A FIRST APPROXIMATION FOR MODELING THE LIQUID DIFFUSION PATHWAY AT THE GREATER CONFINEMENT DISPOSAL FACILITIES*

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ABSTRACT

The greater confinement disposal (GCD) project is an ongoing project examining the disposal of orphan wastes in Area 5 of the Nevada Test Site. One of the major tasks for the project is performance assessment. With regard to performance assessment, a preliminary conceptual model for ground-water flow and radionuclide transport to the accessible environment at the GCD facilities has been developed. One of the transport pathways that has been postulated is diffusion of radionuclides in the liquid phase upward to the land surface. This pathway is not usually considered in a performance assessment, but is included in the GCD conceptual model because of relatively low recharge estimates at the GCD site and the proximity of the waste to the land surface. These low recharge estimates indicate that convective flow downward to the water table may be negligible; thus, diffusion upward to the land surface may then become important. As part of a preliminary performance assessment which considered a base-case scenario and a climate-change scenario, a first approximation for modeling the liquid-diffusion pathway was formulated. The model includes an analytical solution that incorporates both diffusion and radioactive decay. For the base-case scenario, the liquid-diffusion pathway contributed significantly to the cumulative release of radionuclides relative to the convective pathway, but were not higher then releases established by the U.S. Environmental Protection Agency (EPA) in 40 CFR Part 191. In fact, the releases from the liquid-diffusion pathway were significantly below the EPA requirements. For the climate-change scenario, the liquid diffusion results did not contribute significantly to the final results. Overall, these results indicate that, despite the configuration of the GCD facilities that establishes the need for considering the liquid-diffusion pathway, the GCD disposal concept appears to be a technically feasible method for disposing of orphan wastes. Future analyses will consist of investigating the underlying assumptions of the liquid-diffusion model, refining the model if necessary, and reducing uncertainty in the input parameters.

INTRODUCTION

The Department of Energy (DOE) is responsible for disposing of a variety of radioactive wastes, which include high-level wastes, transuranic wastes, and low-level waste (2). The latter also includes greater-than-class-C waste. Some of these wastes do not comply with the waste acceptance criteria for proposed disposal sites such as the Waste Isolation Pilot Project and Yucca Mountain, or are prohibited from shallow land burial. These wastes have been termed "orphan" wastes and require an alternative disposal method (2).

Since 1984, the DOE has been disposing of some of these orphan wastes at the Radioactive Waste Management Site in Area 5 of the Nevada Test Site. The method of disposal utilized at this site is called Greater Confinement Disposal (GCD). The GCD method consists of boreholes 36.6 meters deep and either 3 meters or 3.7 meters in diameter. The bottom 15.2 meters of the boreholes are filled with waste and the upper 21.4 meters are backfilled with native material. The boreholes are within the unsaturated zone, at an estimated 200 meters above the water table. Currently, four boreholes contain approximately 1,000 curies of plutonium and five curies of uranium.

Sandia National Laboratories (SNL) staff conducted a preliminary performance assessment of the GCD site as part of an ongoing project to examine the technical feasibility of this site for orphan waste disposal. This examination was based on comparing the performance of the GCD site against the EPA requirements for the disposal of high-level waste and transuranics, 40 CFR Part 191**. The rationale of the preliminary performance assessment was to use first-approximation models which were either justified or conservative (i.e., overestimated release rates) to examine the technical feasibility of the GCD disposal concept, and to direct further site characterization efforts. These

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** 40 CFR Part 191 was vacated by the U.S. Court of Appeals of the First Circuit and remanded to the U.S. Environmental Protection Agency for repromulgation. It is expected that when this regulation is repromulgated the containment requirements (40 CFR Part 191.13) will not have changed significantly from those in the original regulation. The individual protection and ground water protection requirements will probably change, but it is not known what these changes will be. Therefore, for the preliminary performance assessment, the original individual protection requirements are used.

first-approximation models could later be refined for a final performance assessment, if necessary.

