu\text{Ci/ml})$
Set C+Bentonite	2.001×10^{-3}	1.867×10^{-3}
Set C+Gabbro	5.815×10^{-3}	1.278×10^{-2}
Set C+Granite	6.030×10^{-3}	1.249×10^{-2}
Set C (Control)	5.631×10^{-3}	1.299×10^{-2}

Sample	$^{60}\text{Co } K_d$	$^{137}\text{Cs } K_d$
Set C+Bentonite	2.72×10^1	8.93×10^1
Set C+Gabbro	0.00	2.44×10^{-1}
Set C+Granite	0.00	5.97×10^{-1}

Table IX - ADSORPTION DATA FOR NUTRIENT MEDIA CONTROL

Sample	$^{60}\text{Co}(\mu\text{Ci/ml})$	$^{137}\text{Cs}(\mu\text{Ci/ml})$
Set D+Bentonite	1.949×10^{-3}	6.519×10^{-3}
Set D+Gabbro	7.781×10^{-3}	1.795×10^{-2}
Set D+Granite	7.819×10^{-3}	1.836×10^{-2}
Set D (Control)	1.135×10^{-4}	3.062×10^{-4}

Sample	$^{60}\text{Co } K_d$	$^{137}\text{Cs } K_d$
Set D+Bentonite	3.96×10^1	2.90×10^1
Set D+Gabbro	0.00	9.94×10^{-1}
Set D+Granite	0.00	6.35×10^{-1}

Table X - LEACHANT D ADSORPTION DATA

The competing effects of ^{60}Co on ^{137}Cs and ^{85}Sr on ^{137}Cs are reported in Table VI. In all cases bentonite showed the highest adsorptive capacity followed by granite (for ^{60}Co and ^{137}Cs) and gabbro. The relative difference in K_D 's for the three adsorbents decreased for each group of radionuclides from their non-competitive counterpart. As evidenced by the data in Table VI, the adsorptive capacity of bentonite was decreased by the competing ions while that for gabbro and granite was increased (perhaps due to a non-competitive mechanism for adsorption). Table VIII presents data relating to the effect of synthetic groundwaters on K_D . In every case and for both adsorbents tested (bentonite and granite) the groundwater with the highest conductivity (SCSSS) has the greatest effect on decreasing K_D . This higher figure is easily explained by the saturation of available active sites on the adsorbent by non-active ions present in the highly concentrated brine.

Bentonite showed a greatly reduced adsorptivity for the leached radionuclides contained in leachant A and D. However, it appears that leachant A (DIW) only slightly perturbed the adsorptive capacity of granite and gabbro. In the case of leachant A contacted with gabbro or granite, a < 2 fold reduction was found for K_D 's between this leachant and deionized, demineralized water containing ^{60}Co and ^{137}Cs . The effect of leachants C and D on the adsorption of these

radionuclides to granite or gabbro is significant. The adsorptivity of both gabbro and granite for the radionuclides contained in leachant C or D was decreased more than 100 times by the complexing agents present in the two solutions (as evidenced by the K_D values reported in Tables VI, IX and X). XXXIII and XXXIV.

5. REMARKS

The effect of complexing agents (whether synthetic or otherwise) may cause a reduced attenuation of waste radionuclides leaching from a nuclear waste repository. As evidenced from the results, strong chelating agents present at $10^{-5}M$ can decrease a K_D by up to 3 orders of magnitude. Turco did not decrease K_D 's as much as EDTA, probably because its major constituent (diammonium hydrogen citrate) is not as strong a complexing agent as EDTA.

VI. CONCLUSION

Throughout the course of these experiments, conditions for microbial growth were maintained at a level only possible under the idealized environment of a laboratory. The conditions expected in a full-scale low-level repository are as follows:

- a) a moderately low temperature ($<20^{\circ}\text{C}$);
- b) a low oxygen atmosphere (approaching 0.0ppm O_2); and
- c) devoid of any microbial growth.

Since it is assumed that water will fill the repository, the presence of an unusually high salt content may be expected that would be bacteriocidal or bacteriostatic for all but a few halophiles.

In addition to the conditions listed above, a fourth constraint that may inhibit the microbial growth under repository conditions is the lack of an initial contaminating culture that is capable of utilizing various hydrocarbons as a

substrate.

Also, the low temperature ($<20^{\circ}\text{C}$), high pressure and lack of oxygen will serve to create a microbially hostile environment. Even if various organisms could survive under these conditions and utilize bitumen as their sole carbon source, the rate of their metabolism (and therefore rate of oxidation of bitumen) would be extremely slow.

In addition to the conditions listed above, a fourth constraint that may inhibit the microbial growth under repository conditions is the lack of an initial contaminating culture that may utilize various hydrocarbons as a substrate. The solidification of waste radionuclides with bitumen requires a bitumen temperature of approximately 175°C . The molten bitumen-waste mixture will then be placed in a stainless steel container. The effect of both the molten bitumen and the impermeable container will: 1) kill any contaminating microbes on or near the bitumen; 2) volatilize the most easily metabolized components of the bitumen (light alkane fraction); 3) create an anhydrous environment; and 4) prevent microbial contamination during storage or final disposal.

As illustrated by the results presented in Section III, the anticipated background radioactivity will not affect the net growth of a microbial population. Changes may occur to the overall genotype of the population as a result of an

increased rate of mutation. However, the number of viable organisms will not change due to the radiation expected under repository conditions.

Although microbially enhanced leaching from bitumen was not found, a significant decrease in K_p was observed as a result of microbial action but its importance is secondary to complexation and/or chelation due to synthetic agents. Also, since microbial proliferation will be highly limited, any or all enhanced migration of a waste radionuclide due to microbial action would probably be "swamped" by migration of nuclides chelated prior to conditioning (provided the complex is not thermally labile).

A secondary influence of a microbial population present in situ may be to utilize organic complexes as a metabolite. This in turn would serve to decrease enhanced radionuclide migration due to any previous chelation reactions.

On the basis of these short-term experiments, the effect of microbial action on long-term radioactive waste disposal should be small. If microbial attack does occur, other factors such as diffusion or chelation (to synthetic chelating agents) would "swamp" this effect.

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APPENDIX A - ONTARIO MINISTRY OF THE ENVIRONMENT ANALYSIS*

TEST/COLONY	A	B	C	D1	D2
Gram stain	-	+	-	-	-
Shape	Rod	Rod	Rod	Rod	Rod
Spores	-	+	-	-	-
Motility 30°C	+	+	+	+	-
20°C					-
Catalase	+	+	+	+	+
Oxidase	+	-	-	-	+
Glucose OF (5 Day)	-	-	-	F	-
Growth 20°C	+	+	+		+
TSA 30°C	+	+	+		+
35°C	+	+	Weak		+
42°C	+	+	-		-
MacConkey Agar 30°C	Gr + LAC-	No Gr	No Gr	Gr + LAC+	Gr + LAC-
Skim Growth	+	+	Weak	+	+
Milk Pigment	-	-	-	-	-
Agar Caseinase	-	-	-	-	-
Flouresc.	-	-	-	-	+
Nitrate (5 Day) Reduction	+(Gas)	-	+(NO ₂)	+(NO ₂)	-
Gelatinase	-	+	+		-
Arginine (5 Day) Dihydrolase	-		-		+
Urease	-		-		+
Citrate					+
Growth 6.5% NaCl	Weak+		+		-
ONPG	-		+		-

"+" and "-" represent growth or no growth, respectively.

