SENSITIVITY OF PERFORMANCE ASSESSMENT OF THE ENGINEERED BARRIERS TO NUANCES OF RELEASE RATE CRITERIA

Douglas L.R. Oliver

Mechanical Engineering Department
University of Toledo
Toledo, Ohio 43606
(419)537-2885

ABSTRACT

The United States Nuclear Regulatory Commission (NRC) has established criteria for the long-term performance of proposed high-level waste repositories. As with any regulation, the criteria may be interpreted in several ways. Due to the high capital costs and the emotional political climate associated with any high-level radioactive waste repository, it is important that there be an early consensus regarding interpretations of the criteria, and what assumptions may be used to demonstrate compliance with them. This work uses analytic solutions of mass transport theory to demonstrate how sensitive performance analyses are to various nuances of the NRC release rate criterion for the engineered barriers. The analysis is directed at the proposed repository in basalt at the Hanford site in Washington State.

INTRODUCTION

Before a license may be granted to operate a high level radioactive waste repository in the USA, the proposed repository sub-systems must be shown to meet various long term performance criteria. It is important that both the licensee (the DOE) and the licensor (the NRC) have a common understanding of these criteria. If significant differences of interpretations of the criteria exist after a repository has been designed, serious time delays and cost increases could result. For example, if a repository were designed using a liberal interpretation of a criterion, which the NRC did not originally accept, the repository might need to be redesigned. If not, the NRC would need to accept the more liberal interpretation of the criterion. In either case, serious problems could result. It might be too costly and time consuming to redesign the repository. Also, it might be politically impossible for the NRC to accept the more liberal interpretation at such a late date.

This problem can be avoided by raising technical issues regarding the criteria at an early date. The intent of this work is to raise three such issues which are related to the NRC release rate criterion for the engineered barriers system; 10 CFR 60.113, (1). Specifically, this work applies to the Hanford site in Washington State, (2). The issues raised may have relevance to other proposed repository designs, particularly to repositories located in a saturated medium.

The engineered barriers are to be designed such that the release rates of radionuclides are negligible for the first 300 to 1,000 years after emplacement. Thereafter, the release rates for each radionuclide are not to exceed certain limits, (10 CFR 60.113b). These limits (in Curies per year) are essentially the greater of either: one part in 100,000,000 of the total inventory after 1,000 years, or one part in 100,000 of the inventory for that radionuclide after 1,000 years, (3). For this work, the repository inventory will be assumed to be that listed on table 3.3.10 of Ref. (4), which has a total inventory after 1,000 years of 1.6E+3 Ci/MTHM. Thus, the release rate criterion allows for an annual release rate of the larger of: one part in 100,000 of the radionuclide inventory at 1,000 years or 1.6E-5 Ci/year per MTHM. The specific wording of the criterion follows:

"The release rate of any radionuclide from the engineered barrier system following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1,000 years following permanent closure, or such other fraction as may be approved or specified by the Commission; provided, that this requirement does not apply to any radionuclide which is released at a rate less than 0.1% of the calculated total release rate limit. The calculated total release rate limit shall be taken to be one part in 100,000 per year of the inventory of radioactive waste, originally emplaced in the underground facility, that remains after 1,000 years of radioactive decay." 10 CFR 60.113b

Simple analytic models are employed in this work which may be easily reproduced. The use of simple analytic models facilitates an understanding of gross trends. The penalty for using such simplified models is that some accuracy is lost. However, it is the author's experience that much of the raw data used as input for long term performance assessment models have such large uncertainties as to render the use of complex deterministic computer analyses of marginal value.

THREE ISSUES RELATED TO THE RELEASE CRITERIA

Issue #1: The amount of host rock for which credit may be taken.