As part of the preliminary performance assessment, one of the transport pathways identified was diffusion of radionuclides, dissolved in the ground water, upward to the land surface. The need for considering this pathway resulted from the proximity of the waste to the land surface (21.4 meters) and the relatively low estimates (possibly, as low as 10^{-14} cm/s) of recharge for the area. For most disposal sites this pathway is not considered because of the depth of disposal, or because convective transport downward is the dominant transport path. The following sections present the first-approximation model used to simulate the liquid-diffusion pathway, how this model was implemented in the preliminary performance assessment, and the results of the liquid-diffusion pathway simulations.

MODEL DEVELOPMENT

The liquid-diffusion pathway consisted of the following processes: 1) dissolution of radionuclides into ground water; 2) upward diffusion of radionuclides directly to the land surface; 3) upward diffusion of radionuclides to plant roots, which in turn, transport radionuclides to the land surface; and 4) during diffusive transport, radioactive decay and production from multiple radioactive chains with multiple members. Based on these processes, the following assumptions were invoked for the model:

1. At time zero, all radionuclide species in the initial inventory are available for dissolution in the ground water. This assumption is conservative in that allowance is not made for any delay resulting from the presence of a waste package. Any allowance for a waste package would probably not affect the results significantly since a waste package lifetime would be relatively short compared to the regulatory time frame.
2. For the complete simulation time, the dissolution of each radionuclide is limited by its solubility, and its source is not depleted by radioactive decay or dissolution. Therefore, the concentration of radionuclides (i.e., plutonium and uranium isotopes) at the source is constant. This assumption is conservative because radionuclides are assumed to be available for transport for the complete simulation time. For the uranium isotopes, which have relatively long half-lives, relatively low solubility limits, and relatively large mass inventories, this assumption is reasonable. For the plutonium isotopes, which have relatively short half-lives, this assumption may not be applicable, but it is conservative.
3. The concentration of radionuclides at the ground surface is assumed to be zero. This assumption is conservative in that it provides a relatively large driving force for the flux of radionuclides to the ground surface.
4. During diffusion, the radionuclide species do not adsorb onto the porous medium. This is conservative because the sorption process would only serve to retard, or decrease radionuclide transport. This assumption is also computationally advantageous.
5. Liquid diffusion is one-dimensional, where diffusion occurs upward and downward. In reality, diffusion would occur radially from the source in three dimensions, but it is conservative to assume that the radionuclides diffuse in only one dimension.
6. Downward convective liquid-phase transport is not coupled with upward diffusive liquid transport. This is conservative since downward convective transport would only serve to decrease upward diffusive transport.
7. The molecular diffusion coefficient is not radionuclide-specific; therefore, all radionuclides have the same effective diffusion coefficient. This assumption is justified because all the radionuclides have approximately the same atomic mass, and the molecular diffusion coefficient is a function of atomic mass (3). This is a first-approximation since the exact chemical species (i.e., complexes, compounds) of the isotopes in the ground water is unknown at this time.
8. Radionuclides reach the ground surface either by diffusing 36.6 m upward or by diffusing upward to the root zone and then being transported to the surface via plant roots. The plant roots extend to a depth of 18.3 m and transport radionuclides to the ground surface (i.e., to their foliage) immediately. The ratio of the radionuclide concentration in the foliage to that of the ground water at depth of 18.3 m is 1×10^{-3} (4) and the plant roots occupy 10% of the available volume at that depth. These assumptions are either based on estimates for an arid region, or are conservative.
9. The cumulative release to the ground surface is the sum of diffusive transport directly to the land surface and diffusive transport to the plant roots which then transport radionuclides to the land surface. This is conservative since the mass uptake by plants is not subtracted from the mass transported directly to the land surface.
10. The effective transport porosity of the unsaturated zone is equivalent to the moisture content of the porous medium and is not a function of distance. This is the conventional assumption (5) that the unsaturated nature of the porous medium is accounted for by replacing the porosity by the moisture content.

The above assumptions were translated into mathematical form to construct the liquid- diffusion model. The liquid-diffusion model is broken into two parts, decay and diffusion. The complete solution is given by the product of these two parts,

$$C(x,t) = C_D(x,t)\, C_i(t), \qquad \text{(Eq. 1)}$$

where C(x,t) is the complete solution, $C_D(x,t)$ is the fundamental diffusion solution which is not radionuclide specific, and $C_i(t)$ is the radioactive decay solution which is radionuclide specific.