* Independent analysis was performed for each of the two distinct species found in Culture D

Colony Morphology:
TSA 30°C

- A - Tan, dry, wrinkled.
- B - Cream to white, irregular, spreading, margin.
- C - Tiny, circular, smooth, pale yellow.
- D1- Circular, cream, convex, smooth, entire, margin.
- D2- Circular, cream, convex, smooth.

APPENDIX B - SYNTHETIC GROUNDWATER SOLUTIONS

STANDARD SYNTHETIC GRANITE GROUNDWATER^{4 4}

- 1) The following stock solutions were made up:
- | | | |
|-------------|--------------------------------------|-------|
| a) 11.090 g | MgSO ₄ ·7H ₂ O | /25ml |
| b) 7.115 g | MgCl ₂ ·6H ₂ O | /25ml |
| c) 1.512 g | NaHCO ₃ | /25ml |
| d) 1.965 g | KOH | /25ml |
| e) 0.506 g | KNO ₃ | /25ml |
| f) 0.291 g | KF | /25ml |

2) 0.10 ml of (a), (b), (c), (d), (e) and (f) were pipetted to a 2 L volumetric flask. 1.00 ml of (c) was added and the volume made up to 1700 mls with deionized water (DIW). 0.048 g Ca(OH)₂ was added to a 200 ml volumetric flask filled with about 180 mls of DIW. CO₂ was bubbled through this mixture while stirring until the solution became clear. It was then filled to the mark with DIW and the contents of this 200 ml flask added to the 2 L volumetric flask. The final 100 ml of DIW was added to the volumetric flask to make the volume up to 2.00 L and the solution was stirred for 24 hours in contact with the atmosphere to bring the pH to 6.5±0.5.

STANDARD CANADIAN SHIELD SALINE SOLUTION (SCSSS)⁴⁴

To 100.0 mls of DIW the following dry chemicals were added:

- | | |
|------------|---|
| a) 1.906 g | KCl |
| b) 1.216 g | SrCl ₂ .6H ₂ O |
| c) 3.080 g | Na ₂ SiO ₃ .9H ₂ O |
| d) 0.276 g | NaHCO ₃ |
| e) 1.370 g | NaNO ₃ |

10.0 mls of this stock solution was added to a 2 L volumetric flask and made up to 2.0 litres. 110.050 g CaCl₂.2H₂O, 4.056 g MgSO₄.7H₂O and 25.420 g NaCl were added and the entire mixture was stirred thoroughly.

BASALT GROUNDWATER^{4 4}Stock Solution A

The following was combined in a 200 ml volumetric flask:

- | | |
|------------|---|
| a) 100 ml | DIW |
| b) 8.000 g | NaHCO ₃ |
| c) 5.485 g | Na ₂ SO ₄ ·10H ₂ O |
| d) 0.994 g | MgSO ₄ ·7H ₂ O |
| e) 1.490 g | KCl |
| f) 0.232 g | KF |

This solution was stirred until dissolved and the volume made up to 200 mls.

The following was then combined in a 2 L volumetric flask:

- | | |
|-------------|--|
| a) 1800 ml | DIW |
| b) 0.0109 g | CaSO ₄ ·1/2H ₂ O |

and stirred until dissolved.

Stock Solution B

The following was then combined in a 200 ml volumetric flask:

- | | |
|------------|--------------------------------------|
| a) 180 ml | DIW |
| b) 1.871 g | CaCl ₂ ·2H ₂ O |

This was mixed until dissolved and the volume made up to 2 L.

To the stirred 2 L volumetric flask the following was added:

- | | |
|-----------|------------------|
| a) 4.0 ml | Stock Solution A |
| b) 4.0 ml | Stock Solution B |

and made up to 2.0 litres, then stirred overnight.

WN-1 Saline Solution^{4 4}

- 1) The following stock solution was made-up in a 200 ml volumetric flask (filled to the mark with DIW)
 - a) 0.537 g KCl
 - b) 1.882 g NaHCO₃
 - c) 0.901 g NaNO₃
- 2) 20.0 mls of this stock solution was pipetted into a 2 L volumetric flask, the following dry chemicals added, then made up the mark with DIW.
 - d) 0.056 g FeSO₄.7H₂O
 - e) 0.150 g SrCl₂.6H₂O
 - f) 13.111 g CaCl₂.2H₂O
 - g) 1.232 g MgSO₄.7H₂O
 - h) 9.520 g NaCl
 - i) 1.282 g Ca(OH)₂

APPENDIX C - LEACH TEST DATA

TEST A1 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.686E-01	0.0623	0.284E-03	0.284E-03	0.384E-03	0.734E-12
2.	0.164E+00	0.1495	0.682E-03	0.397E-03	0.537E-03	0.211E-11
3.	0.200E+00	0.1822	0.831E-03	0.149E-03	0.201E-03	0.209E-11
4.	0.226E+00	0.2056	0.937E-03	0.106E-03	0.144E-03	0.200E-11
5.	0.244E+00	0.2220	0.101E-02	0.751E-04	0.101E-03	0.186E-11
6.	0.257E+00	0.2333	0.106E-02	0.512E-04	0.692E-04	0.171E-11
7.	0.266E+00	0.2420	0.110E-02	0.399E-04	0.539E-04	0.158E-11
14.	0.295E+00	0.2677	0.122E-02	0.167E-04	0.226E-04	0.967E-12
21.	0.355E+00	0.3227	0.147E-02	0.358E-04	0.484E-04	0.937E-12
28.	0.423E+00	0.3844	0.175E-02	0.402E-04	0.543E-04	0.997E-12
35.	0.476E+00	0.4324	0.197E-02	0.312E-04	0.422E-04	0.101E-11
42.	0.541E+00	0.4921	0.224E-02	0.389E-04	0.525E-04	0.109E-11
49.	0.622E+00	0.5656	0.258E-02	0.479E-04	0.647E-04	0.123E-11
56.	0.717E+00	0.6515	0.297E-02	0.559E-04	0.755E-04	0.143E-11
63.	0.796E+00	0.7240	0.330E-02	0.472E-04	0.638E-04	0.157E-11
93.	0.108E+01	0.9800	0.447E-02	0.389E-04	0.526E-04	0.195E-11

