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**ANALYSIS OF RADIONUCLIDE RELEASE
FROM LOW-LEVEL RADIOACTIVE WASTE REPOSITORY**

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ABSTRACT

The analytical repository source term (REPS) computer code is developed for the safety assessment of radioactive waste repository. For reliable prediction of the leach rates for various radionuclides, degradation of concrete structures, corrosion rate of waste container, degree of corrosion on the container surface, and the characteristics of radionuclides are considered in the REPS code. A simplified safety assessment is carried out on rock-cavern type disposal of LLW. The results of preliminary assessment show that Cs-137, Ni-63, and Sr-90 are dominant. For the parametric uncertainty and sensitivity analysis, Latin hypercube sampling and rank correlation techniques are applied. The results of the potential public health impacts show that radiological doses to general public are negligible and that near-field performance should be given more attention.

REPS

INTRODUCTION

Korea has nine nuclear power plants (8 PWRs and 1 CANDU) in operation with generating capacity of 7,616 MWe. In addition, two PWRs are under construction, three units (1 CANDU and 2 PWRs) are under design, and five more CANDUs and eight more PWRs are planned to be completed by 2006. The cumulative amount of operational waste from nuclear power plants in Korea amounted to about 34,000 drums by the end of 1991 and will reach up to about 105,000 drums by 2000 as shown in Figure 1. A centralized repository for low-level radioactive waste (LLW) is scheduled to be constructed by the end of 1995 with an initial capacity of 50,000 drums. The subsequent expansions are also scheduled to increase the capacity to 250,000 drums, and eventually up to 1 million drums.

The potential public health impacts associated with rock-cavern type disposal of LLW have been preliminarily assessed to ensure that radiological doses to persons who might be exposed do not exceed any regulatory limits.

The performance of far field is difficult to be predicted conclusively because of the high demands on site characterization. Considering the uncertainty regarding performance assessment of far field, it is desirable at this moment to analyze near-field performance for a preliminary performance assessment. This paper presents the analytical repository source term (REPS) computer code developed for the performance assessment of geologic repository.

METHODOLOGY

Process Description.

Most low-level radioactive wastes from nuclear power plants in Korea are processed by cement solidification. The objective of the low-level radioactive waste repository source term model is to develop a system model capable of predicting radionuclide release rates from cemented waste drums disposed of in underground repository. Repository source-term model will be used for the performance assessment of geologic repository as described in Figure 2. The system model is divided in three compartments: degradation of concrete structures, corrosion of waste container, and waste

form leaching. Each of these compartments is described by submodels which will be coupled into the system model.

A primary objective of the REPS modeling is to make the solution procedure flexible enough to allow incorporation of new models to represent alternative disposal methods for high-level radioactive waste while retaining the basic procedures used for modeling low-level radioactive waste disposal. Therefore, the computer code is structured by a series of modules that represent physical processes. Figure 3 shows the concepts of the REPS code.

No penetration of groundwater may occur while the concrete structure retains its integrity. The degradation of a reinforced concrete depends on environmental characteristics of a specific disposal site. In this preliminary assessment, it is conservatively assumed that groundwater can infiltrate into the repository soon after the repository closure.

Corrosion Model

The primary cause for a loss of containment ability of steel container is corrosion. Corrosion of the radioactive waste container may be predicted from mechanistic models or empirical models. Several mechanistic models for corrosion have been developed (1,2,3). However, empirical models are preferred to mechanistic models to predict the container corrosion rate because the corrosion process of steel depends on too many parameters such as groundwater Eh, pH, major ions present, type of steel, location of defects in the steel, surface impurities, etc. For this reason, the corrosion model of REPS is empirical in nature and uses a corrosion data base. The general form of corrosion depth equation is

$$d = k t^n \quad [1]$$

where d is the corrosion depth, k is a corrosion rate constant from the corrosion data base, t is time, and n is an empirical constant which gives the time dependence of corrosion depth. Modelers often set n equal to 1 for long-term corrosion depth prediction.

The degree of corrosion, C_R , that is defined as the ratio of the surface area of the waste form exposed to water to the total surface area of a container, is assumed to be

expressed by the following experimental formula (4)

$$C_R(t) = \exp(\alpha + \beta t) / (1 + \exp(\alpha + \beta t)) \quad [2]$$

where α and β are empirical constants determined by fitting experimental data to eq.[2].

Leaching Mechanisms

Leach rates, i.e., the rates at which radionuclides are released from the solid waste form into the contacting groundwater, constitute the source term to radionuclide hydro-geological transport models. The analysis is based on time-dependent leaching at constant temperature, appropriate for non-heat-generating low- and intermediate-level radioactive wastes. Three leaching mechanisms included in the REPS model are solubility-limited release, congruent release, and solid diffusion controlled release.

• Solubility-Limited Release

The equation for the solubility-limited diffusive release of radionuclide i from a spherical waste form surface is

$$K_i \frac{\partial C_i(r, \tau)}{\partial \tau} = D \frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 \frac{\partial C_i(r, \tau)}{\partial r} \right] - \lambda_i K_i C_i(r, \tau); \quad r > r_0, \tau > 0 \quad [3]$$

The initial condition is

$$C_i(r, 0) = 0, \quad r > r_0 \quad [4]$$

The boundary conditions are

$$C_i(r_0, \tau) = C_{sc} \gamma_i, \quad \tau > 0 \quad [5]$$

$$C_i(\infty, \tau) = 0, \quad \tau > 0 \quad [6]$$

where $C_i(r, \tau)$ is the concentration of radionuclide i in the groundwater, g/m^3

K_i is the retardation coefficient of radionuclide i , dimensionless

D is the diffusion coefficient of radionuclide i , m^2/yr

λ_i is the decay constant of radionuclide i , yr^{-1}

r_0 is the radius of spherical waste form, m

C_{se} is the solubility of element e, g/m³

γ_i is the inventory ratio of isotope to element, dimensionless

and τ is the time after leaching begins, yr.

The desired solution of eqs.[3] - [6] is obtained using the Laplace transform and convolution theorem. (5)

$$C_i(r, \tau) = \frac{C_{se} \gamma_i r_0}{2r} \left\{ e^{(r-r_0)\sqrt{K_i \lambda_i / D}} \operatorname{erfc} \left[\frac{(r-r_0)}{2} \sqrt{K_i / (D \tau)} + \sqrt{\lambda_i \tau} \right] \right. \\ \left. + e^{-(r-r_0)\sqrt{K_i \lambda_i / D}} \operatorname{erfc} \left[\frac{(r-r_0)}{2} \sqrt{K_i / (D \tau)} - \sqrt{\lambda_i \tau} \right] \right\}, \quad r > r_0, \tau > 0 \quad [7]$$

The quantity of concern is the total mass flux $m_i(\tau)$ of species i from the waste form surface into the surrounding porous rock.