For the diffusive part of the model, the governing equation is given by Fick's second law,

$$\frac{\partial C_D}{\partial t} = D\frac{\partial^2 C_D}{\partial x^2}, \qquad \text{(Eq. 2)}$$

where D is the effective diffusion coefficient for diffusion in the porous medium, t is time, and x is distance from the top of the waste to the land surface. D is further defined by

$$D = \frac{\theta D_M}{\tau}, \qquad \text{(Eq. 3)}$$

where θ is the moisture content of the partially water saturated porous medium, D_M is the molecular diffusion coefficient of the solute, and τ is the tortuosity factor for the porous medium.

The initial and boundary conditions for the diffusion part of the liquid-diffusion model are:

$$C_D(x,0) = 0,$$
$$C_D(0,t) = C_0, \text{ and} \qquad \text{(Eq. 4)}$$
$$C_D(L,t) = 0,$$

where x = 0 denotes the top of the waste and x = L the land surface.

With these conditions, the analytical solution to Eq. (2) is

$$C_D(x,t) = \frac{C_o}{2}\left[\sum_{n=0}^{\infty} erfc\left(\frac{x+2nL}{2\sqrt{Dt}}\right) - \sum_{n=1}^{\infty} erfc\left(\frac{2nL-x}{2\sqrt{Dt}}\right)\right], \qquad \text{(Eq. 5)}$$

The radioactive decay part of the liquid diffusion model is based on the following differential equations that describe decay of an n-member radionuclide chain,

$$\frac{dC_1}{dt} = -\lambda_1 C_1,$$
$$\frac{dC_i}{dt} = \lambda_{i-1}C_{i-1} - \lambda_i C_i, \quad 1 > i \le n. \qquad \text{(Eq. 6)}$$

where C_i and λ_i are the concentration and decay rate of the i-th member, respectively. The initial conditions for Eq. (6) are

$$C_i = C_i(0), \; 1 \le i \le n, \qquad \text{(Eq. 7)}$$

which are consistent with the initial condition for the diffusive solution given above. The solution to Eq. (6) is given by Gelbard (6) as

$$C_i(t) = \sum_{j=1}^{i} \alpha_j b_i^{(j)} \exp(-\lambda_j t), \quad 1 \le i \le n, \qquad \text{(Eq. 8)}$$

where

$$b_i^{(j)} = \begin{cases} 0, & i < j, \\ 1, & i = j, \\ \prod_{k=j}^{i-1} \dfrac{\lambda_k}{\lambda_{k+1} - \lambda_j}, & i > j. \end{cases} \qquad \text{(Eq. 9)}$$

and

$$\alpha_i = \begin{cases} C_i(0), & i = 1 \\ C_i(0) - \sum_{j=1}^{i-1} \alpha_j b_i^{(j)}, & 1 < i \le n. \end{cases} \qquad \text{(Eq. 10)}$$

To compare with the requirements of interest in the preliminary performance assessment, the integrated flux of radionuclides past the accessible environment (i.e., the land surface, in this case) over 10,000 years is needed. This quantity is defined as

$$T_i(t) = \int_0^t -D\frac{\partial C_D(x,t)}{\partial x}\Big|_{x=L} C_i(t)\, dt. \qquad \text{(Eq. 11)}$$

The derivative in Eq. (11) is given by

$$\frac{\partial C_D(x,t)}{\partial x} = \frac{C_o}{2\sqrt{\pi Dt}}\left[\sum_{n=0}^{\infty} \exp\left(\frac{-(x+2nL)^2}{4Dt}\right) + \sum_{n=1}^{\infty} \exp\left(\frac{-(2nL-x)^2}{4Dt}\right)\right] \qquad \text{(Eq. 12)}$$

The following section describes how the liquid-diffusion model was incorporated and used in the preliminary performance assessment of the GCD site.

MODEL APPLICATION

The basis for the preliminary performance assessment was the High-Level Waste Performance Assessment Methodology (7) previously developed at SNL for the U.S. Nuclear Regulatory Commission. The methodology includes scenario selection, process and pathway identification, consequence analysis, uncertainty analysis, and sensitivity analysis. In the following discussion, each part of the methodology is discussed as it relates to the liquid-diffusion model.