Table XI - LEACH TEST DATA - TEST SET A1 - ⁶⁰Co

TEST A2 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.125E+01	1.1811	0.532E-02	0.532E-02	0.719E-02	0.258E-09
2.	0.155E+01	1.4669	0.661E-02	0.129E-02	0.174E-02	0.199E-09
3.	0.159E+01	1.4978	0.675E-02	0.139E-03	0.188E-03	0.138E-09
4.	0.165E+01	1.5533	0.700E-02	0.250E-03	0.338E-03	0.111E-09
5.	0.168E+01	1.5892	0.716E-02	0.162E-03	0.218E-03	0.933E-10
6.	0.171E+01	1.6148	0.728E-02	0.115E-03	0.156E-03	0.802E-10
7.	0.174E+01	1.6373	0.738E-02	0.101E-03	0.137E-03	0.707E-10
14.	0.185E+01	1.7446	0.786E-02	0.691E-04	0.933E-04	0.401E-10
21.	0.199E+01	1.8778	0.846E-02	0.857E-04	0.116E-03	0.310E-10
28.	0.216E+01	2.0365	0.918E-02	0.102E-03	0.138E-03	0.274E-10
35.	0.232E+01	2.1920	0.988E-02	0.100E-03	0.135E-03	0.253E-10
42.	0.253E+01	2.3856	0.108E-01	0.125E-03	0.168E-03	0.250E-10
49.	0.277E+01	2.6091	0.119E-01	0.144E-03	0.194E-03	0.257E-10
56.	0.307E+01	2.8931	0.130E-01	0.183E-03	0.247E-03	0.276E-10
63.	0.331E+01	3.1262	0.141E-01	0.150E-03	0.203E-03	0.286E-10
93.	0.437E+01	4.1262	0.186E-01	0.150E-03	0.203E-03	0.338E-10

Table XII - LEACH TEST DATA - TEST SET A2 - ⁶⁰Co

TEST A3 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.126E+01	1.1858	0.534E-02	0.534E-02	0.722E-02	0.260E-09
2.	0.144E+01	1.3568	0.612E-02	0.770E-03	0.104E-02	0.170E-09
3.	0.149E+01	1.4036	0.633E-02	0.211E-03	0.285E-03	0.121E-09
4.	0.162E+01	1.5266	0.688E-02	0.554E-03	0.748E-03	0.108E-09
5.	0.169E+01	1.5927	0.718E-02	0.298E-03	0.402E-03	0.937E-10
6.	0.173E+01	1.6280	0.734E-02	0.159E-03	0.215E-03	0.816E-10
7.	0.174E+01	1.6430	0.741E-02	0.675E-04	0.911E-04	0.712E-10
14.	0.181E+01	1.7075	0.770E-02	0.415E-04	0.561E-04	0.385E-10
21.	0.191E+01	1.7996	0.811E-02	0.593E-04	0.800E-04	0.285E-10
28.	0.202E+01	1.9047	0.858E-02	0.677E-04	0.913E-04	0.239E-10
35.	0.218E+01	2.0570	0.927E-02	0.981E-04	0.132E-03	0.223E-10
42.	0.243E+01	2.2887	0.103E-01	0.149E-03	0.201E-03	0.230E-10
49.	0.270E+01	2.5511	0.115E-01	0.169E-03	0.228E-03	0.245E-10
56.	0.296E+01	2.7932	0.126E-01	0.156E-03	0.210E-03	0.257E-10
63.	0.323E+01	3.0503	0.137E-01	0.166E-03	0.224E-03	0.273E-10
93.	0.423E+01	3.9937	0.180E-01	0.142E-03	0.191E-03	0.317E-10

Table XIII - LEACH TEST DATA - TEST SET A3 - ^{60}Co

TEST B1 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.492E+00	0.5415	0.229E-02	0.229E-02	0.309E-02	0.477E-10
2.	0.608E+00	0.6689	0.283E-02	0.539E-03	0.727E-03	0.364E-10
3.	0.658E+00	0.7234	0.306E-02	0.231E-03	0.312E-03	0.284E-10
4.	0.698E+00	0.7674	0.324E-02	0.186E-03	0.251E-03	0.239E-10
5.	0.729E+00	0.8022	0.339E-02	0.147E-03	0.199E-03	0.209E-10
6.	0.757E+00	0.8333	0.352E-02	0.131E-03	0.177E-03	0.188E-10
7.	0.784E+00	0.8630	0.365E-02	0.125E-03	0.169E-03	0.173E-10
14.	0.960E+00	1.0565	0.447E-02	0.117E-03	0.158E-03	0.130E-10
21.	0.113E+01	1.2446	0.526E-02	0.114E-03	0.153E-03	0.120E-10
28.	0.125E+01	1.3798	0.583E-02	0.817E-04	0.110E-03	0.111E-10
35.	0.135E+01	1.4806	0.626E-02	0.609E-04	0.822E-04	0.102E-10
42.	0.144E+01	1.5856	0.670E-02	0.634E-04	0.857E-04	0.973E-11
49.	0.155E+01	1.7013	0.719E-02	0.699E-04	0.944E-04	0.960E-11
56.	0.165E+01	1.8130	0.767E-02	0.674E-04	0.911E-04	0.954E-11
63.	0.174E+01	1.9087	0.807E-02	0.578E-04	0.781E-04	0.940E-11
93.	0.204E+01	2.2418	0.948E-02	0.470E-04	0.634E-04	0.878E-11

Table XIV - LEACH TEST DATA - TEST SET B1 - ^{60}Co

TEST B2 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.175E+01	1.6500	0.744E-02	0.744E-02	0.100E-01	0.503E-09
2.	0.192E+01	1.8143	0.818E-02	0.741E-03	0.100E-02	0.304E-09
3.	0.199E+01	1.8815	0.848E-02	0.302E-03	0.408E-03	0.218E-09
4.	0.205E+01	1.9331	0.871E-02	0.233E-03	0.314E-03	0.173E-09
5.	0.207E+01	1.9550	0.881E-02	0.987E-04	0.133E-03	0.141E-09
6.	0.210E+01	1.9789	0.892E-02	0.107E-03	0.145E-03	0.121E-09
7.	0.211E+01	1.9943	0.899E-02	0.698E-04	0.942E-04	0.105E-09
14.	0.219E+01	2.0678	0.932E-02	0.473E-04	0.638E-04	0.564E-10
21.	0.226E+01	2.1288	0.959E-02	0.393E-04	0.531E-04	0.398E-10
28.	0.233E+01	2.1977	0.991E-02	0.444E-04	0.599E-04	0.319E-10
35.	0.239E+01	2.2528	0.102E-01	0.355E-04	0.479E-04	0.268E-10
42.	0.244E+01	2.2993	0.104E-01	0.299E-04	0.404E-04	0.232E-10
49.	0.249E+01	2.3479	0.106E-01	0.313E-04	0.423E-04	0.208E-10
56.	0.253E+01	2.3878	0.108E-01	0.257E-04	0.347E-04	0.188E-10
63.	0.256E+01	2.4114	0.109E-01	0.152E-04	0.205E-04	0.170E-10
93.	0.263E+01	2.4770	0.112E-01	0.984E-05	0.133E-04	0.122E-10

Table XV - LEACH TEST DATA - TEST SET B2 - ^{60}Co

TEST B3 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.145E+01	1.3054	0.599E-02	0.599E-02	0.810E-02	0.326E-09
2.	0.165E+01	1.4853	0.681E-02	0.825E-03	0.112E-02	0.211E-09
3.	0.175E+01	1.5759	0.723E-02	0.415E-03	0.562E-03	0.158E-09
4.	0.179E+01	1.6148	0.741E-02	0.179E-03	0.242E-03	0.125E-09
5.	0.181E+01	1.6292	0.747E-02	0.660E-04	0.893E-04	0.102E-09
6.	0.183E+01	1.6456	0.755E-02	0.752E-04	0.102E-03	0.864E-10
7.	0.184E+01	1.6551	0.759E-02	0.436E-04	0.590E-04	0.749E-10
14.	0.191E+01	1.7204	0.789E-02	0.428E-04	0.580E-04	0.405E-10
21.	0.198E+01	1.7813	0.817E-02	0.399E-04	0.540E-04	0.289E-10
28.	0.203E+01	1.8332	0.841E-02	0.340E-04	0.460E-04	0.230E-10
35.	0.207E+01	1.8660	0.856E-02	0.215E-04	0.290E-04	0.190E-10
42.	0.214E+01	1.9284	0.885E-02	0.409E-04	0.553E-04	0.169E-10
49.	0.220E+01	1.9811	0.909E-02	0.346E-04	0.468E-04	0.153E-10
56.	0.230E+01	2.0692	0.949E-02	0.577E-04	0.781E-04	0.146E-10
63.	0.234E+01	2.1088	0.967E-02	0.260E-04	0.351E-04	0.135E-10
93.	0.246E+01	2.2163	0.102E-01	0.165E-04	0.223E-04	0.101E-10