Since

$$m_i(\tau) = 4 \pi r_0^2 \left\{ -\epsilon D \frac{\partial C(r_0, \tau)}{\partial r} \right\}, \quad \tau > 0 \quad [8]$$

one obtains from eqs. [7] and [8]

$$m_i(\tau) = 4 \pi r_0 C_{se} \gamma_i D \epsilon \left\{ 1 + \sqrt{\frac{K_i r_0^2}{\pi D t}} e^{-\lambda_i \tau} \right. \\ \left. + \sqrt{\frac{\lambda_i K_i r_0^2}{D}} \operatorname{erf} \sqrt{\lambda_i \tau} \right\} \cdot \{h(\tau) - h(\tau - T_i)\}, \quad \text{g/yr ; } \tau > 0 \quad [9]$$

where $h(\tau)$ is the Heaviside step function and ϵ is the porosity of the diffusing medium.

The leach time T_i of radionuclide i is determined by the following equation

$$\frac{d M_i(\tau)}{d \tau} = -m_i(\tau) - \lambda_i M_i(\tau), \quad 0 < \tau \leq T_i \quad [10]$$

with side conditions

$$M_i(0) = M_i^0 \quad [11]$$

$$M_i(T_i) = 0 \quad [12]$$

where M_i^0 is the initial inventory of radionuclide i per a waste package, $M_i(\tau)$ is the inventory at time τ in the waste form, and $m_i(\tau)$ is the leach rate of radionuclide i at time τ .

• Inventory-Limited Release

For soluble species, for example, cesium, strontium, and iodine, one can only estimate the range of leach rates, since the solubilities of their usual compounds may be too large to limit their leaching. A lower limit to the leach rate would be the leach rate of the waste matrix, if they release congruently with the matrix. The congruently released species (or inventory-limited release) has the same fractional leach rate at any time τ after beginning of leaching as the waste matrix, if both leach rates are normalized to the instantaneous inventory in the undissolved waste. Then the leach rate $m_i(\tau)$ of the congruently released species i at time τ , normalized to the inventory $M_i(\tau)$ of species i at time τ is the same as the leach rate of the cement waste matrix at time τ , normalized to the inventory $M_c(\tau)$ of the cement waste matrix at time τ :

$$m_i(\tau) = 4 \pi r_0^2 L_c(\tau) \frac{M_i(\tau)}{M_c(\tau)} [h(\tau) - h(\tau - T_c)] \quad [13]$$

The leach rate L_c of cement per unit surface area of the waste form is to be obtained experimentally.

• Release Limited by Solid Diffusion

For extremely low solubility species such as Co-60, leaching is limited by the diffusive transport rate of the species in solid waste form. The diffusion equation in cylindrical geometry is

$$\frac{\partial C_i}{\partial \tau} = D \left[\frac{\partial^2 C_i}{\partial r^2} + \frac{1}{r} \frac{\partial C_i}{\partial r} + \frac{\partial^2 C_i}{\partial z^2} \right] - \lambda_i C_i, \quad 0 \leq r \leq a, \quad -b \leq z \leq b \quad [14]$$

where a is the radius of the cylindrical waste form, and b is half-height of the cylinder. The initial condition is

$$C_i(r, z, 0) = C_i^0 \quad [15]$$

where C_i^0 is an initial concentration. The boundary conditions are

$$C_i(a, z, \tau) = 0, \quad \tau > 0 \quad [16]$$

$$C_i(r, \pm b, \tau) = 0, \quad \tau > 0 \quad [17]$$

The solution of eqs.[14] - [17] was obtained by Nestor (6):

$$C_i = \frac{8C_i^0}{\pi a} e^{-\lambda_i \tau} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{(-1)^n J_0(r \alpha_{in})}{(2n-1) \alpha_m J_1(a \alpha_m)} \cos \beta_n z \exp[-D \tau (\alpha_m^2 + \beta_n^2)] \quad [18]$$

where J_0 is the zero-order Bessel function,

J_1 is the first-order Bessel function,

α_m is the positive root of $J_0(a \alpha_m) = 0$,

and $\beta_n = (2n-1) \pi / 2b$.

Finally the time-dependent diffusion-controlled leach rate of species i from the surface of a cylindrical waste form is

$$m_i(\tau) = 16C_i^0 D e^{-\lambda_i \tau} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \left[\frac{4b}{(2n-1)^2 \pi} + \frac{\pi}{b \alpha_m^2} \right] \exp[-D \tau (\alpha_m^2 + \beta_n^2)] \{h(\tau) - h(\tau - T_i)\} \quad [19]$$

Leach Model

The fractional release rate $f_i(t)$ as a function of time t after emplacement from a waste drum to surrounding porous medium is

$$f_i(t) = \frac{m_i(t; t_0, T_i) C_R(t)}{M_i^0} \quad [20]$$

where $m_i(t; t_0, T_i)$ is the leach rate of radionuclide i from a waste form at time t after emplacement,

t_0 is the container penetration time or the starting time of leaching after emplacement,

T_i is the leach time or the duration of leaching for radionuclide i ,

and $C_R(t)$ is the exposed surface area ratio given in eq.[2].

$m_i(t; t_0, T_i)$ has a non-zero value for $t_0 \leq t \leq T_i + t_0$.

For three leaching models included in the REPS code, the leach rate $m_i(t; t_0, T_i)$ is as follows:

- Solubility-limited model

From eq.[9]

$$m_i(t ; t_0, T_i) = 4\pi r_0 C_{se} \gamma_i D \epsilon \left\{ 1 + \sqrt{\frac{K_i r_0^2}{\pi D (t - t_0)}} e^{-\lambda_i (t - t_0)} + \sqrt{\frac{\lambda_i K_i r_0^2}{D}} \operatorname{erf} \sqrt{\lambda_i (t - t_0)} \right\} \{h(t - t_0) - h(t - t_0 - T_i)\} \quad [21]$$

- Inventory-limited model

From eq.[13]

$$m_i(t ; t_0, T_i) = 4 \pi r_0^2 L_c (t - t_0) \frac{M_i^0 e^{-\lambda_i t}}{M_c^0} \{h(t - t_0) - h(t - t_0 - T_i)\} \quad [22]$$

- Solid-diffusion model

From eq.[19]

$$m_i(t ; t_0, T_i) = 16 C_i^0 D e^{-\lambda_i t} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \left[\frac{4b}{(2n-1)^2 \pi} + \frac{\pi}{b \alpha_m^2} \right] e^{-D(t-t_0)(\alpha_m^2 + \beta_n^2)} \cdot \{h(t - t_0) - h(t - t_0 - T_i)\} \quad [23]$$

The cumulative fractional release of radionuclide *i* from a waste drum is

$$F_i(t) = \int_0^t f_i(t') dt' \quad [24]$$

RESULTS

Radionuclide Leach Rates

As an illustration of the REPS model, the calculated leach rates of Cs-137 and Sr-85 are compared with leaching test results by Moore (7). The inventory-limited leaching model in the REPS code is included to predict the leaching of soluble radionuclides. For this illustration, the leach rate of cement waste form L_c is assumed to be 0.001 g/cm² day (8).

