MODELING OF GAS-PHASE RADIONUCLIDES RELEASE FROM LOW-LEVEL WASTE DISPOSAL FACILITIES

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ABSTRACT

This is an analysis of the release of gas-phase radionuclides from engineered barriers in a LLW disposal facilities to the atmosphere. Modeling of the time-dependent flow of gases subject to diffusion, advection, chemical reactions, and radioactive decay for the gases of concern has been undertaken.

This model was applied to analyze the release of ^{222}Rn from a earth-mounded concrete bunker. The results showed that any facility with a drain pipe and connected piping for monitoring provides a direct pathway for the fast release of significant amounts of ^{222}Rn to the atmosphere under atmospheric pressure pumping. For the migration of ^{222}Rn through concrete barrier and earthen cover system, there appears to be virtually no escape of this radionuclide as long as the clay layer is intact with enough moisture. This would hold whether the concrete barrier is intact or not.

INTRODUCTION

One of the most important approaches for evaluating the acceptability of a specific facility in the disposal of low-level radioactive waste (LLW) is to conduct performance assessments of the proposed facility prior to site confirmation and facility construction and operation. In conducting such performance assessments, all potential post-closure pathways of radionuclide release, migration, and exposure must be identified and described to assess the long-term potential impact of disposal facility on the surrounding environment.

Currently performance assessments of low-level radioactive waste (LLW) disposal facilities concentrate on the water pathway. It is standard practice to consider that for undisturbed performance of LLW disposal facilities, groundwater contamination is the only important generic pathway(1).

Radiochemical studies(2,3) at the LLW burial site in West Valley, New York, have indicated the potential significance of the gaseous releases of radionuclides. Analyses of that facility showed that venting of radioactive gases through the trench cover with dispersion into the atmosphere was the most significant transport pathway. The gases released were ^{85}Kr, ^{222}Rn, ^{3}HH, $^{14}CO_2$, and ^{14}C and ^{3}H as CH_4 and other hydrocarbons.

Contaminated gases may be introduced into the containment structure as a result of the failure or corrosion of disposal canisters or the decomposition of organic wastes. Some amount of gaseous and/or organic waste is expected to exist in all disposal facilities, therefore gas-phase release of radionuclides needs to be characterized.

Due to the public's unfavorable view on the shallow land burial of LLW, a major share of the LLW disposal facilities being planned by the States and state compacts are to be above- or near grade bunkered facilities. According to the current design of these facilities, often there is a flow drain in the engineered concrete structure and a standpipe for monitoring which is connected to the flow drain. This feature provides a direct gas transport pathway to the atmosphere from within the facilities (Fig. 1(4)).

None of the existing performance assessment models considers airborne releases of elements and volatile compounds from within the disposal facility. For all of the codes the analysis of the air pathway in performance assessments, i.e., concentration of radionuclides released from the source, must be calculated externally. Current performance assessment computer codes without the capability to analyze the gaseous release pathway may not provide meaningful projections of the radiological impacts of newly proposed facilities.

Another important aspect of analyzing gaseous radionuclides releases is to assess their impacts on the source term. If radionuclides in the waste are released to the environment through gases, the source inventory that will be available for the transport through groundwater is reduced. Since groundwater contamination is conjectured to be the most significant pathway for radiation exposure for an undisturbed facility(1), gaseous releases which generally are followed by rapid dilution in the air could be beneficial in reducing the dose to individual members of the public. Potential reductions in the source term through gaseous releases have not previously been analyzed in LLW performance assessments.

The purpose of these studies is to investigate the mechanisms of gaseous release of radionuclides from within a engineered disposal facility and to develop a computer model for assessing such releases.

ANALYSIS OF GASEOUS RELEASES OF RADIONUCLIDES

Radioactive gases are generated within LLW disposal facilities due to microbial activity, corrosion of steel in the waste containers and of metallic components, radiolysis, and radioactive decay(5). The characteristics of gas generation depend on the nature of the waste, the radionuclides it contains, the type of waste package and differences in the local chemistry of the environment within the disposal facility.