Table XVI - LEACH TEST DATA - TEST SET B3 - ^{60}Co

TEST C1 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.135E+01	1.3196	0.586E-02	0.586E-02	0.791E-02	0.312E-09
2.	0.144E+01	1.4130	0.627E-02	0.414E-03	0.560E-03	0.179E-09
3.	0.149E+01	1.4627	0.649E-02	0.221E-03	0.298E-03	0.128E-09
4.	0.151E+01	1.4833	0.658E-02	0.917E-04	0.124E-03	0.985E-10
5.	0.153E+01	1.4365	0.664E-02	0.585E-04	0.790E-04	0.802E-10
6.	0.153E+01	1.5043	0.668E-02	0.343E-04	0.464E-04	0.675E-10
7.	0.154E+01	1.5115	0.671E-02	0.321E-04	0.434E-04	0.584E-10
14.	0.157E+01	1.5398	0.683E-02	0.179E-04	0.242E-04	0.303E-10
21.	0.162E+01	1.5910	0.706E-02	0.325E-04	0.439E-04	0.216E-10
28.	0.166E+01	1.6240	0.721E-02	0.209E-04	0.283E-04	0.169E-10
35.	0.168E+01	1.6498	0.732E-02	0.163E-04	0.220E-04	0.139E-10
42.	0.172E+01	1.6820	0.746E-02	0.205E-04	0.276E-04	0.121E-10
49.	0.175E+01	1.7154	0.761E-02	0.212E-04	0.286E-04	0.108E-10
56.	0.179E+01	1.7580	0.780E-02	0.270E-04	0.365E-04	0.988E-11
63.	0.183E+01	1.7971	0.798E-02	0.248E-04	0.335E-04	0.918E-11
93.	0.190E+01	1.8661	0.828E-02	0.102E-04	0.138E-04	0.670E-11

Table XVII - LEACH TEST DATA - TEST SET C1 - ^{60}Co

TEST C2 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.470E+00	0.4610	0.204E-02	0.204E-02	0.276E-02	0.378E-10
2.	0.587E+00	0.5757	0.255E-02	0.507E-03	0.686E-03	0.295E-10
3.	0.658E+00	0.6448	0.285E-02	0.305E-03	0.413E-03	0.246E-10
4.	0.695E+00	0.6814	0.301E-02	0.162E-03	0.219E-03	0.206E-10
5.	0.724E+00	0.7095	0.314E-02	0.124E-03	0.168E-03	0.179E-10
6.	0.748E+00	0.7333	0.324E-02	0.105E-03	0.143E-03	0.159E-10
7.	0.762E+00	0.7475	0.331E-02	0.630E-04	0.852E-04	0.142E-10
14.	0.832E+00	0.8153	0.361E-02	0.428E-04	0.579E-04	0.844E-11
21.	0.903E+00	0.8855	0.392E-02	0.443E-04	0.600E-04	0.664E-11
28.	0.969E+00	0.9504	0.420E-02	0.410E-04	0.555E-04	0.573E-11
35.	0.103E+01	1.0136	0.448E-02	0.400E-04	0.541E-04	0.522E-11
42.	0.108E+01	1.0629	0.470E-02	0.311E-04	0.421E-04	0.478E-11
49.	0.114E+01	1.1189	0.495E-02	0.354E-04	0.479E-04	0.454E-11
56.	0.163E+01	1.6014	0.708E-02	0.305E-03	0.412E-03	0.814E-11
63.	0.169E+01	1.6600	0.734E-02	0.370E-04	0.501E-04	0.777E-11
93.	0.187E+01	1.8366	0.812E-02	0.260E-04	0.352E-04	0.645E-11

Table XVIII - LEACH TEST DATA - TEST SET C2 - ^{60}Co

TEST C3 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.108E+01	0.9899	0.451E-02	0.451E-02	0.610E-02	0.185E-09
2.	0.119E+01	1.0883	0.496E-02	0.448E-03	0.607E-03	0.112E-09
3.	0.125E+01	1.1436	0.521E-02	0.252E-03	0.341E-03	0.822E-10
4.	0.127E+01	1.1666	0.531E-02	0.105E-03	0.141E-03	0.642E-10
5.	0.128E+01	1.1772	0.536E-02	0.484E-04	0.655E-04	0.523E-10
6.	0.130E+01	1.1889	0.542E-02	0.535E-04	0.724E-04	0.444E-10
7.	0.131E+01	1.2000	0.547E-02	0.504E-04	0.682E-04	0.388E-10
14.	0.135E+01	1.2378	0.564E-02	0.246E-04	0.333E-04	0.206E-10
21.	0.142E+01	1.3043	0.594E-02	0.432E-04	0.585E-04	0.153E-10
28.	0.145E+01	1.3344	0.608E-02	0.196E-04	0.266E-04	0.120E-10
35.	0.149E+01	1.3652	0.622E-02	0.200E-04	0.271E-04	0.100E-10
42.	0.152E+01	1.3900	0.633E-02	0.162E-04	0.219E-04	0.868E-11
49.	0.155E+01	1.4196	0.647E-02	0.193E-04	0.261E-04	0.776E-11
56.	0.164E+01	1.5077	0.687E-02	0.573E-04	0.776E-04	0.766E-11
63.	0.167E+01	1.5306	0.697E-02	0.149E-04	0.202E-04	0.702E-11
93.	0.171E+01	1.5707	0.716E-02	0.609E-05	0.824E-05	0.500E-11

Table XIX - LEACH TEST DATA - TEST SET C3 - ^{60}Co

TEST D1 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.115E+01	1.1184	0.498E-02	0.498E-02	0.672E-02	0.226E-09
2.	0.126E+01	1.2206	0.544E-02	0.455E-03	0.613E-03	0.134E-09
3.	0.131E+01	1.2711	0.566E-02	0.225E-03	0.303E-03	0.971E-10
4.	0.133E+01	1.2905	0.575E-02	0.864E-04	0.116E-03	0.751E-10
5.	0.135E+01	1.3109	0.584E-02	0.911E-04	0.123E-03	0.620E-10
6.	0.137E+01	1.3313	0.593E-02	0.908E-04	0.122E-03	0.533E-10
7.	0.139E+01	1.3491	0.601E-02	0.792E-04	0.107E-03	0.469E-10
14.	0.145E+01	1.4078	0.627E-02	0.373E-04	0.503E-04	0.255E-10
21.	0.152E+01	1.4749	0.657E-02	0.427E-04	0.576E-04	0.187E-10
28.	0.156E+01	1.5172	0.676E-02	0.269E-04	0.363E-04	0.148E-10
35.	0.159E+01	1.5427	0.687E-02	0.162E-04	0.219E-04	0.123E-10
42.	0.162E+01	1.5767	0.702E-02	0.216E-04	0.291E-04	0.107E-10
49.	0.165E+01	1.6065	0.716E-02	0.190E-04	0.256E-04	0.950E-11
56.	0.172E+01	1.6667	0.742E-02	0.383E-04	0.517E-04	0.895E-11
63.	0.173E+01	1.6793	0.748E-02	0.802E-05	0.108E-04	0.807E-11
93.	0.178E+01	1.7261	0.769E-02	0.695E-05	0.937E-05	0.578E-11