Scenario Selection

In the preliminary performance assessment, the liquid diffusion pathway was considered for two different scenarios, a base-case scenario and a climate-change scenario. The consequence of each scenario was calculated separately and these consequences were not consolidated into one probabilistic result for comparison with the requirements. The base-case scenario was based on current climatic conditions at the GCD site, and the climate-change scenario was based on a twenty fold increase from the base case in the estimated recharge, with a corresponding decrease in water table depth.

Process and Pathway Identification/Consequence Analysis

Several different processes and pathways were identified for the preliminary performance assessment. Besides the liquid-diffusion pathway discussed previously, convective transport downward through the unsaturated zone and horizontally in the saturated zone was also considered. Therefore, consequence analysis for the preliminary performance assessment involved the use of several different models and associated computer codes. To simulate the liquid-diffusion pathway, the analytical solutions presented above were implemented in the form of a computer code. The integral presented in Eq. (11) was solved numerically in the computer code.

Within each scenario, the liquid-diffusion pathway was considered only when the diffusive flux upward was found to be greater than the convective flux downward. The diffusive flux is given by the expression $[-D(\partial,C/\partial x)]$ and the convective flux is equal to q/θ, where q is the Darcy velocity found from assuming steady-state flow and is θ the moisture content. Comparing the time-dependent diffusive flux with the steady-state convective flux could not be done without assuming a non-zero time for the diffusive flux. Therefore, as a first approximation and for comparison purposes only, the diffusive flux was found for steady-state conditions. The expression for steady-state diffusive flux was

$$-D\,\frac{dC_s}{dx} = \frac{D}{L}, \qquad \text{(Eq. 13)}$$

where C_s is the steady-state concentration. Thus, the liquid-phase diffusion model was implemented when

$$\frac{D}{L} \geq \frac{q}{\theta}\ . \qquad \text{(Eq. 14)}$$

As indicated previously, the technical feasibility of the GCD concept was compared against the requirements in 40 CFR Part 191. This regulation contains requirements concerning individual protection, ground-water protection, and containment of radionuclides within the accessible environment. Therefore, consequence analysis involved evaluating three performance measures. However, for the liquid-diffusion pathway, only the individual protection and containment requirements are applicable. The individual protection requirements limit the maximum annual dose to any member of the public for 1,000 years and the containment requirements limit the cumulative release, in terms of curies, of specific radionuclides to the accessible environment for 10,000 years. For the liquid-diffusion pathway, the accessible environment is defined as the ground surface. As discussed previously for the diffusion pathway, the ground surface can be reached either directly or via plant roots. Therefore, the cumulative release to ground surface is the sum of the release from each route. The cumulative releases of radionuclides to the ground surface are normalized by release limits obtained from 40 CFR Part 191. The release limits are based on the amount of disposed waste. The normalized release estimates are summed over all radionuclides to produce what is referred to as the "EPA sum." The EPA sum is the basis for comparison with the containment requirements.

Uncertainty Analysis

Knowledge of the values of the input parameters for the liquid diffusion model is uncertain; therefore, these parameters, with the exception of D_M, were treated as random variables, each with a specified probability density function (pdf). The Monte Carlo simulation technique was used to propagate this uncertainty through the models. For the liquid-diffusion model, the following input is required: 1) the solubility of the plutonium and uranium isotopes, 2) moisture content of the porous medium, 3) molecular diffusion coefficient for each species, and 4) tortuosity factor for the porous medium. Each required input is discussed in the following sections.

The solubilities of the plutonium and uranium isotopes were described by two pdf's: one for the plutonium isotopes and one for the uranium isotopes. These were based on solubility calculations using the composition of water taken

from a well located near the GCD site. The pdf of the solubility of the Pu isotopes was uniform and ranged from 2×10^{-5} to 6×10^{-5} g/g (Fig. 1). The pdf of the solubility of the U isotopes was loguniform and ranged from 1×10^{-11} to 1×10^{-6} g/g (Fig. 2). Estimates of the moisture content at the GCD site, based on available information, produced a lognormal distribution shown in Fig. 3. The molecular diffusion coefficient for each species was taken to be a constant, 1×10^{-9} m^2/s. This was based on molecular diffusion estimates given by Weast (8) for species of molecular weight less than 500.