Most interpretations of the above release rate criterion assume that the release rate from the

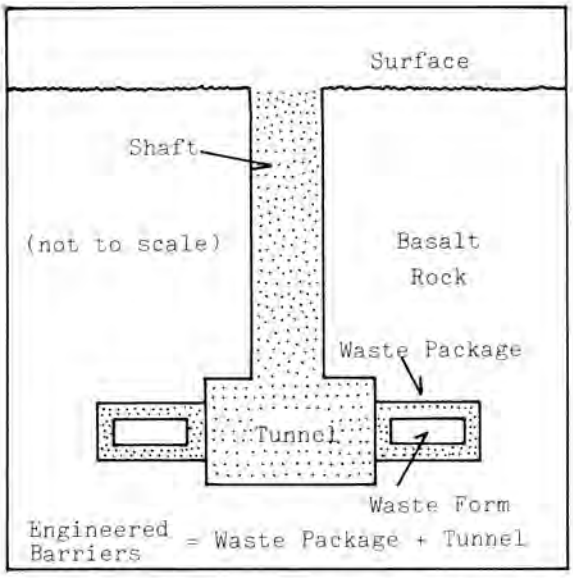

Fig. 1. Schematic of a Repository

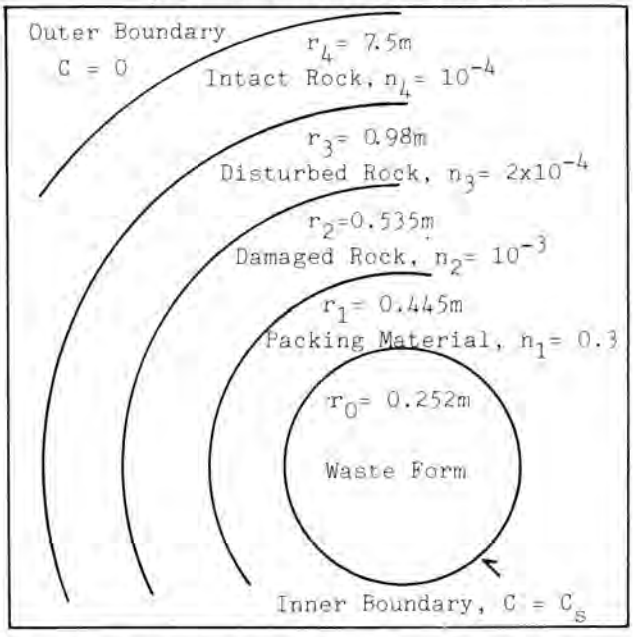

Fig. 2. Domain for the Release Rate Calculations in DOE, 1986.

engineered barriers should be measured at the interface between the engineered barriers and the intact host rock, (see Fig. 1). However, it is not clear how much credit may be taken for the host rock which surrounds the waste package, (as long as the release rate is measured at the packing material / host rock interface). Presently, the DOE is taking credit for a total of seven meters of host rock which surrounds the waste package. The amount of rock for which credit may be taken is of critical importance to the current DOE release rate models. This fact may be demonstrated using a simple steady-state analysis of a long-lived, solubility-limited radionuclide, such as Carbon-14. The current DOE model assumes that the primary transport mechanism near the waste package is radial diffusion through the packing material and several layers of rock, (see section 6.4.2.4.4 of Ref. (2)). Using the assumptions of the DOE, the steady-state release rate for a long-lived, solubility-limited radionuclide may be shown to be equal to, (Appendix A):

$$\text{Release Rate} = [2000\,\pi\,L\,C_s\,M\,SA] \,/\, [(\int_{r_o}^{r} \frac{dr}{D\,r})(INV)] \quad (1)$$

where: C_s is the average saturation concentration (moles/liter),

D is the effective diffusion coefficient (m^2/yr),

INV is the inventory of the wasteform in metric tonnes of heavy metal, (MTHM),

L is the length of the waste container (meters),

M is the atomic weight of the radionuclide,

SA is the specific activity (Ci/gram),

r_o and r, are the respective outer radii of the wasteform and the region of the rock for which credit is taken.