Table XX - LEACH TEST DATA - TEST SET D1 - ^{60}Co

TEST D2 CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.120E+01	1.1558	0.515E-02	0.515E-02	0.697E-02	0.241E-09
2.	0.135E+01	1.2984	0.579E-02	0.636E-03	0.860E-03	0.152E-09
3.	0.142E+01	1.3684	0.610E-02	0.312E-03	0.422E-03	0.113E-09
4.	0.145E+01	1.3980	0.623E-02	0.132E-03	0.178E-03	0.882E-10
5.	0.150E+01	1.4386	0.641E-02	0.181E-03	0.245E-03	0.747E-10
6.	0.153E+01	1.4747	0.657E-02	0.161E-03	0.218E-03	0.655E-10
7.	0.155E+01	1.4924	0.665E-02	0.793E-04	0.107E-03	0.575E-10
14.	0.163E+01	1.5681	0.699E-02	0.482E-04	0.651E-04	0.317E-10
21.	0.171E+01	1.6461	0.734E-02	0.497E-04	0.672E-04	0.233E-10
28.	0.177E+01	1.6993	0.757E-02	0.339E-04	0.458E-04	0.186E-10
35.	0.182E+01	1.7533	0.781E-02	0.343E-04	0.465E-04	0.159E-10
42.	0.186E+01	1.7920	0.799E-02	0.247E-04	0.334E-04	0.138E-10
49.	0.192E+01	1.8500	0.825E-02	0.369E-04	0.500E-04	0.126E-10
56.	0.196E+01	1.8852	0.840E-02	0.224E-04	0.303E-04	0.115E-10
63.	0.198E+01	1.9053	0.849E-02	0.128E-04	0.173E-04	0.104E-10
93.	0.204E+01	1.9639	0.875E-02	0.870E-05	0.118E-04	0.749E-11

Table XXI - LEACH TEST DATA - TEST SET D2 - ^{60}Co

TEST D3 - CO-60

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.308E+00	0.3050	0.135E-02	0.135E-02	0.182E-02	0.165E-10
2.	0.391E+00	0.3875	0.171E-02	0.365E-03	0.491E-03	0.133E-10
3.	0.432E+00	0.4280	0.189E-02	0.179E-03	0.241E-03	0.109E-10
4.	0.451E+00	0.4470	0.198E-02	0.844E-04	0.114E-03	0.888E-11
5.	0.474E+00	0.4691	0.207E-02	0.977E-04	0.132E-03	0.782E-11
6.	0.499E+00	0.4945	0.219E-02	0.112E-03	0.151E-03	0.724E-11
7.	0.533E+00	0.5278	0.233E-02	0.147E-03	0.198E-03	0.707E-11
14.	0.638E+00	0.6319	0.279E-02	0.658E-04	0.886E-04	0.507E-11
21.	0.731E+00	0.7237	0.320E-02	0.580E-04	0.780E-04	0.443E-11
28.	0.807E+00	0.7994	0.353E-02	0.478E-04	0.644E-04	0.406E-11
35.	0.867E+00	0.8585	0.380E-02	0.374E-04	0.503E-04	0.374E-11
42.	0.933E+00	0.9234	0.408E-02	0.409E-04	0.551E-04	0.361E-11
49.	0.103E+01	1.0246	0.453E-02	0.640E-04	0.861E-04	0.381E-11
56.	0.109E+01	1.0835	0.479E-02	0.372E-04	0.501E-04	0.373E-11
63.	0.113E+01	1.1161	0.494E-02	0.206E-04	0.277E-04	0.351E-11
93.	0.122E+01	1.2043	0.533E-02	0.130E-04	0.175E-04	0.277E-11

Table XXII - LEACH TEST DATA - TEST SET D3 - ^{60}Co

TEST A1 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.289E+01	1.5712	0.716E-02	0.716E-02	0.968E-02	0.466E-09
2.	0.316E+01	1.7194	0.784E-02	0.676E-03	0.913E-03	0.279E-09
3.	0.326E+01	1.7718	0.808E-02	0.239E-03	0.323E-03	0.198E-09
4.	0.332E+01	1.8022	0.821E-02	0.138E-03	0.187E-03	0.153E-09
5.	0.335E+01	1.8205	0.830E-02	0.831E-04	0.112E-03	0.125E-09
6.	0.337E+01	1.8329	0.835E-02	0.567E-04	0.767E-04	0.106E-09
7.	0.339E+01	1.8427	0.840E-02	0.448E-04	0.606E-04	0.916E-10
14.	0.347E+01	1.8858	0.860E-02	0.281E-04	0.379E-04	0.480E-10
21.	0.363E+01	1.9703	0.898E-02	0.550E-04	0.743E-04	0.349E-10
28.	0.381E+01	2.0730	0.945E-02	0.669E-04	0.904E-04	0.290E-10
35.	0.397E+01	2.1559	0.983E-02	0.540E-04	0.729E-04	0.251E-10
42.	0.414E+01	2.2477	0.102E-01	0.598E-04	0.808E-04	0.227E-10
49.	0.437E+01	2.3738	0.108E-01	0.821E-04	0.111E-03	0.217E-10
56.	0.463E+01	2.5165	0.115E-01	0.929E-04	0.126E-03	0.214E-10
63.	0.486E+01	2.6417	0.120E-01	0.816E-04	0.110E-03	0.209E-10
93.	0.575E+01	3.1259	0.142E-01	0.736E-04	0.994E-04	0.198E-10

Table XXIII - LEACH TEST DATA - TEST SET A1 - ^{137}Cs

TEST A2 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.681E+01	3.8286	0.173E-01	0.173E-01	0.233E-01	0.271E-08
2.	0.761E+01	4.2732	0.193E-01	0.200E-02	0.270E-02	0.169E-08
3.	0.789E+01	4.4310	0.200E-01	0.711E-03	0.960E-03	0.121E-08
4.	0.803E+01	4.5136	0.203E-01	0.372E-03	0.503E-03	0.940E-09
5.	0.812E+01	4.5631	0.206E-01	0.223E-03	0.301E-03	0.769E-09
6.	0.818E+01	4.5959	0.207E-01	0.148E-03	0.200E-03	0.650E-09
7.	0.825E+01	4.6363	0.209E-01	0.182E-03	0.246E-03	0.567E-09
14.	0.858E+01	4.8206	0.217E-01	0.119E-03	0.160E-03	0.307E-09
21.	0.897E+01	5.0373	0.227E-01	0.140E-03	0.188E-03	0.223E-09
28.	0.943E+01	5.2976	0.239E-01	0.168E-03	0.226E-03	0.185E-09
35.	0.990E+01	5.5622	0.251E-01	0.170E-03	0.230E-03	0.163E-09
42.	0.105E+02	5.9154	0.267E-01	0.227E-03	0.307E-03	0.154E-09
49.	0.113E+02	6.3448	0.286E-01	0.276E-03	0.373E-03	0.152E-09
56.	0.122E+02	6.8690	0.310E-01	0.337E-03	0.456E-03	0.156E-09
63.	0.130E+02	7.3066	0.329E-01	0.282E-03	0.380E-03	0.156E-09
93.	0.164E+02	9.2055	0.415E-01	0.285E-03	0.385E-03	0.168E-09