The cumulative fractional release of Cs-137 as a function of time calculated from the REPS code is shown in Figure 4. For a comparison, a result of Cs-137 leaching experiment by Moore is also shown. The predicted cumulative release is in good agreement with the leaching test result.

Similarly, the time-dependent cumulative fractional release of Sr-85 obtained by Moore is in good agreement with the calculated cumulative fractional release by the congruent release model of the REPS code as shown in Figure 5.

Effect of Corrosion

Three different fractional release rates of Cs-137 for three waste container corrosion scenarios are shown in Figure 6. The first scenario is groundwater penetration by localized corrosion on carbon steel drum surface. A corrosion rate of 262 $\mu\text{m}/\text{yr}$ is chosen from the corrosion data base of the REPS code for this illustration. The calculated penetration time and leach time are 4.5 years after emplacement and 45.9 years after leaching starts, respectively. The second scenario is groundwater penetration by uniform corrosion. The assumed corrosion rate is 26.8 $\mu\text{m}/\text{yr}$. For this case the calculated penetration time is 44.8 years after emplacement. The last case is the gradual increase of corroded surface area on waste drum. $\alpha = -5.3$ and $\beta = 0.12$ in eq.[2] are assumed (4). The calculated leach time is 90.1 years.

The first scenario results in the most conservatively high fractional release rate of Cs-137 because of the assumption that the entire surface of drum is exposed to groundwater as soon as the first corrosion pit penetrates the drum. Because of the relatively lower corrosion rate of uniform corrosion, the release of Cs-137 for the second scenario is delayed and shows a lower maximum release rate. The last scenario may result in a probable and less conservative estimation of radionuclide release from waste drums in a geologic repository.

Radionuclide Release from Repository

The calculated annual release rates of 9 important radionuclides out of repository engineered barriers from 250,000 drums of nuclear power plant waste disposed of in underground repository are shown in Figure 7. Among various nuclides, long half-life nuclides such as Cs-137, Sr-90, and Tc-99 in fission products, Co-60, Ni-59, and Ni-63 in activation

products, and Pu-241, Am-241, and Cm-244 in actinides are selected as the representative radionuclides as shown in Table 1. The maximum initial radioactivity expected for the first low-level radioactive waste repository with the capacity of 250,000 drums is expected less than 134,000 Ci. About 50 % of the initial radioactive contents will result from Cs-137, Co-60, and Ni-63.

Migration in Backfill

In Figure 8, the annual release rate of Cs-137 from 250,000 waste drums into backfill and the expected release rate out of backfill with a thickness of 5 cm, 20 cm, or 100 cm are illustrated. Radionuclide migration through backfill materials is governed by advection, dispersion, retardation, and radioactive decay. Several mathematical models were developed for this migration concept. For this simple illustration, one-dimensional migration model in porous medium is used (5). The results show that backfill materials can contribute sufficiently to safety by retardation effect. The sorption distribution coefficient of $100 \text{ cm}^3/\text{g}$ and the diffusion coefficient of $0.031 \text{ m}^2/\text{yr}$ are used.

Parametric Uncertainty and Sensitivity Analysis

For the parametric uncertainty and sensitivity analysis in the risk assessment of low-level radioactive waste repository(9), Latin hypercube sampling and rank correlation techniques are applied for the parameter values shown in Table 2. Latin hypercube sampling technique is cost-effective for large computer programs because it generates smaller error to estimate output distribution even with smaller number of runs than Crude Monte Carlo technique. Rank correlation technique is good for generating dependence structure between samples of input parameters.

In Figure 9, the annual release rate of Cs-137 which is the most important radionuclide for short-term to medium-term radiological effects of repository, is calculated for 30 sample runs (9). Due to conservatively chosen parameter values, some of the 30 sample runs result in very high annual release rate of Cs-137. The maximum release rate out of repository into

geologic media among 30 sample runs is 170 Ci/yr, or 0.5% release of initial Cs-137 inventory per year.

From the results of uncertainty and sensitivity analysis, it is found that the degradation time of concrete structure and the corrosion rate of steel drum are strongly sensitive to overall uncertainty in few tens of years after repository closure and that in hundreds years after closure the leach rate of cement waste matrix is strongly sensitive to overall uncertainty in case of Cs-137.

Risk Assessment

Among various exposure scenarios evaluated for the risk assessment, intruder scenario results in the worst exposure scenario. In this risk assessment, an imaginary town adjacent to the repository is considered as shown in Table 3.

The assumed exposure pathways in the intruder scenario are:

- a) extensive use of contaminated groundwater from an imaginary aquifer underlying the repository by the whole population of an imaginary town
- b) food grown on site.

The annual dose to an individual is calculated for the following biosphere pathways (9, 10):

- a) drinking water
- b) milk
- c) beef
- d) vegetables.

Groundwater flow model is shown conceptually in Figure 10.

However, even for the worst-case scenario such as the intruder town just next to the repository site, huge contaminated aquifer, extensive development of groundwater resources, no credit to natural barrier (no sorption retardation, no dispersion, and instantaneous transfer of released radioactivity from engineered barriers to biosphere), and very conservatively chosen parameters of the biosphere model, the maximum value of the annual dose to an individual among 30 sample runs is 3.96 mrem/yr if aquifer size is assumed 100 times of

annually pumped volume of groundwater.

According to our previous analysis(11), the maximum dose rate from low-level radioactive waste repository would be less than 1 mrem/yr when natural barrier was taken into consideration. Therefore the interpretation of the results of this analysis requires some caution. In this study, we conservatively neglect the natural barrier to show that low-level radioactive waste repository would cause no serious harm to human even in unrealistic intruder scenario, and that the performance of far field needs to be proven for minimum requirements to ensure adequate repository performance.

CONCLUSIONS

A low-level radioactive waste repository source term code is developed and the preliminary calculation shows that the REPS code can estimate reasonably the leaching behaviors of cesium and strontium. The effect of exposed surface area ratio on the release of radionuclides from the waste drums can not be neglected. Therefore details on the container corrosion, especially localized corrosion, should be studied.

A simplified safety assessment is carried out on rock-cavern type disposal for LLW. The results of the preliminary assessment show that Cs-137, Ni-63, and Sr-90 are dominant. From the results of uncertainty and sensitivity analysis of Cs-137 release, it is found that the degradation time of concrete structure and the corrosion rate of steel drum are very sensitive to overall uncertainty at early stage after repository closure and that the leach rate of cement waste matrix is sensitive to overall uncertainty on the long term. The results of the potential public health impacts associated with the low-level radioactive waste repository show that radiological doses to persons who might be exposed do not exceed the expected regulatory limits. It is also demonstrated that near-field performance should be given more attention.