Equation 1 does not account for any transient effects. One of the most important differences between Eq. 1 and the analysis of the DOE, as presented in Ref. (2), is that the release rate predicted by Eq. 1 will not predict any transient peak release rate. For Carbon-14, a non-sorbing solubility-limited radionuclide, the transient peak release rate predicted by the DOE analysis is roughly double that of the steady-state rate predicted by Eq. 1. With this qualification, Eq. 1 may be used to demonstrate the sensitivity of the predicted release rate to the amount of host rock for which credit is

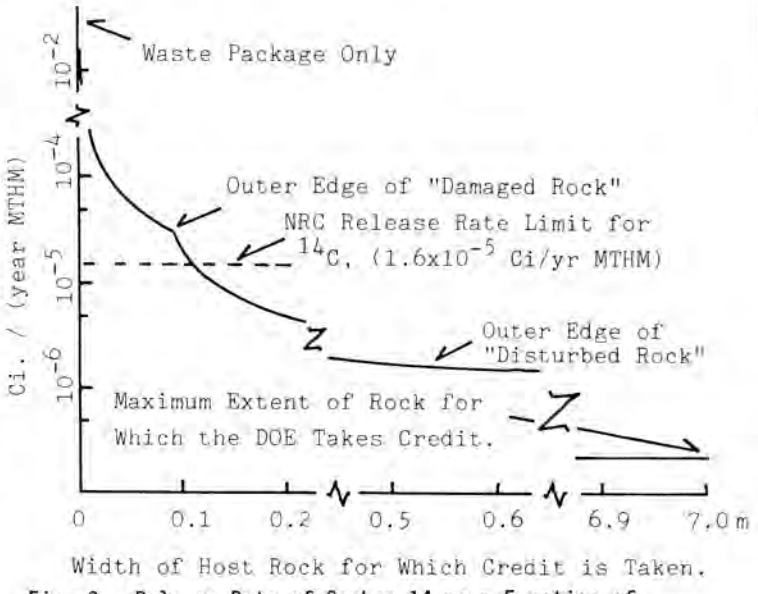

Fig. 3. Release Rate of Carbon-14 as a Function of the Width of the Host Rock for Which Credit is Taken.

Figure 3 demonstrates the effect of the thickness of the rock layer for which credit is

taken. The input data used in the calculations were based on data implied in Fig. 6-15 and Tables 6-24 through 6-26 of Ref. (2). The outer radius, at which a zero concentration boundary condition was imposed, was decreased in Eq. 1 until no credit was taken for the rock layer which surrounds the waste package. As Fig. 3 illustrates, the predicted release rates are a strong function of the thickness of the rock layer for which credit is taken. The assumed properties of the surrounding rock will also affect the predicted release rates. If significant credit is to be taken for the host rock, there should be a common understanding between the DOE and the NRC regarding how much credit may be taken, and what assumptions may be made regarding the properties of the host rock.

Issue #2; Production dominated radionuclides.

In the DOE analysis of radionuclide release from the waste package, radionuclides which have a half-life of less than 100 years were neglected from consideration. This was based on the assumption that any radionuclide with a half-life of less than 100 years would decay to insignificant levels before the container corroded. There is a set of radionuclides for which the levels of radioactivity increase with time. One of these radionuclides is Lead-210, which has a half life of about 22.3 years. The production rate and the inventory of Lead-210 increases with time, (Fig. 4). This class of radionuclides was excluded from the release rate analysis presented by the DOE.

Due to the short half-life of Lead-210, it has a very high specific activity (76 Ci/gram). Thus, the use of a solubility-limited analysis might seriously overpredict its release rate. The release is also likely to be limited by the rate of production of Lead-210. (Only as much lead may be released from the wasteform as is being produced in the wasteform). As Lead-210 diffuses from the wasteform, through the packing material and the surrounding rock, its inventory is continually being depleted by radioactive decay. Much of what is released from the wasteform will decay before it enters the host rock.