Table XXIV - LEACH TEST DATA - TEST SET A2 - ^{137}Cs

TEST - A3 CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.421E+01	2.3652	0.107E-01	0.107E-01	0.144E-01	0.103E-08
2.	0.460E+01	2.5825	0.116E-01	0.980E-03	0.132E-02	0.616E-09
3.	0.475E+01	2.6685	0.120E-01	0.388E-03	0.523E-03	0.438E-09
4.	0.482E+01	2.7080	0.122E-01	0.178E-03	0.240E-03	0.339E-09
5.	0.486E+01	2.7309	0.123E-01	0.103E-03	0.139E-03	0.275E-09
6.	0.489E+01	2.7490	0.124E-01	0.817E-04	0.110E-03	0.233E-09
7.	0.492E+01	2.7629	0.125E-01	0.627E-04	0.846E-04	0.201E-09
14.	0.507E+01	2.8500	0.128E-01	0.561E-04	0.757E-04	0.107E-09
21.	0.530E+01	2.9800	0.134E-01	0.837E-04	0.113E-03	0.781E-10
28.	0.555E+01	3.1178	0.141E-01	0.887E-04	0.120E-03	0.641E-10
35.	0.594E+01	3.3347	0.150E-01	0.140E-03	0.189E-03	0.587E-10
42.	0.667E+01	3.7482	0.169E-01	0.266E-03	0.359E-03	0.618E-10
49.	0.739E+01	4.1528	0.187E-01	0.260E-03	0.352E-03	0.650E-10
56.	0.811E+01	4.5547	0.205E-01	0.259E-03	0.349E-03	0.684E-10
63.	0.887E+01	4.9819	0.225E-01	0.275E-03	0.371E-03	0.727E-10
93.	0.117E+02	6.5802	0.297E-01	0.240E-03	0.324E-03	0.860E-10

Table XXV - LEACH TEST DATA - TEST SET A3 - ^{137}Cs

TEST B1 -CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.119E+01	0.7822	0.331E-02	0.331E-02	0.447E-02	0.994E-10
2.	0.144E+01	0.9477	0.401E-02	0.700E-03	0.945E-03	0.730E-10
3.	0.159E+01	1.0438	0.441E-02	0.406E-03	0.549E-03	0.590E-10
4.	0.169E+01	1.1087	0.469E-02	0.275E-03	0.371E-03	0.499E-10
5.	0.176E+01	1.1555	0.489E-02	0.198E-03	0.267E-03	0.434E-10
6.	0.182E+01	1.2005	0.508E-02	0.190E-03	0.257E-03	0.390E-10
7.	0.189E+01	1.2429	0.526E-02	0.180E-03	0.242E-03	0.359E-10
14.	0.215E+01	1.4146	0.598E-02	0.104E-03	0.140E-03	0.232E-10
21.	0.234E+01	1.5392	0.651E-02	0.752E-04	0.102E-03	0.183E-10
28.	0.245E+01	1.6123	0.682E-02	0.442E-04	0.596E-04	0.151E-10
35.	0.253E+01	1.6633	0.703E-02	0.308E-04	0.416E-04	0.128E-10
42.	0.262E+01	1.7222	0.728E-02	0.356E-04	0.481E-04	0.115E-10
49.	0.272E+01	1.7916	0.758E-02	0.419E-04	0.566E-04	0.106E-10
56.	0.280E+01	1.8452	0.780E-02	0.324E-04	0.437E-04	0.988E-11
63.	0.286E+01	1.8835	0.796E-02	0.231E-04	0.312E-04	0.915E-11
93.	0.303E+01	1.9950	0.844E-02	0.157E-04	0.212E-04	0.696E-11

Table XXVI - LEACH TEST DATA - TEST SET B1 - ^{137}Cs

TEST B2 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.477E+01	2.6815	0.121E-01	0.121E-01	0.163E-01	0.133E-08
2.	0.521E+01	2.9281	0.132E-01	0.111E-02	0.150E-02	0.792E-09
3.	0.540E+01	3.0349	0.137E-01	0.481E-03	0.650E-03	0.567E-09
4.	0.552E+01	3.1039	0.140E-01	0.311E-03	0.420E-03	0.445E-09
5.	0.557E+01	3.1298	0.141E-01	0.116E-03	0.157E-03	0.362E-09
6.	0.563E+01	3.1628	0.143E-01	0.149E-03	0.201E-03	0.308E-09
7.	0.567E+01	3.1826	0.143E-01	0.892E-04	0.120E-03	0.267E-09
14.	0.587E+01	3.2998	0.149E-01	0.754E-04	0.102E-03	0.144E-09
21.	0.604E+01	3.3934	0.153E-01	0.603E-04	0.814E-04	0.101E-09
28.	0.624E+01	3.5029	0.158E-01	0.705E-04	0.952E-04	0.809E-10
35.	0.640E+01	3.5947	0.162E-01	0.591E-04	0.798E-04	0.682E-10
42.	0.652E+01	3.6644	0.165E-01	0.449E-04	0.606E-04	0.590E-10
49.	0.664E+01	3.7307	0.168E-01	0.426E-04	0.576E-04	0.524E-10
56.	0.675E+01	3.7903	0.171E-01	0.384E-04	0.519E-04	0.474E-10
63.	0.682E+01	3.8289	0.173E-01	0.248E-04	0.335E-04	0.430E-10
93.	0.702E+01	3.9463	0.178E-01	0.176E-04	0.238E-04	0.309E-10

Table XXVII - LEACH TEST DATA - TEST SET B2 - ^{137}Cs

TEST B3 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.388E+01	2.0887	0.958E-02	0.958E-02	0.130E-01	0.835E-09
2.	0.435E+01	2.3396	0.107E-01	0.115E-02	0.156E-02	0.524E-09
3.	0.460E+01	2.4752	0.114E-01	0.622E-03	0.842E-03	0.391E-09
4.	0.469E+01	2.5195	0.116E-01	0.203E-03	0.275E-03	0.304E-09
5.	0.471E+01	2.5336	0.116E-01	0.648E-04	0.877E-04	0.246E-09
6.	0.475E+01	2.5538	0.117E-01	0.925E-04	0.125E-03	0.208E-09
7.	0.477E+01	2.5649	0.118E-01	0.510E-04	0.691E-04	0.180E-09
14.	0.493E+01	2.6480	0.121E-01	0.545E-04	0.737E-04	0.958E-10
21.	0.507E+01	2.7252	0.125E-01	0.506E-04	0.685E-04	0.677E-10
28.	0.519E+01	2.7886	0.128E-01	0.415E-04	0.562E-04	0.531E-10
35.	0.526E+01	2.8272	0.130E-01	0.253E-04	0.342E-04	0.437E-10
42.	0.541E+01	2.9089	0.133E-01	0.535E-04	0.724E-04	0.385E-10
49.	0.550E+01	2.9581	0.136E-01	0.323E-04	0.437E-04	0.342E-10
56.	0.574E+01	3.0841	0.141E-01	0.826E-04	0.112E-03	0.325E-10
63.	0.585E+01	3.1428	0.144E-01	0.384E-04	0.520E-04	0.300E-10
93.	0.621E+01	3.3376	0.153E-01	0.298E-04	0.403E-04	0.229E-10