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Table Captions

Table 1. Radionuclide Inventory of LLW Repository (250,000 Drums)

Table 2. Parameter Values Used in the Analysis

Table 3. Imaginary Town Adjacent to LLW Repository

Figure Captions

Figure 1. LLW Arisings in Korea

Figure 2. Performance Assessment of LLW Disposal and the REPS Model

Figure 3. Flow Diagram of the REPS Code

Figure 4. Cumulative Fractional Release of Cs-137 from a Cement Waste Form

Figure 5. Cumulative Fractional Release of Sr-85 from a Cement Waste Form

Figure 6. Fractional Release Rates of Cs-137 Based on Three Waste Container Corrosion Scenarios

Figure 7. Radionuclide Release from Repository (250,000 Drums)

Figure 8. Release Rate of Cs-137 out of Backfill

Figure 9. Results of Parametric Uncertainty Analysis

Figure 10. Groundwater Flow Model

Table 1. Radionuclide inventory of LLW repository (250,000 drums)

Nuclide	Inventory [Ci]	Nuclide	Inventory [Ci]
H-3	158	Sb-125	258
C-14	169	I-129	0.63
Fe-55	23,065	Cs-134	11,808
Co-60	22,679	Cs-137	33,978
Ni-59	364	Eu-154	2.8
Ni-63	12,075	Pu-238	6.1
Sr-90	288	Pu-239	9.1
Nb-94	9.4	Pu-241	104
Tc-99	12	Am-241	11
Ru-106	1,811	Cm-244	2.6

Table 2. Parameter values used in the analysis

Parameter		Value	Distribution Type
Degradation time		0 - 300 years	lognormal
Corrosion rate		26.8 - 262 $\mu\text{m}/\text{yr}$	loguniform
Waste-form leach rate		10^{-3} - 10^{-4} $\text{g}/\text{cm}^2 \text{ day}$	uniform
Porosity		0.248 - 0.475	uniform
Drum thickness	Mean	1.2 mm	normal
	Standard deviation	0.12 mm	
Mass of waste form per drum	Mean	3.1×10^5 g	normal
	Standard deviation	6.2×10^4 g	
Diffusion coefficient in solid		10^{-10} cm^2/s	constant
Diffusion coefficient in liquid		315.36 cm^2/yr	constant

Table 3. Imaginary town adjacent to LLW repository

Population		9500 persons
Area		52 km^2
Wells	- depth	100 m
	- pumping rate	$200 \text{ m}^3/\text{yr}$
	- minimum distance btwn wells	500 m
	- number of wells	200
Water consumption rate by man		200 l / day
	- drinking	$0.549 \text{ m}^3/\text{yr}$
Water consumption rate by cattle		$29 \text{ m}^3/\text{yr}$