The pdf for the tortuosity was assumed to be uniform, with a range from 1 to 110 (Fig. 4). For unsaturated media, it has been suggested by several investigators (9,10) that a relationship between tortuosity and moisture content exists. Intuitively, this correlation is inverse; when the moisture content decreases, the tortuosity increases. Campbell (10) presents an empirical inverse relationship between moisture content and tortuosity, but the degree to which these parameters are correlated is a matter of judgement. For the preliminary performance assessment, the moisture content and the tortuosity were assumed to be strongly correlated, but not directly related. More specifically, the correlation coefficient between tortuosity and moisture content was assumed to be -0.9. The upper limit of the uniform distribution (i.e., 110) was determined based on the relationship given by Campbell (10), using the smallest estimate of moisture content. The lower limit (i.e., 1) for tortuosity is the

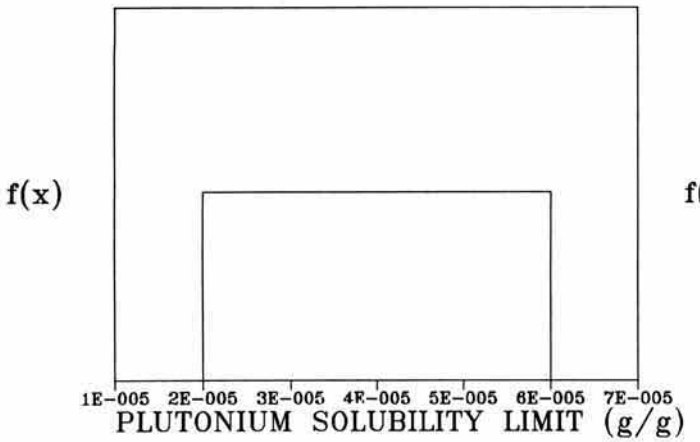

Fig. 1. Probability distribution function of plutonium solubility limit.

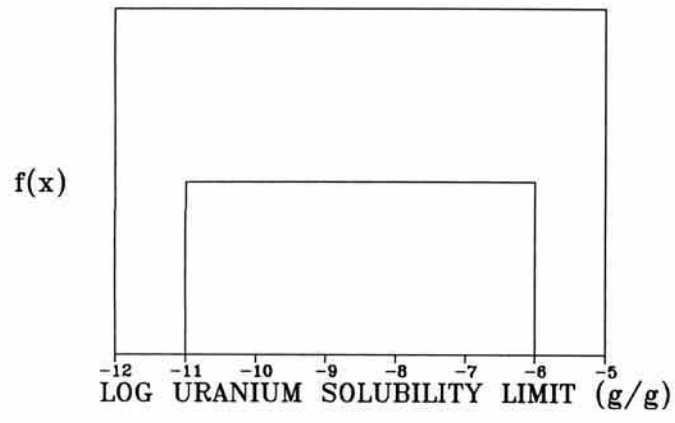

Fig. 2. Probability distribution function of uranium solubility limit.

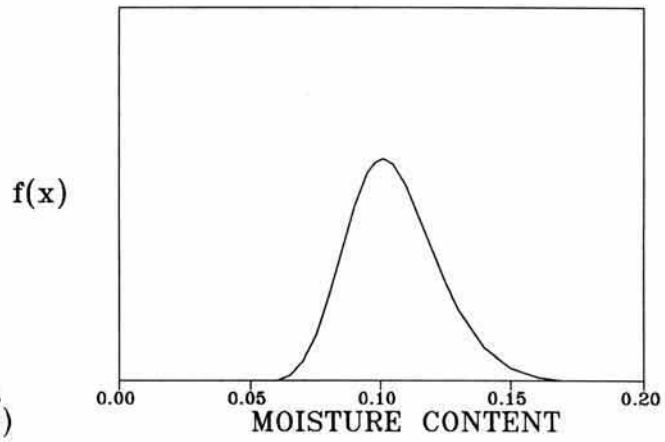

Fig. 3. Probability distribution function of moisture content.