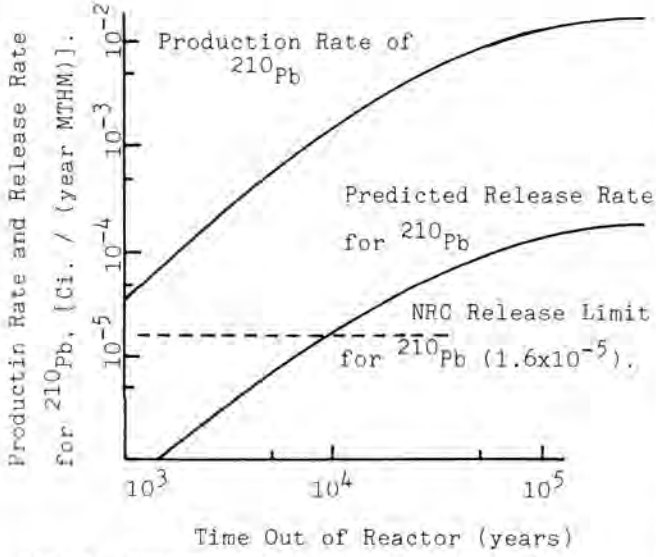

Fig. 4. Production Rate and Release Rate for Lead-210.

A simple equation may be derived to estimate the net release as a function of the radius, (see Appendix B). The analysis of Appendix B depends on accurate estimations of the porosity, the effective diffusion coefficients, etc. However, an estimate of the release rate of Lead-210 has been made using parameters detailed in Appendix B. On Fig. 5 the relative concentration level and the relative release rates are plotted as a function of the distance from the waste form surface.

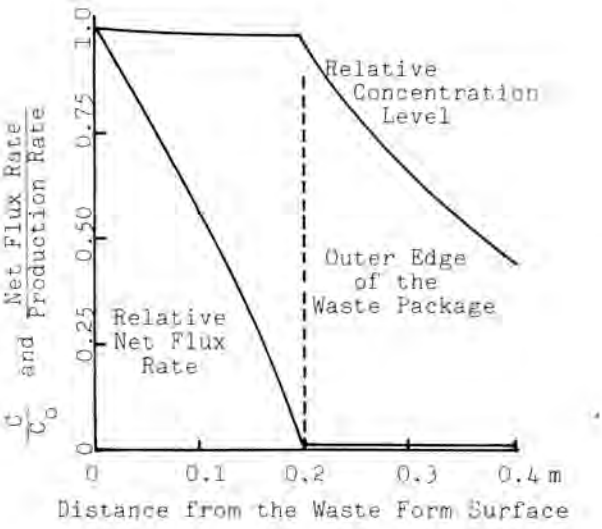

Fig. 5. Relative Concentration Levels and Release Rate for Lead-210.

Using the assumed parameters, 99.1% of the Lead-210 which is released from the wasteform will decay in the packing material. The remainder diffuses into the surrounding rock. Even if only 0.9% of the lead enters the rock, the NRC release rate criterion would be violated when the production rate exceeds 1.8E-3 Ci/(year x MTHM). This production rate is reached in about 10,000 years. The predicted release rates will increase with the production rate until the production rate begins to decline, after over 100,000 years, (Fig. 3).

The porosities and diffusion coefficients assumed for use in the preceeding analysis may be non-conservative with respect to the predicted release rates. However, phenomenon such as sorption in the packing material will encourage more rapid decay in the packing material. If credit is to be taken for sorption of lead in the packing material, it may be difficult to demonstrate the continued ability of the packing material to sorb lead after thousands of years of deposition and decay of Lead-210.