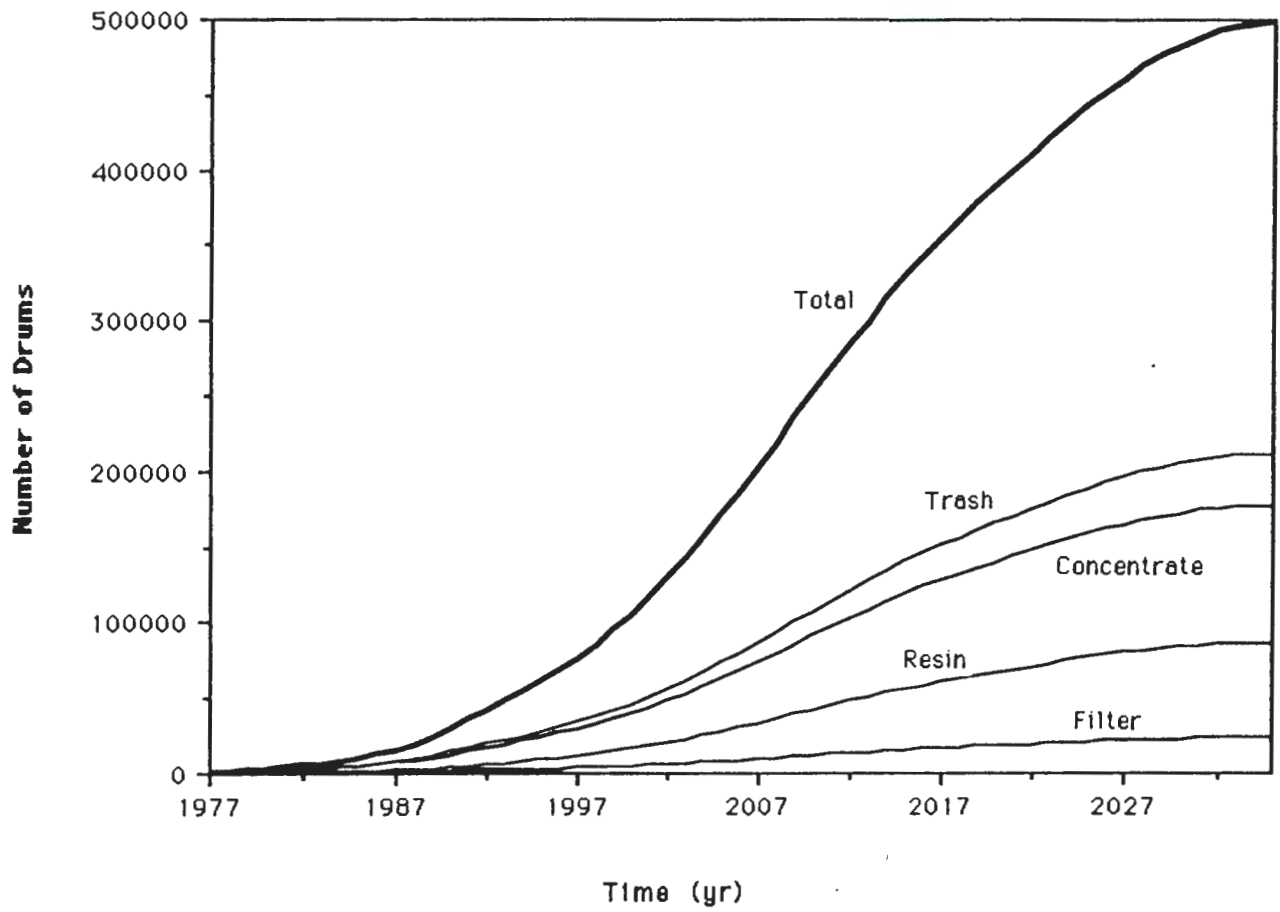


Figure 1

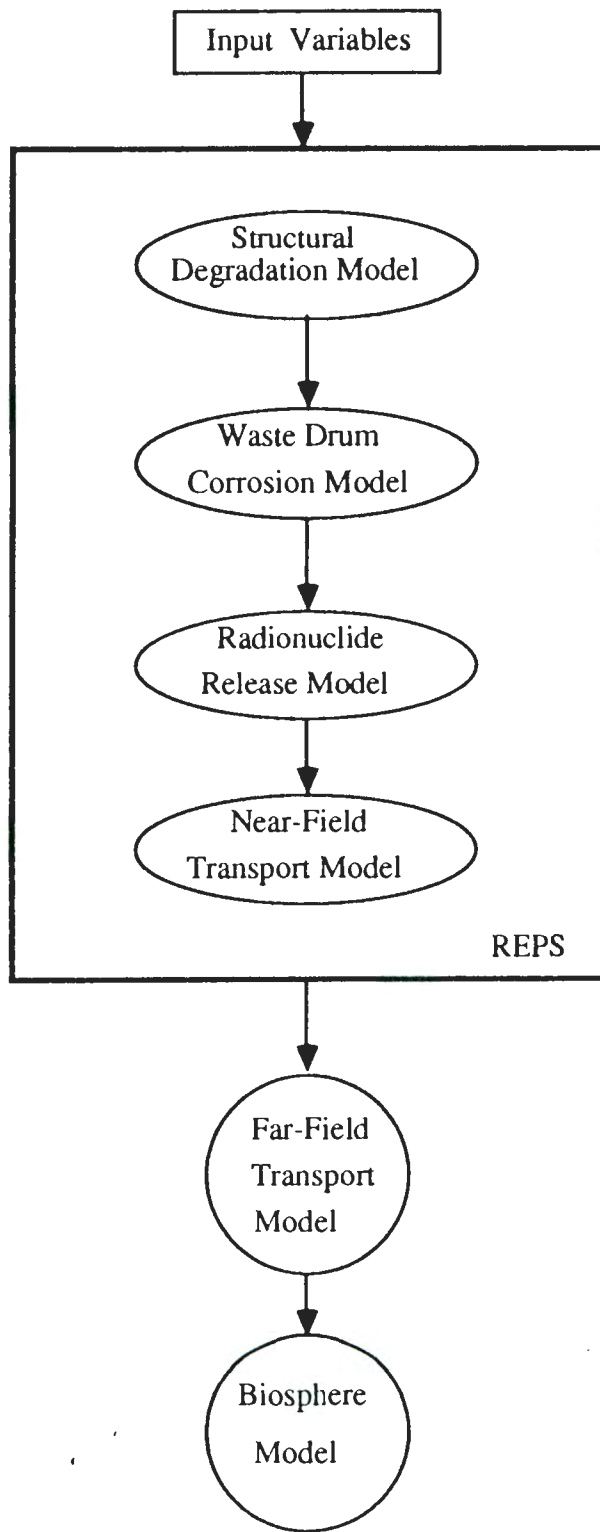


Figure 2

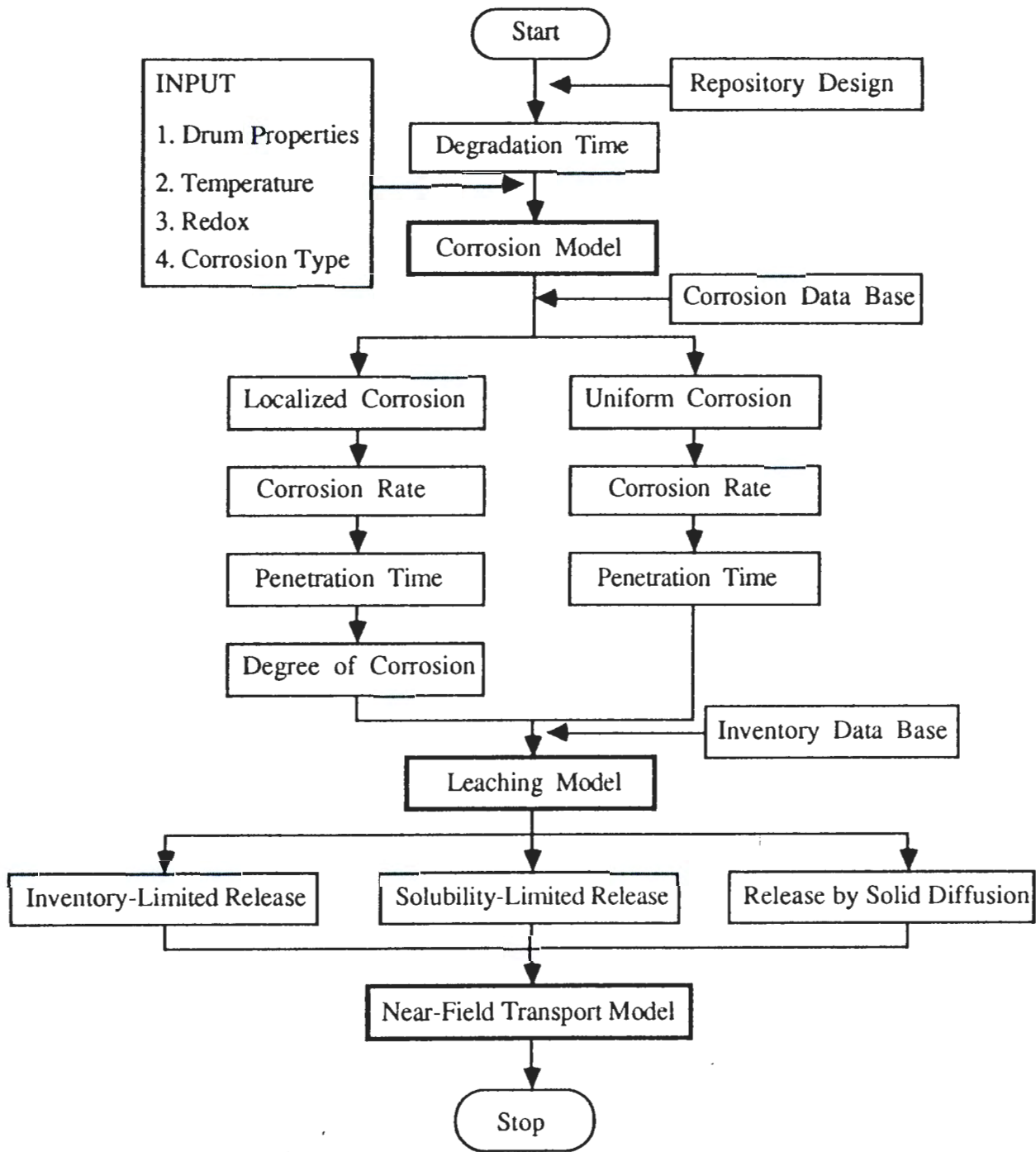


Figure 5.