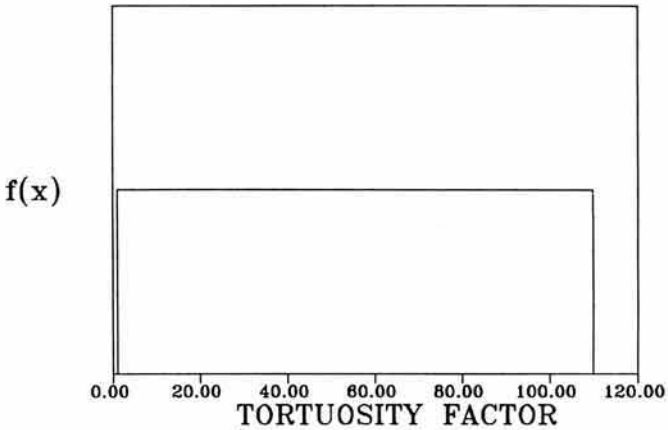

Fig. 4. Probability distribution function of tortuosity factor.

lowest possible value for a porous medium, and is therefore, extremely conservative.

The pdf's for all the input parameters were sampled using the Latin Hypercube Sampling (LHS) technique (11). This sampling method results in a set of input vectors, each vector consisting of a specific numerical value for each of the uncertain parameters. Each vector is then used as the input to the consequence analysis models and a value of the performance measure (e.g., EPA sum) is obtained for each of the sample vectors. For the base-case and the climate-change scenarios, 4000 vectors were used.

Sensitivity Analysis

The results of the consequence analysis are used to perform sensitivity analysis. Sensitivity analysis is used to identify the most important input parameters associated with the model output. The results of a sensitivity analysis can be used to set priorities for site-characterization activities. For the liquid-diffusion model the number of input parameters is relatively small and the model is relatively simple; thus, sensitivity analysis is simplified somewhat.

RESULTS

Of the 4,000 samples generated for the base-case scenario, 47 resulted in implementation of the liquid-diffusion model. That is, for those particular sample values, the flux of ground water toward the water table was less than the diffusive flux of radionuclides toward the ground surface. Of these 47 samples, 13 resulted in non-zero EPA sums. Of the total 4,000 samples, approximately 3,910 result in EPA sums of zero. Thus, of all the non-zero EPA sums, approximately 15% are the consequence of diffusion in the liquid phase. The 13 non-zero EPA sums from the liquid-diffusion pathway contained the third, fourth, fifth, sixth and seventh highest overall EPA sums. However, these EPA sums were not higher than those based on the EPA's containment requirements. Releases resulting from the convection pathway were also not higher than those established from the containment requirements. In fact, for the base-case scenario, the resulting EPA sums appeared to be significantly below the EPA containment requirements.

For the EPA's individual protection requirements, the strategy used to compare with the probabilistic containment requirements was employed. For the base-case scenario, the analyses indicated that, over 1,000 years, in less than 1% of the simulations, minute quantities of radionuclides reach the ground surface via diffusion in the liquid phase combined with uptake via plant roots. These quantities range from 4×10^{-10} pc/l to 6×10^{-3} pc/l and result in an annual effective dose equivalent of about 5.5×10^{-4} millirems. The individual protection requirements limit the annual does equivalent to 75 millirems to any critical organ and 25 millirems to the whole body, well above the annual effective dose equivalent found from the liquid-diffusion pathway.

For the climate-change scenario the containment requirements are the only requirements with which a comparison is made because individual protection requirements apply only to the base-case scenario. The liquid-diffusion pathway dominated the convective pathway in only 13 of the 4,000 samples, about 25% as often as in the base-case scenario. This was expected, since the increased downward flux of water tends to make liquid-phase diffusion a less significant pathway for radionuclide migration. Of these 13 samples, none appeared to result in significant releases to the accessible environment.

For sensitivity analysis, the EPA sums from the liquid-diffusion pathway as a function of tortuosity factor, for the base-case scenario are shown in Fig. 5. For tortuosity factors greater than ten, all corresponding EPA sums were zero. As expected, the higher EPA sums correspond to lower tortuosity factors. In fact, the only significant EPA sums correspond to tortuosity factors less than two, which are extremely conservative values for a porous medium. These same EPA sums as a function of moisture content are shown in Fig. 6. For moisture contents less than 0.12, all corresponding EPA sums were zero. Again, as expected, the higher EPA sums correspond to the higher moisture contents. Also as expected, the combination of low tortuosity and high moisture content produce the larger EPA sums.