Although 10-CFR-60.113b does not specifically address the issue of production dominated radionuclides, such as Lead-210, the intent of the criterion does not seem to apply to those radionuclides which have inventories that increase with time. In addition, the criterion does not address the maximum duration for which it was intended to apply. From the above analysis, it is clear that there needs to be a common understanding of which radionuclides were intended to be regulated by 10 CFR 60.113b, and the duration for which the criterion applies.

Issue #3: Release through the borehole plug into the emplacement tunnel.

The repository design presented by the DOE in Ref. (2), proposes that the waste packages be placed in short horizontal boreholes. These boreholes are excavated from the side of a long horizontal emplacement tunnel. Each tunnel will have hundreds of these boreholes placed in pairs spaced about 6.7 meters apart, (Fig. 1). The analysis used by the DOE assumed that the primary transport mechanism for release from the wasteform will be radial diffusion. Inspection of Fig. 1 reveals an alternative release route, through the borehole plug into the emplacement room tunnel. Assuming (per the DOE model) that diffusion is the dominant transport mechanism in the region near the waste package, the steady-state release rate through the plug will be roughly:

Release rate =

$$(C_s - C_{tunnel})(SA \times M \times 1000 \times D \times area) \quad (2)$$

$$/ \ (length \times INV)$$

Data for the plug region are not generally available. However, the effective diffusion coefficient may be assumed to be similar to that of the packing material. From Table 6-26 of Ref. 2 the effective diffusion coefficient for the packing material may be shown to be 9.5E-3 m^2/year. The cross sectional area of the borehole is 0.62m^2. The length of the borehole plug is not explicitly given, it is assumed to be 1.0 meter. With these values, the release rate for Carbon-14 (assuming a mean solubility of 2.E-6 moles/liter), through the plug may be estimated to be about 4.E-4 Ci/(year x MTHM).

Comparison with Fig. 3 reveals that if the rock surrounding the wastepackage is as tight as is assumed by the DOE, then most of the radionuclides could exit the wastepackage through the borehole plug. Thus, the assumption that the primary path of radionuclide migration is only radial could lead to a gross underestimation of release rates. If release through the tunnel is modeled, the predicted release rates could be orders of magnitude different than those predicted by Eq. 1.

The above analysis could over-predict the release rate into the tunnel region if the radionuclide concentration level in the tunnel were near the saturation concentration. To estimate the concentration level in the tunnel, the radionuclide transport rates in the tunnel must be estimated. Estimation of the transport rates in the tunnel region is no small task, due to the heterogeneous nature of the excavation-damaged rock which would surround such a long (about 1 km) tunnel. If no credit is claimed for the isolation capability of the tunnel, then the appropriate (conservative) concentration level to assume for the tunnel region is the ambient concentration level (near zero). With this conservative assumption, it may be difficult to demonstrate that the NRC release rate criterion can be met.

There needs to be a clear understanding between the DOE and the NRC regarding which assumptions are appropriate with respect to release from the waste package into the tunnel region and thence from the repository.

CONCLUSION

Three issues have been raised regarding interpretations of the NRC release rate criterion for a high level radioactive waste repository in basalt. It was demonstrated that there was potential for diverse interpretations of how the criterion is to be applied. Since the criterion is quantitative in nature, it is important that all concerned parties come to an agreement regarding what the specific intents of the criterion are; and which assumptions may be used to demonstrate compliance with the criterion. Without such a common agreement, the release rates predicted by the various parties could vary by several orders of magnitude. While disagreements in interpretation are inevitable, they may be reduced and the negative consequences of these disagreements may be mitigated by raising relevant issues as early as possible.

REFERENCES

1. NRC, 10 Code of Federal Regulations (CFR), part 60.113, January 1, 1986.

2. DOE, DOE/RW-0070, "Environmental Assesssment - Hanford Site," United States Department of Energy / Office of Civilian Radioactive Waste Management, vol. II, May 1986.