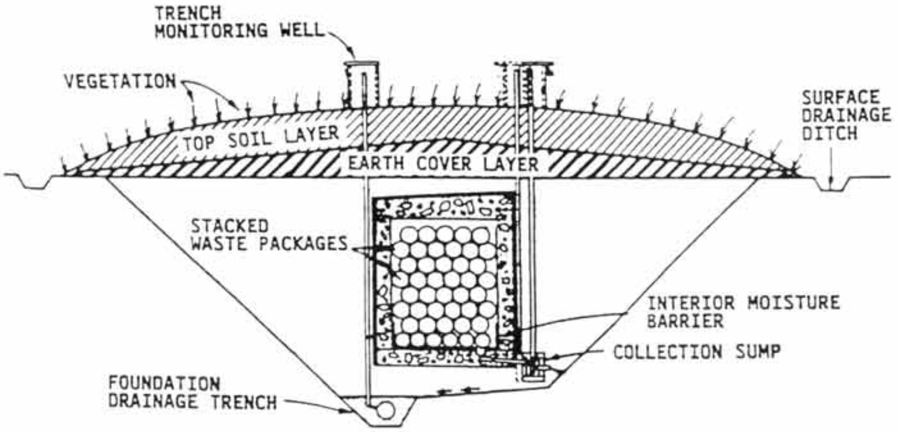

Fig. 1. Earth-mounded concrete bunker for disposal of low-level radioactive wastes (4).

Once these gases are generated, they can leak out from waste containers and migrate through the disposal facility. Gases can be released from an undisturbed disposal facility through two main migration modes: 1. diffusion as impurities in air and water vapor, and 2. advection of air and water vapor from the disposal facility due to atmospheric pressure variations. If a disposal facility is exposed to water infiltration or partially saturated with water, the interactions between air and water become important. These phenomena which tend to retard migration of radionuclides in the gaseous phase can take place away from the source location through the interaction of gaseous molecules with the ground water, and soil particles present at the site.

The mathematical model in this study includes the time-dependent effect of diffusion, advection, chemical reactions, and radioactive decay for each gas of concern(6). The resulting space- and time-dependent partial differential equation needs to be solved through the model geometry with appropriate boundary conditions.

Mathematically, the model is expressed as:

$$\frac{\partial C_i}{\partial t} = \frac{\partial}{\partial x}\left[D_i(x)\frac{\partial C_i}{\partial x} - v(x,t)C_i\right] + \sum_{j=1}^{n} k_j \mu_{ij} \prod_{m=1}^{n_{eq}} C^{|\mu_{im}|} - \lambda_i C_i + \lambda_{i-1} C_{i-1}$$

where

C_i	concentration of species **i** as a function of time and space
x	spatial variable,
i	number of the individual species (of quantity **neq**),
j	reaction number,
n	number of reactions,
D_i	diffusion coefficient of species **i** at position x
v	velocity of the gas or fluid,
n	total number of reactions,
k_j	reaction rate constant of reaction **j**,
μ_{ij}	stoichiometric coefficient for species**i** in reaction **j**.
λ_i	decay constant for species **i**

The complexity of both the time and spatial dependencies and the inhomogeneities of parameters within the model geometry preclude explicit analytic solution of the model equation.

The method of lines has been applied in this study for the numerical solution of the model equation. This method allows one to convert time-dependent partial differential equations into systems of time dependent ordinary differential equations (ODE's) by a spatial discretization procedure. The resulting system of ODEs is solved by the LSODE package. LSODE is a revised version of Gear's DIFSUB program with improvements in flexibility, portability and ease of use(7).

In describing the flow of gas by advection, the effects of atmospheric pressure variation on flow velocity (so-called atmospheric pressure pumping) was determined a priori and utilized in the model. The flow velocity in the open drainage system can be calculated based on the ideal gas law considering the total volume of air affected under barometric pressure variation. In a porous medium, if pressure gradients are established due to atmospheric pressure variations, then there will be an associated flow of gas. This can be described by Darcy's law. The time-dependent pressure field in the porous medium due to pressure variation at the outer surface also can be determined based on the conservation of mass in a bulk volume of a porous medium(8).

In an atmospheric pressure pumping situation, the direction of gas flow can be reversed. Therefore, both forward and backward advection are possible. A switch is provided to implement this situation in the finite differencing scheme to maintain numerical stability.