Table XXVIII - LEACH TEST DATA - TEST SET B3 - ^{137}Cs

TEST C1 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.290E+01	1.6936	0.752E-02	0.752E-02	0.102E-01	0.514E-09
2.	0.310E+01	1.8112	0.804E-02	0.522E-03	0.705E-03	0.294E-09
3.	0.320E+01	1.8719	0.831E-02	0.269E-03	0.364E-03	0.209E-09
4.	0.324E+01	1.8951	0.841E-02	0.103E-03	0.140E-03	0.161E-09
5.	0.326E+01	1.9091	0.847E-02	0.621E-04	0.838E-04	0.131E-09
6.	0.328E+01	1.9167	0.851E-02	0.336E-04	0.454E-04	0.110E-09
7.	0.329E+01	1.9248	0.854E-02	0.359E-04	0.485E-04	0.948E-10
14.	0.335E+01	1.9610	0.870E-02	0.230E-04	0.310E-04	0.492E-10
21.	0.346E+01	2.0226	0.898E-02	0.391E-04	0.528E-04	0.349E-10
28.	0.354E+01	2.0700	0.919E-02	0.300E-04	0.406E-04	0.274E-10
35.	0.360E+01	2.1065	0.935E-02	0.231E-04	0.312E-04	0.227E-10
42.	0.368E+01	2.1524	0.955E-02	0.291E-04	0.394E-04	0.198E-10
49.	0.376E+01	2.2013	0.977E-02	0.310E-04	0.419E-04	0.177E-10
56.	0.387E+01	2.2628	0.100E-01	0.390E-04	0.526E-04	0.164E-10
63.	0.397E+01	2.3199	0.103E-01	0.362E-04	0.489E-04	0.153E-10
93.	0.414E+01	2.4199	0.107E-01	0.148E-04	0.200E-04	0.113E-10

Table XXIX - LEACH TEST DATA - TEST SET C1 - ^{137}Cs

TEST C2 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.103E+01	0.6035	0.267E-02	0.267E-02	0.361E-02	0.647E-10
2.	0.120E+01	0.7047	0.312E-02	0.447E-03	0.605E-03	0.441E-10
3.	0.130E+01	0.7674	0.339E-02	0.277E-03	0.375E-03	0.349E-10
4.	0.136E+01	0.7986	0.353E-02	0.138E-03	0.187E-03	0.283E-10
5.	0.139E+01	0.8204	0.363E-02	0.963E-04	0.130E-03	0.239E-10
6.	0.143E+01	0.8419	0.372E-02	0.948E-04	0.128E-03	0.210E-10
7.	0.146E+01	0.8577	0.379E-02	0.700E-04	0.948E-04	0.187E-10
14.	0.160E+01	0.9394	0.415E-02	0.516E-04	0.698E-04	0.112E-10
21.	0.172E+01	1.0129	0.448E-02	0.464E-04	0.628E-04	0.868E-11
28.	0.182E+01	1.0721	0.474E-02	0.374E-04	0.506E-04	0.730E-11
35.	0.192E+01	1.1297	0.500E-02	0.364E-04	0.493E-04	0.648E-11
42.	0.199E+01	1.1711	0.518E-02	0.262E-04	0.354E-04	0.580E-11
49.	0.213E+01	1.2509	0.553E-02	0.504E-04	0.681E-04	0.568E-11
56.	0.233E+01	1.3713	0.606E-02	0.761E-04	0.103E-03	0.597E-11
63.	0.240E+01	1.4110	0.624E-02	0.250E-04	0.339E-04	0.562E-11
93.	0.258E+01	1.5158	0.670E-02	0.155E-04	0.209E-04	0.439E-11

Table XXX - LEACH TEST DATA - TEST SET C2 - ^{137}Cs

TEST C3 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.288E+01	1.5732	0.717E-02	0.717E-02	0.970E-02	0.467E-09
2.	0.317E+01	1.7305	0.788E-02	0.716E-03	0.970E-03	0.282E-09
3.	0.331E+01	1.8105	0.825E-02	0.364E-03	0.493E-03	0.206E-09
4.	0.336E+01	1.8379	0.837E-02	0.125E-03	0.169E-03	0.159E-09
5.	0.339E+01	1.8500	0.843E-02	0.551E-04	0.746E-04	0.129E-09
6.	0.341E+01	1.8631	0.849E-02	0.597E-04	0.809E-04	0.109E-09
7.	0.343E+01	1.8767	0.855E-02	0.621E-04	0.841E-04	0.949E-10
14.	0.353E+01	1.9314	0.880E-02	0.355E-04	0.481E-04	0.503E-10
21.	0.370E+01	2.0239	0.922E-02	0.602E-04	0.815E-04	0.368E-10
28.	0.377E+01	2.0611	0.939E-02	0.242E-04	0.328E-04	0.286E-10
35.	0.385E+01	2.1038	0.958E-02	0.278E-04	0.376E-04	0.239E-10
42.	0.391E+01	2.1374	0.974E-02	0.219E-04	0.296E-04	0.205E-10
49.	0.399E+01	2.1803	0.993E-02	0.279E-04	0.377E-04	0.183E-10
56.	0.420E+01	2.2976	0.105E-01	0.764E-04	0.103E-03	0.178E-10
63.	0.427E+01	2.3319	0.106E-01	0.223E-04	0.302E-04	0.163E-10
93.	0.438E+01	2.3941	0.109E-01	0.944E-05	0.128E-04	0.116E-10

Table XXXI - LEACH TEST DATA - TEST SET C3 - 137Cs

TEST D1 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.311E+01	1.8064	0.805E-02	0.805E-02	0.108E-01	0.588E-09
2.	0.341E+01	1.9822	0.883E-02	0.783E-03	0.106E-02	0.354E-09
3.	0.356E+01	2.0713	0.923E-02	0.397E-03	0.535E-03	0.258E-09
4.	0.363E+01	2.1131	0.941E-02	0.186E-03	0.251E-03	0.201E-09
5.	0.369E+01	2.1463	0.956E-02	0.148E-03	0.200E-03	0.166E-09
6.	0.372E+01	2.1644	0.964E-02	0.803E-04	0.108E-03	0.141E-09
7.	0.375E+01	2.1822	0.972E-02	0.793E-04	0.107E-03	0.123E-09
14.	0.389E+01	2.2614	0.101E-01	0.504E-04	0.680E-04	0.659E-10
21.	0.403E+01	2.3456	0.104E-01	0.536E-04	0.722E-04	0.472E-10
28.	0.413E+01	2.4003	0.107E-01	0.348E-04	0.469E-04	0.371E-10
35.	0.420E+01	2.4398	0.109E-01	0.251E-04	0.339E-04	0.307E-10
42.	0.427E+01	2.4851	0.111E-01	0.288E-04	0.389E-04	0.265E-10
49.	0.435E+01	2.5276	0.113E-01	0.271E-04	0.365E-04	0.235E-10
56.	0.449E+01	2.6091	0.116E-01	0.518E-04	0.699E-04	0.219E-10
63.	0.452E+01	2.6254	0.117E-01	0.104E-04	0.140E-04	0.197E-10
93.	0.461E+01	2.6796	0.119E-01	0.805E-05	0.109E-04	0.139E-10