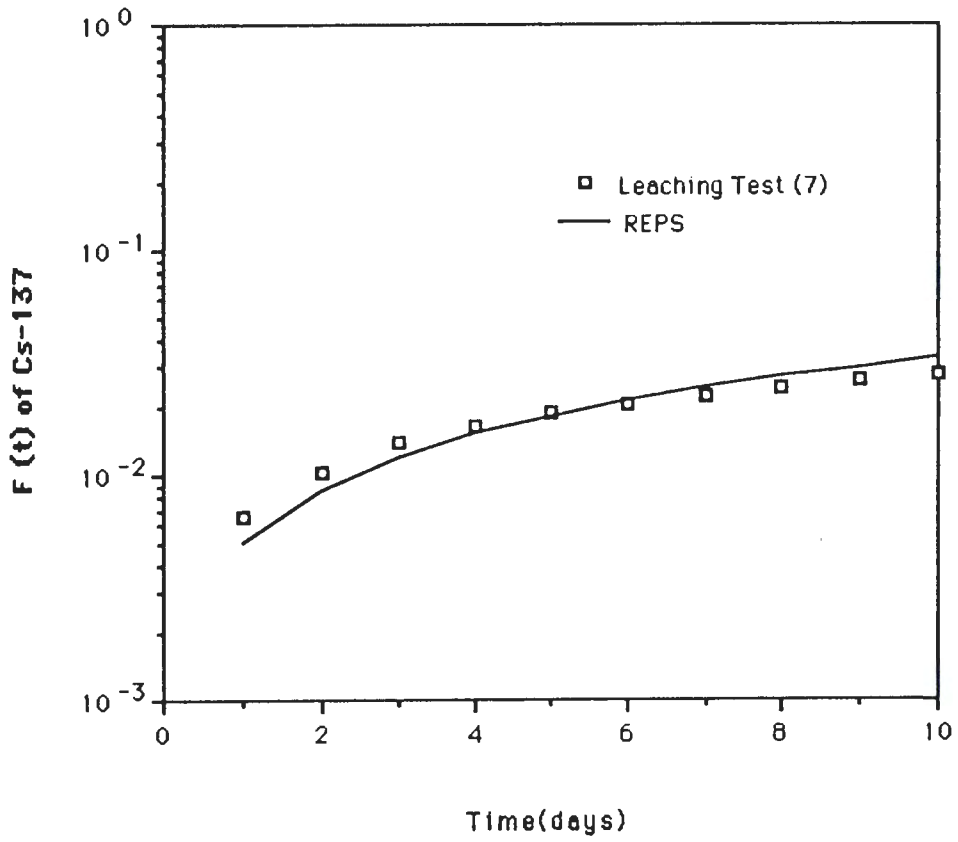


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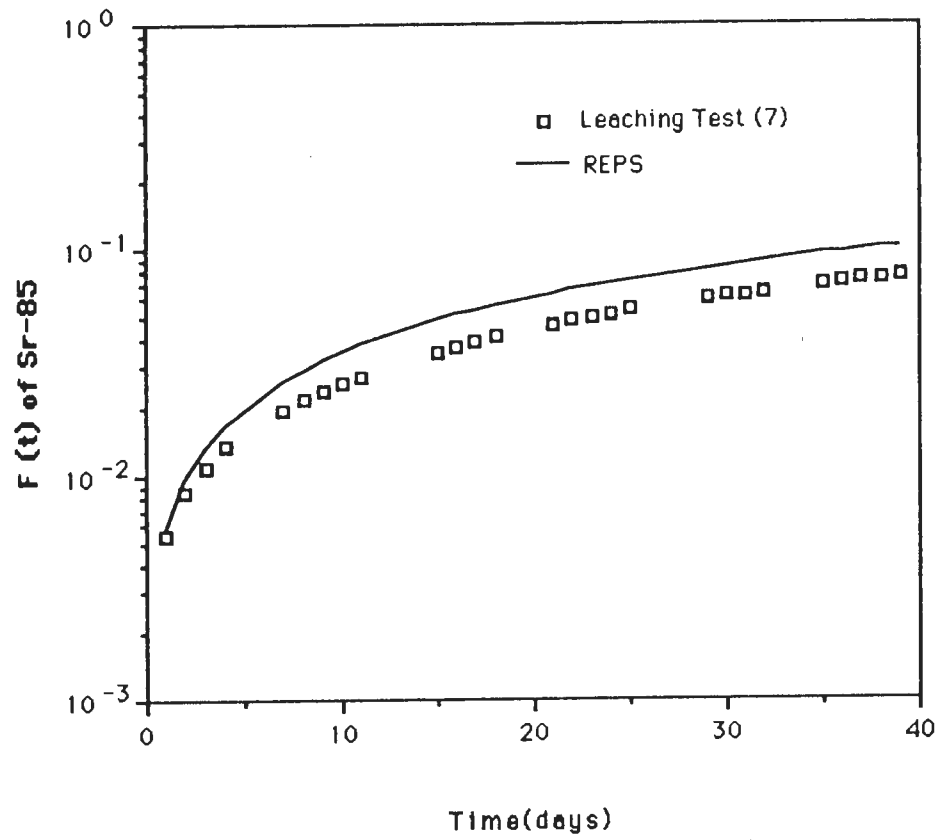