ANALYSIS OF ^{222}Rn RELEASE

The previously described model was used to analyze the time-dependent transport characteristics of ^{222}Rn through one type of a LLW disposal facility (e.g., an Earth-Mounded Concrete Bunker) in a one-dimensional description. In an earth-mounded concrete bunker, gases can either migrate through a concrete barrier or earthen cover system or be transported through the drainage system. The current design of the facility with a drain pipe and connected standpipe for monitoring provides a direct release pathway to atmosphere.

The source inventory of ^{222}Rn within an earth-mounded concrete bunker was modeled based on a description of one waste container (55 gallon drum) assuming that all the ^{226}Ra activity is contained in the drum in cement form. The value for the ^{226}Ra concentration in the drum was estimated[*] on the basis of the waste disposed during 1987 through 1989 at the Beatty, NV, shallow land burial disposal site(9).

Due to its short half-life (3.82 days), only a portion of the ^{222}Rn generated from the decay of ^{226}Ra will migrate to the open space within a drum and subsequently escape to the outside of the drum through a vent. The effective ^{222}Rn source inventory within a cement matrix was assumed to be the activity contained within a distance of one diffusion length[**] from the surface of the waste-form (Fig. 2). The concentration was calculated by dividing the activity within an effective source volume by the open space volume (11% of the total volume) within a drum. The open space in a drum is that which was generated when the poured cement solidified. For ^{222}Rn in an open space to escape from the drum, it was further assumed that all the ^{222}Rn had to diffuse the distance of one half a drum height.

Migration of gas within a vault (including the release of ^{222}Rn from a waste container) was described only by diffusion. The model geometry for the ^{222}Rn source within a vault includes a half-sized waste container, vent penetration, open space within a vault between waste containers. The average open space area per waste container was used as the flow area for the open space diffusion. The contributions of ^{222}Rn releases from surrounding waste containers were added as an external source term. The external source generation rate was calculated as the ^{222}Rn release rate from a waste container averaged over the dimension of the container height.

Next an estimate was made of how much of the ^{222}Rn that is released to the open air inside the vault would be available for release to the general atmosphere. Again, the diffusion length of ^{222}Rn in air was used to define the effective inventory of ^{222}Rn and this inventory was used for analyzing the release through the drain system or the concrete barrier and earthen cover system.

Figure 3 shows the schematic of the model geometry of the analysis from a waste drum to the atmosphere through the subsurface flow drain system. For the simulation of flow through the drain system, the model accounts for the pea gravel layer at the bottom of the vault structure, the primary drainage system and the cell drain sump monitor standpipe. The releases through the flow drain system and subsequent standpipe were described both by diffusion and advection. It was assumed that the drain and standpipe were dry. The ratio of flow area changes was accounted for in the model to overcome the limitations of the one-dimensional description of an open system flow including the variations in flow velocity within the drain system. The schematic of the model geometry through the earthen cover system is given in Fig. 4.

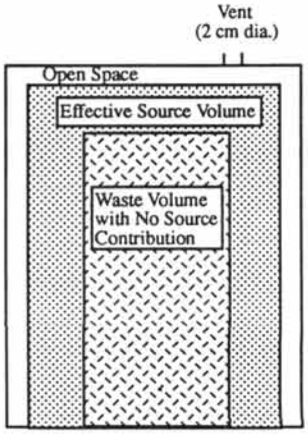

Fig. 2. Effective Rn-222 source volume within a waste container.

The release of ^{222}Rn to the atmosphere has been estimated using the condition of a pressure variation which can be expected during a severe storm. The rate of pressure variation is assumed to be cyclic (represented by a sine function) with maximum changing rate of 3 mb/hr over a period of three days. The changes in ^{222}Rn concentration within the drainage system and across the concrete barrier and earthen cover systems and in the flux at the surface of the soil cover have been simulated in the model.