3. Bensky, M.S. and D. L. Oliver, "Transient Diffusional Release from Waste Packages in a Repository in Basalt," Scientific Basis for Nuclear Waste Management IX, Sponsored by the Materials Research Society; Stockholm, Sweden, September, 1985.

4. DOE, DOE/ET-0028, "Technology for Commercial Radioactive Waste Management," vol. I, 1979.

Appendix A: Release Rate for Steady Radial Diffusion of a Non-Decaying Solute.

Fick's law for diffusion through a porous medium may be written as:

$$Flux = -D\frac{dC}{dx}, \quad A\text{-}1$$

where: C is the concentration in (moles/liter),
D is the effective diffusion coefficient (m^2/ year),
x is a generic coordinate (m).

For the special case of steady-radial diffusion of a long-lived, solubility-limited radionuclide, Eq. A-1 may be written as:

$$(Flux_{net}) = -2\pi L[D\ r\ \frac{dC}{dr}], \quad A\text{-}2$$

where: r is the radial coordinate (m),
L is the wastepackage length (m).

For a steady-state solution, the net flux is constant (not a function of time or radial coordinate), thus Eq. A-2 may be replaced by:

$$(Flux_{net}) = 2\pi L[C(r_o)-C(r)]/[\int_{r_o}^{r} \frac{dr}{Dr}], \quad A\text{-}3$$

where: r_o is the outer radius of the wasteform, (0.252 m).

Equation A-3 has dimensions of (moles x m^3)/(liter x year). An equivalent form of Eq. A-3 that is compatable with the release rate criterion, (Ci / year MTHM) is:

$$\text{Release Rate} = [2000\pi L\ SA\ M][C(r_o)-C(r)] \Big/ [(INV)_{r_o} \int^{r} \frac{dr}{Dr}]. \qquad A\text{-}4$$

Where: M is the atomic weight (grams/mole),
SA is the specific activity (Curies/gram),
INV is the wasteform inventory (MTHM).

The specific parameters used in this analysis are based on the parameters implied in section 6.4.2.4.4 of Ref. 2. The rock region was modeled as three connected annuli. The packing material was modeled as an annuli surrounded by the rock (Fig. 2). The effective diffusion coefficients used for each region were the product of the porosity and the diffusion coefficients as listed on Table 6-26 of Ref. 2. The concentration level at the wasteform surface assumed for Carbon-14 was the mean of the solubility range listed on Table 6-25 of the same work, (2.E-6 moles/liter). The concentration level imposed at the outer radius was zero. The specific activity (SA), of Carbon-14 is 4.5 Curies per gram. The wasteform is assumed to contain 1.8 metric tonnes of heavy metal, (INV = 1.8 MTHM). The wastepackage length was assumed to be four meters, (L = 4m).

Appendix B: Release of a Production-Limited Radionuclide.

The steady-state equation for radial diffusion (cylindrical coordinates), of a non-sorbing species with radioactive decay is:

$$D\left(\frac{d^2C}{dr^2} + \frac{1}{r}\frac{dC}{dr}\right) = \lambda n C\ , \qquad B\text{-}1$$

where: D is the effective diffusion coefficient,
n is the porosity, and
λ is the decay constant.

In the previous analysis, several discrete rock regions were considered (i.e. the "damaged rock", "disturbed rock", and "undisturbed rock" regions). For this analysis, the rock will be treated as a homogeneous region. That is the "damaged rock" will be assumed to be infinite in extent. This simplification is not required to obtain an analytic solution to Eq. B-1, but it significantly reduces the complexity of the analysis. If the rock is assumed to be a homogeneous region of an infinite thickness, the boundary conditions imposed on Eq. B-1 are:

$$\text{Net Production Rate} = \text{Net Release Rate} = -2\pi L\, D_o r_o \left.\frac{dC}{dr}\right|_{r=r_o}\ , \qquad B\text{-}2$$

$$C(r_1^-) = C(r_1^+), \qquad B\text{-}3$$

$$D_o \left.\frac{dC}{dr}\right|_{r=r_1^-} = D_1 \left.\frac{dC}{dr}\right|_{r=r_1^+}\ , \qquad B\text{-}4$$

$$\lim_{r \to \infty} C(r) = 0, \text{ (due to radioactive decay)}, \qquad B\text{-}5$$

where the subscripts o and 1 denote the packing material and the rock parameters respectively. Also, r_o and r_1 denote the inner radii of the packing material and the rock regions.