Figure 5

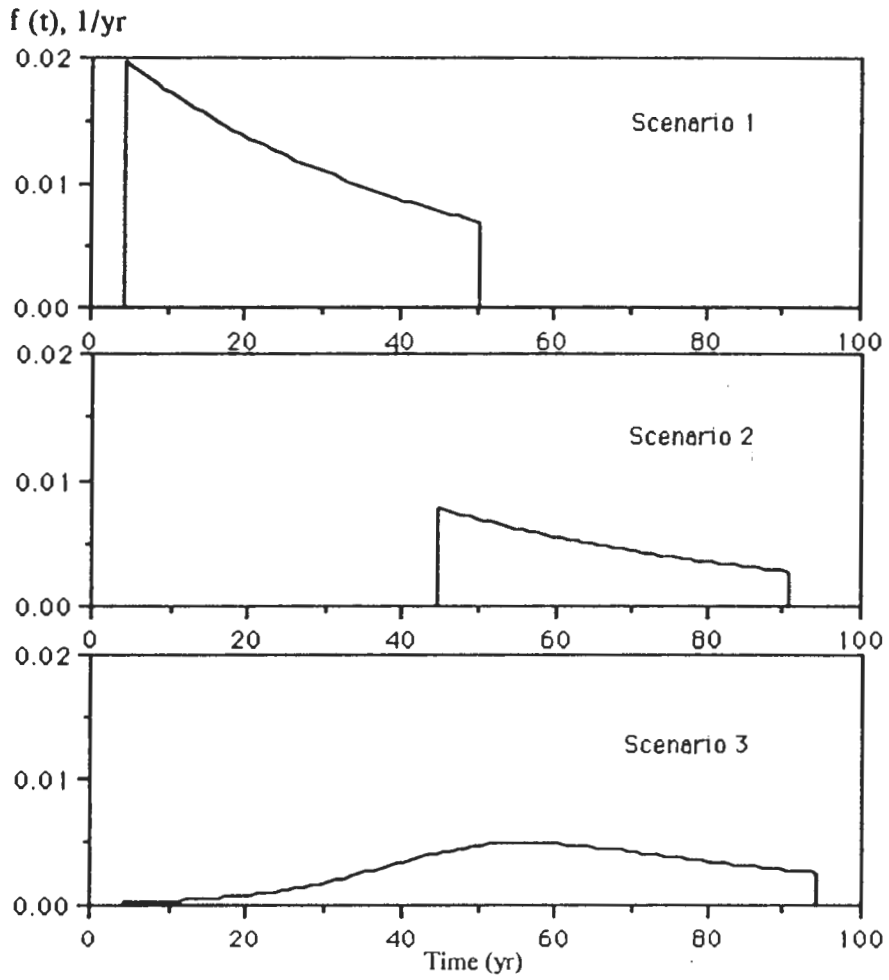


Figure 6.

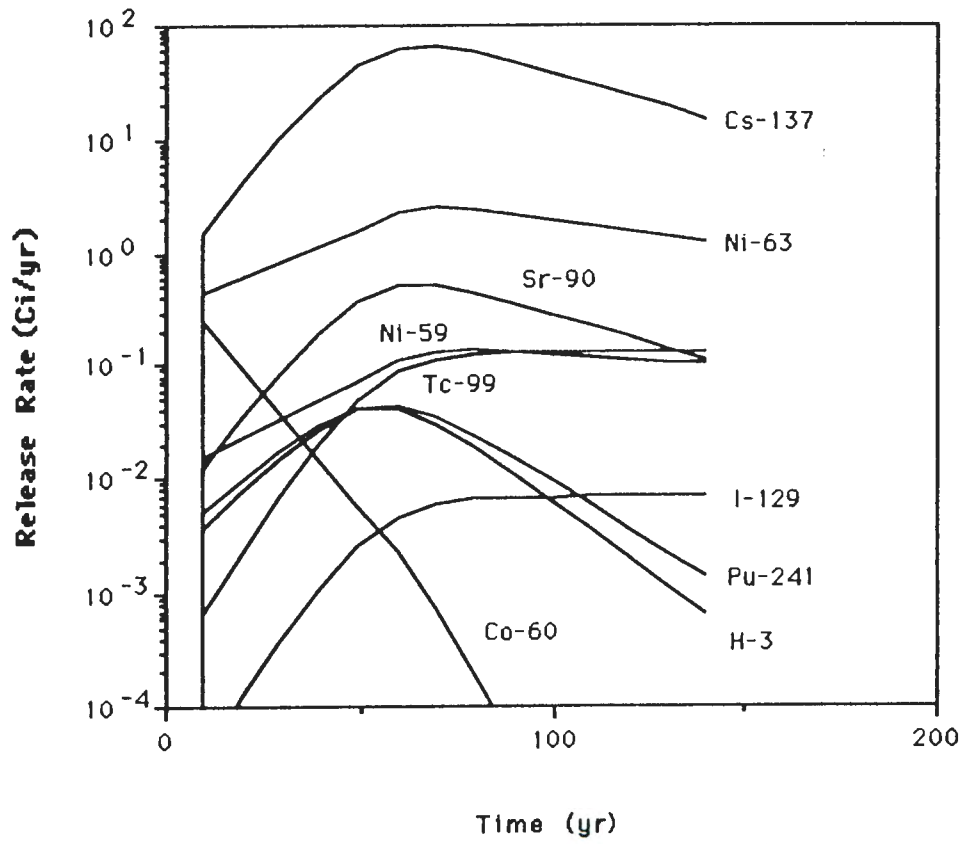


Figure 7.

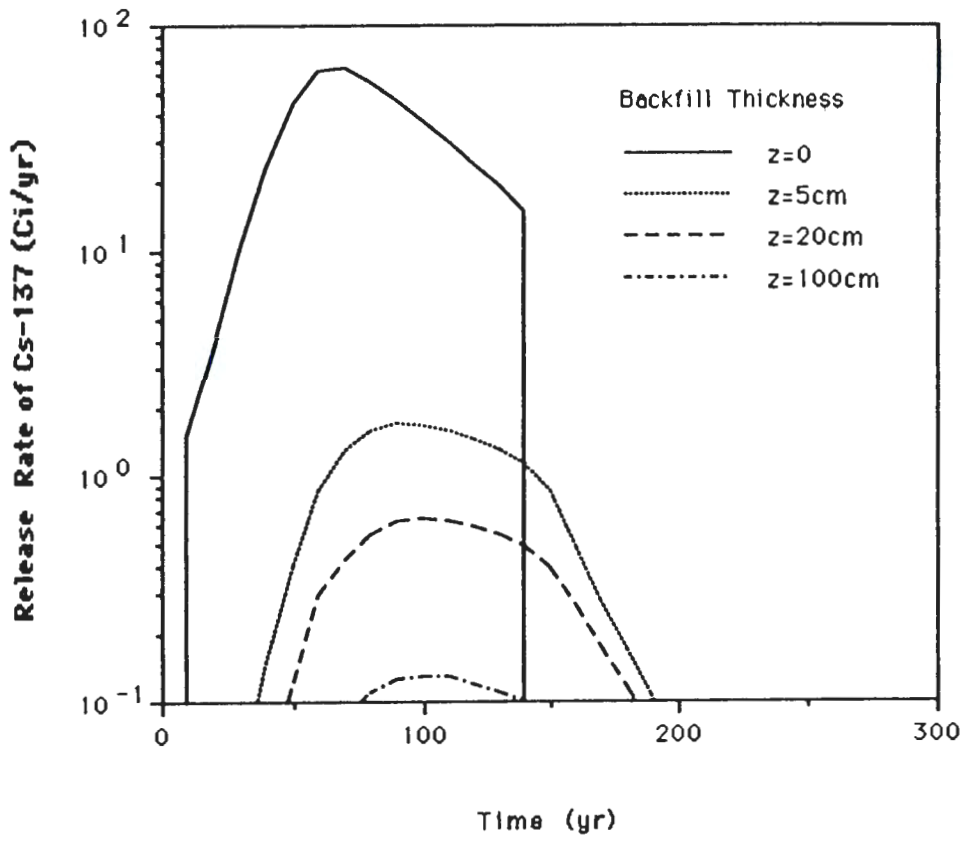


Figure 8.