Table XXXII - LEACH TEST DATA - TEST SET D1 - 137Cs

TEST D2 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.368E+01	2.1138	0.942E-02	0.942E-02	0.127E-01	0.807E-09
2.	0.411E+01	2.3629	0.105E-01	0.111E-02	0.150E-02	0.504E-09
3.	0.434E+01	2.4947	0.111E-01	0.587E-03	0.794E-03	0.375E-09
4.	0.445E+01	2.5577	0.114E-01	0.281E-03	0.380E-03	0.295E-09
5.	0.456E+01	2.6180	0.117E-01	0.269E-03	0.363E-03	0.248E-09
6.	0.460E+01	2.6447	0.118E-01	0.119E-03	0.161E-03	0.211E-09
7.	0.464E+01	2.6665	0.119E-01	0.971E-04	0.131E-03	0.183E-09
14.	0.484E+01	2.7815	0.124E-01	0.732E-04	0.990E-04	0.998E-10
21.	0.505E+01	2.9004	0.129E-01	0.757E-04	0.102E-03	0.723E-10
28.	0.517E+01	2.9691	0.132E-01	0.437E-04	0.591E-04	0.569E-10
35.	0.530E+01	3.0436	0.136E-01	0.474E-04	0.641E-04	0.478E-10
42.	0.539E+01	3.0973	0.138E-01	0.342E-04	0.463E-04	0.412E-10
49.	0.553E+01	3.1764	0.142E-01	0.504E-04	0.682E-04	0.372E-10
56.	0.560E+01	3.2207	0.144E-01	0.282E-04	0.382E-04	0.335E-10
63.	0.565E+01	3.2468	0.145E-01	0.166E-04	0.224E-04	0.302E-10
93.	0.580E+01	3.3314	0.148E-01	0.126E-04	0.170E-04	0.216E-10

Table XXXIII - LEACH TEST DATA - TEST SET D2 - ^{137}Cs

TEST D3 - CS-137

TOTAL TIME DAYS	TOTAL ACTIVITY RELEASED MICROCURIES	FRACTIONAL RELEASED PERCENT	CUMULATIVE FRACTION LEACHED CM	INCREMENTAL LEACH RATE CM/DAY	MASS LEACH RATE G/CM**2*DAY	DIFFUSION COEFFICIENT CM**2/SEC
1.	0.111E+01	0.6547	0.290E-02	0.290E-02	0.390E-02	0.762E-10
2.	0.136E+01	0.8022	0.355E-02	0.652E-03	0.878E-03	0.572E-10
3.	0.148E+01	0.8709	0.385E-02	0.304E-03	0.409E-03	0.449E-10
4.	0.154E+01	0.9087	0.402E-02	0.167E-03	0.225E-03	0.367E-10
5.	0.160E+01	0.9405	0.416E-02	0.141E-03	0.189E-03	0.314E-10
6.	0.164E+01	0.9647	0.427E-02	0.107E-03	0.145E-03	0.276E-10
7.	0.170E+01	1.0014	0.443E-02	0.162E-03	0.219E-03	0.255E-10
14.	0.195E+01	1.1450	0.506E-02	0.907E-04	0.122E-03	0.166E-10
21.	0.214E+01	1.2616	0.558E-02	0.736E-04	0.991E-04	0.135E-10
28.	0.230E+01	1.3550	0.599E-02	0.590E-04	0.795E-04	0.117E-10
35.	0.243E+01	1.4309	0.633E-02	0.479E-04	0.645E-04	0.104E-10
42.	0.256E+01	1.5071	0.666E-02	0.482E-04	0.648E-04	0.961E-11
49.	0.278E+01	1.6351	0.723E-02	0.808E-04	0.109E-03	0.970E-11
56.	0.289E+01	1.6986	0.751E-02	0.401E-04	0.540E-04	0.916E-11
63.	0.295E+01	1.7377	0.768E-02	0.247E-04	0.333E-04	0.852E-11
93.	0.313E+01	1.8440	0.815E-02	0.157E-04	0.211E-04	0.650E-11

Table XXXIV - LEACH TEST DATA - TEST SET D3 - ^{137}Cs

APPENDIX D - LEACHANT CONDUCTIVITY AND PH

	Initial	Final 1 Week's contact
<u>Set A</u>		
pH	4.10	5.80
Conductivity	$1.00 \times 10^{-6} \text{ Mho/cm}$	$4.90 \times 10^{-4} \text{ Mho/cm}$
<u>Set B</u>		
pH	6.39	6.25
Conductivity	$4.80 \times 10^{-3} \text{ Mho/cm}$	$6.5 \times 10^{-3} \text{ Mho/cm}$
<u>Set C</u>		
pH	6.39	5.65
Conductivity	$4.80 \times 10^{-3} \text{ Mho/cm}$	$5.5 \times 10^{-3} \text{ Mho/cm}$
<u>Set D</u>		
ph	5.62	4.99
Conductivity	$1.85 \times 10^{-2} \text{ Mho/cm}$	$1.95 \times 10^{-2} \text{ Mho/cm}$

APPENDIX E - ANALYSIS OF VARIANCE OF ^{60}Co AND ^{137}Cs -SAMPLE MEANS
AT T \geq 14DAYS

^{60}Co :

<u>Set</u>	<u>Mean</u>	<u>St Deviation</u>	<u>St Error</u>	<u>95% Confidence Interval for Mean</u>
A	0.0083	0.0050	0.0010	0.0064 - 0.0103
B	0.0087	0.0018	0.0003	0.0080 - 0.0094
C	0.0064	0.0013	0.0003	0.0059 - 0.0070
D	0.0064	0.0018	0.0003	0.0057 - 0.0071

F Ratio=4.856

Homogenous subsets:

Subset 1

Group	Set D	Set C
Mean	0.0064	0.0064

Subset 2

Group	Set A	Set B
Mean	0.0083	0.0087

^{137}Cs :

<u>Set</u>	<u>Mean</u>	<u>St Deviation</u>	<u>St Error</u>	<u>95% Confidence Interval for Mean</u>
A	0.0190	0.0087	0.0017	0.0156 - 0.0225
B	0.0124	0.0040	0.0008	0.0108 - 0.0140
C	0.0083	0.0022	0.0004	0.0074 - 0.0092
D	0.0105	0.0031	0.0006	0.0093 - 0.0117

F Ratio=21.756

Homogenous Subsets:

Subset 1

Group	Set C	Set D
Mean	0.0083	0.0105

Subset 2

Group	Set D	Set B
Mean	0.0105	0.0124

Subset 3

Group	Set A
Mean	0.0190