Figure 5 presents estimates of variations in ^{222}Rn concentrations within the flow drain system. As may be noted, pressure pumping is expected to cause dramatic changes in the concentrations of this radionuclide. When the pumping effect is very high, the ^{222}Rn concentrations within the drain system (or standpipe) approach the same order of the concentration that is present inside the engineered barrier (10^5 - 10^6 pCi/cm^3). The occupational limit of ^{222}Rn concentration is 4 WLM (Working Level Month), which is close to 0.03 pCi/cm^3. Even though this limit has no practical applicability for overall performance assessment, it is obvious that the ^{222}Rn concentration within the drain pipe can be very high. Figure 6 shows estimates of changes in the radon flux at the surface of the soil cover (outside the standpipe) which indicate significant potential releases. Estimates of ^{222}Rn concentrations across the

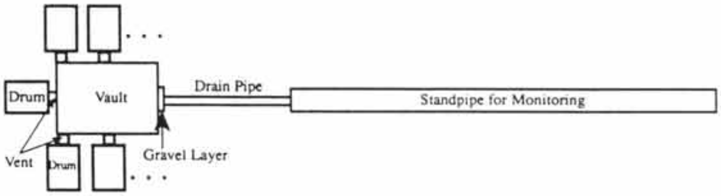

Fig. 3. Schematic of the radon transport model geometry for the release through primary drainage.

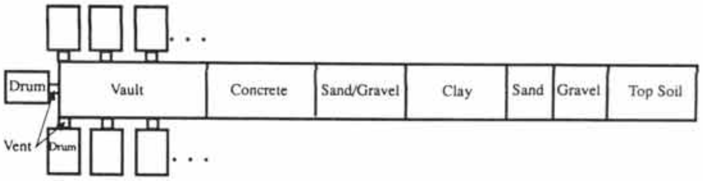

Fig. 4. Schematic of the radon transport model geometry for the release through concrete and earthen cover.

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[*] Total activity of ^{226}Ra disposed of in three years was divided by the total volume of waste disposed of in three years. The resulting activity was 2.663 pCi/cm^3. This site was chosen because it contained higher concentrations of ^{226}Ra than the LLW disposal facilities at Barnwell, SC, and Richland, WA.

[**] The diffusion length is difined as $L=(D/\varepsilon\lambda)^{1/2}$ where, D is the diffusion coefficient; ε is the porosity; and λ is the decay constant. The wasteform was assumed to be concrete.

earthen cover system are given in Fig. 7. In this calculation, the concrete barrier was assumed to be either intact or completely failed. The results indicate that the migration of gaseous radionuclides through the concrete barrier and earthen cover materials virtually ceases once the radionuclides enter the clay layer. The small variations in ^{222}Rn concentration through the earthen materials were caused by the differences in ^{226}Ra concentration as natural background in each medium.

Figure 8 represents the results of ^{222}Rn flux at the surface of the soil cover for the condition of less severe pressure variation. In this case, the maximum rate of pressure variation was 0.7 mb/hr over a period of seven days. Results indicate that the advective transport is fast enough such that release is not very sensitive to differences in the magnitude of pressure variation rate.

CONCLUSIONS

The current design of an earth-mounded concrete bunker with a drain pipe and connected piping for monitoring provides a direct pathway for the fast release of gaseous radionuclides to the atmosphere. For this release, the pumping effect of barometric pressure changes is far more important than diffusion as a release mechanism.

For the migration of ^{222}Rn, there appears to be virtually no escape of this radionuclide to the atmosphere through earthen cover materials as long as the clay layer is intact with enough moisture. This would be true whether the concrete barrier is intact or completely failed.

FUTURE WORK

For the transport of ^{222}Rn through concrete or earthen cover materials, absorption or surface adsorption of gas in the moisture can significantly affect the diffusion. The interaction of ^{222}Rn with water will be considered in future studies of ^{222}Rn transport through concrete and earthen materials.

To characterize gaseous release of radionuclides from a LLW disposal facility, analyses of ^{14}C and ^{3}H releases, which are the major components of radioactive gases in LLW disposal facilities, are essential. The transport and release of ^{14}C and ^{3}H containing gases will be modeled and analyzed in the future work. Potential source inventory reduction for these radionuclides through airborne releases will be also investigated.

Comparative performance evaluations will be exercised for the air and the water pathways to assess the relative importance of the gaseous release pathway to overall radiological performance assessment of a LLW disposal facility.