^{239}Pu and ^{240}Pu were identified as the primary contributors to the cumulative release (i.e., EPA sum) for the liquid-diffusion results. This is expected because the solubility of the Pu isotopes is much greater than that of the U isotopes. Thus, the concentration of Pu (in g/g) is greater

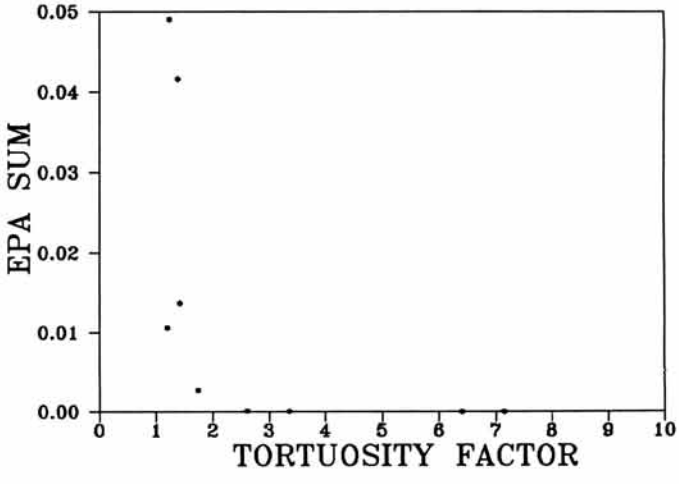

Fig. 5. EPA sum as a function of tortuosity factor for the base-case scenario.

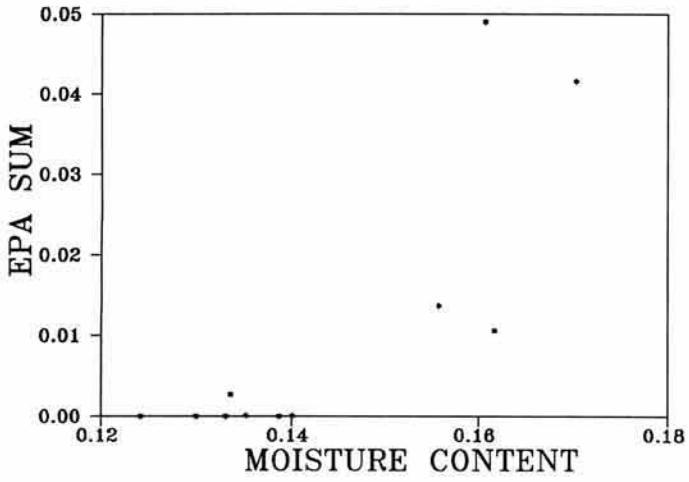

Fig. 6. EPA sum as a function of moisture content for the base-case scenario.

than the concentration of U. In addition, the activity (curies/gram) of the Pu isotopes tends to be much higher than that of the U isotopes. Therefore, because the releases are in terms of curies, the release of, for example, one gram of a Pu isotope results in a higher EPA sum than does the release of one gram of an U isotope.

SUMMARY AND CONCLUSIONS

A conservative, first-approximation model for a liquid-diffusion pathway has been applied as part of a preliminary performance assessment of the GCD facilities. This analytical model considers diffusion and radioactive decay. For the base-case scenario (i.e., current climatic conditions) the results indicated that the liquid-diffusion pathway contributed significantly to the resultant EPA sums, but did not produce EPA sums that were larger than those established by the EPA's containment requirements. Also, the diffusive transport of radionuclides did not result in doses higher than those established in the EPA's individual protection requirements. For the climate-change scenario, the liquid-diffusion pathway was not significant. These results indicate that although the configuration of the GCD facilities established the need for considering the liquid-diffusion pathway, for this pathway, the GCD disposal concept appears to be a technically feasible method for disposing of orphan wastes. Future analyses will consist of investigating the underlying assumptions of the liquid-diffusion model, refining the model if necessary, and reducing uncertainty in the input parameters.

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