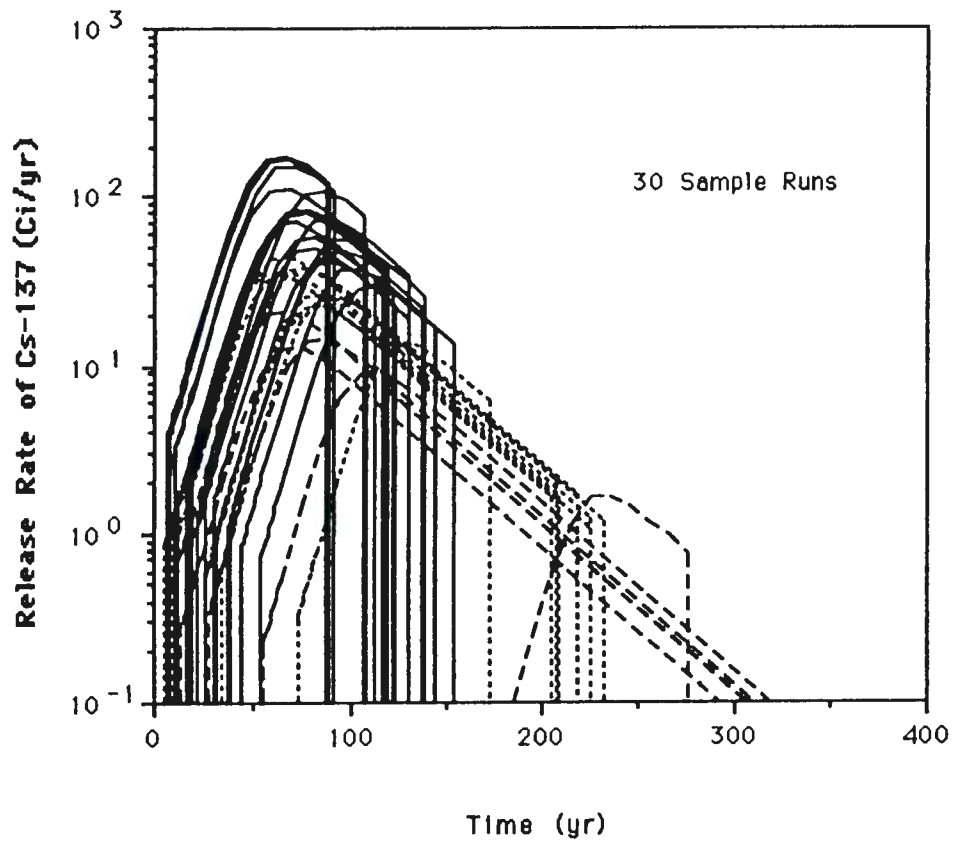


Figure 9

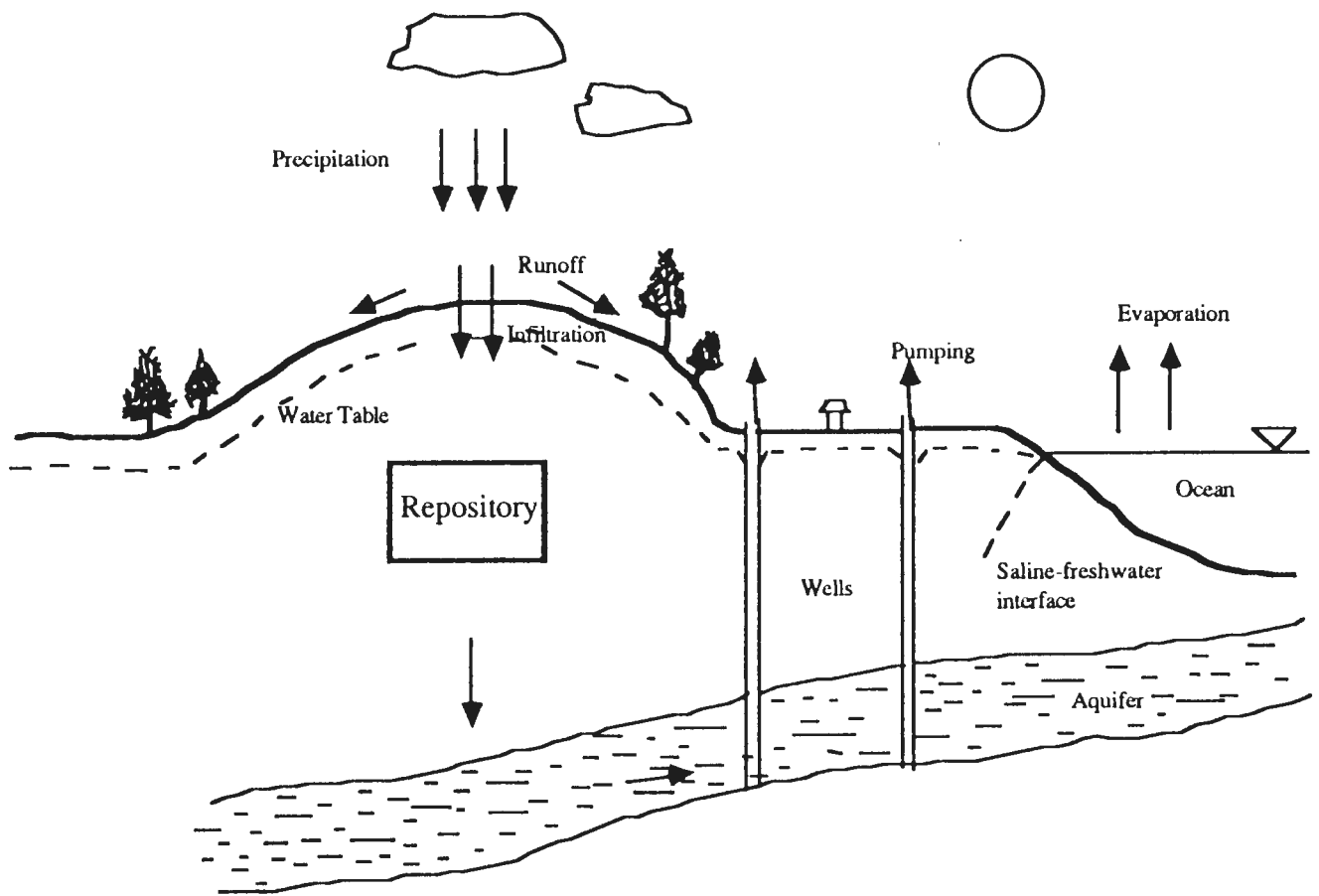


Figure 10.

CORRESPONDENCE DISTRIBUTION COVERSHEET

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SUBJECT: WASTE MANAGEMENT MANUSCRIPT WM 20217-004

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