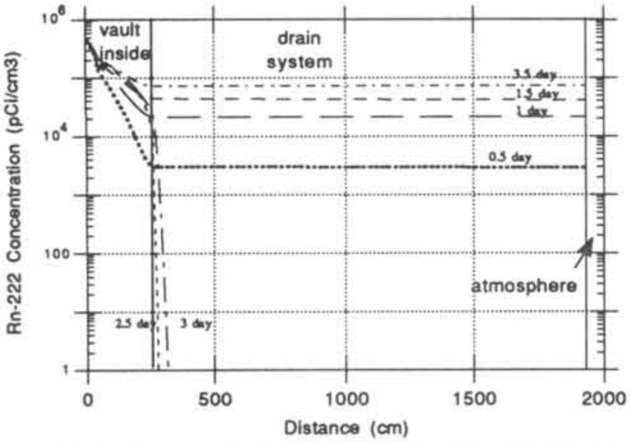

Fig. 5. Rn-222 concentrations within a facility (dP/dt = 3 sinwt (mb/hr), period = 3 days).

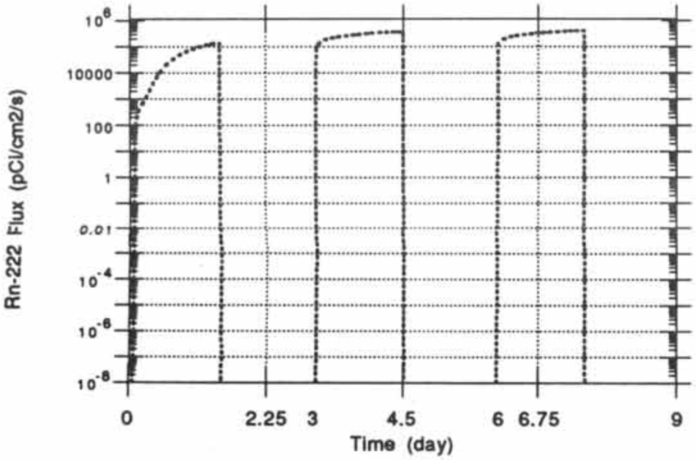

Fig. 6. Rn-222 surface flux changes (dP/dt = 3 sinwt (mb/hr), period = 3 days).

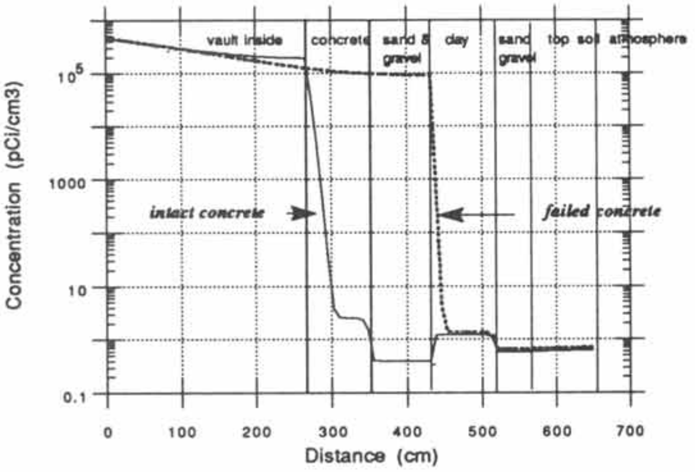

Fig. 7. Rn-222 concentration across concrete and earthen cover.

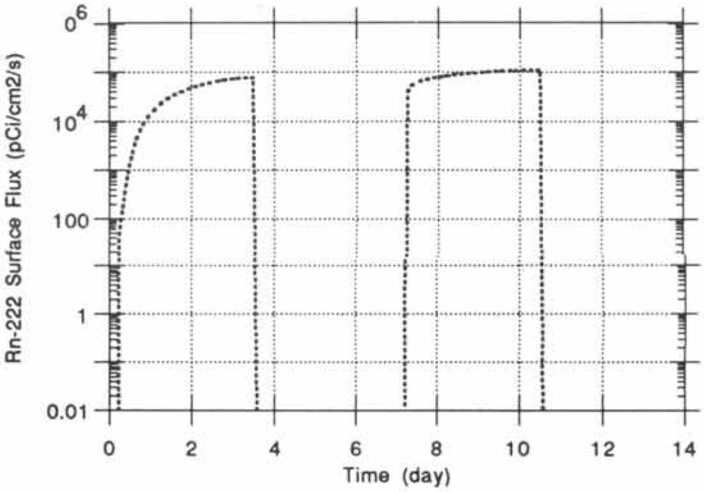

Fig. 8. Rn-222 surface flux changes (dP/dt = 0.7 sinwt (mb/hr), period = 7 days).

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