Equation B-1 may be transformed using the following dimensionless parameters:

$$Da = \frac{n_o r_o^2 \lambda}{D_o} \quad \text{(Damkoeler number)},$$

$$f = D_o/D_1 \quad \text{(ratio of effective diffusion coefficients)},$$

$$R = r/r_o \quad \text{(dimensionless radius)},$$

$$\alpha = \frac{n_1 D_o}{n_o D_1}$$

$$\frac{d^2C}{dR^2} + \frac{1}{R}\frac{dC}{dR} = Da\ C, \quad 1 < R < R_1\ , \qquad B\text{-}6$$

$$\frac{d^2C}{dR^2} + \frac{1}{R}\frac{dC}{dR} = \alpha\ Da\ C, \quad R > R_1\ . \qquad B\text{-}7$$

The boundary conditions imposed on Eqs. B-6 and B-7 are:

$$\text{Net Production Rate} = -2\pi L\, D_o \left.\frac{dC}{dR}\right|_{R=1}, \qquad B\text{-}8$$

$$f\left.\frac{dC}{dR}\right|_{R=R_1^-} - \left.\frac{dC}{dR}\right|_{R=R_1^+} = 0, \qquad B\text{-}9$$

$$C(R_1^-) - C(R_1^+) = 0, \qquad B\text{-}10$$

$$\lim_{R \to \infty} C(R) = 0. \qquad B\text{-}11$$

The solution to Eqs. B-6 and B-7 is:

$$C(R) = A_1 I_o(\sqrt{Da}\ R) + A_2 K_o(\sqrt{Da}\ R),\ 1 < R < R_1, \qquad B\text{-}12$$

$$C(R) = A_3 K_o(\sqrt{\alpha\ Da}\ R), \quad R > R_1, \qquad B\text{-}13$$

where: $I_o(R)$ is the modified Bessel function of the first kind and $K_o(R)$ is the modified Bessel function of the second kind.

If the coefficients A_1, A_2, and A_3 are placed into a vector format, the vector $\tilde{A}_j$ is given by:

$$\tilde{A}_j = \begin{Bmatrix} A_1 \\ A_2 \\ A_3 \end{Bmatrix},$$

with $\tilde{A}_j$ satisifying the following set of equations:

$$\tilde{M}_{ij}\ \tilde{A}_j = \tilde{d}_i, \text{ with;}$$

$$M_{11} = \sqrt{Da}\ I_1(\sqrt{Da}\ ), \quad M_{12} = -\sqrt{Da}\ K_1(\sqrt{Da}\ ),$$

$$M_{13} = 0,$$

$$M_{21} = f\ I_1(\sqrt{Da}\ R_1), \quad M_{22} = -f\ K_1(\sqrt{Da}\ R_1),$$

$$M_{23} = \sqrt{\alpha} K_1(\sqrt{\alpha Da}\ R_1);$$

$$M_{31} = I_o(\sqrt{Da}\ R_1), \quad M_{32} = K_o(\sqrt{Da}\ R_1),$$

$$M_{33} = -K_o(\sqrt{\alpha Da}\ R_1),$$

and,

$$\tilde{d}_j = \begin{Bmatrix} [\text{net production rate}]/[-2\pi\ L\ D_o] \\ 0 \\ 0 \end{Bmatrix}$$

The ratio of the net release across any concentric surface to the production rate may be shown to be:

Release/Production = B-15

$= R\ [A_1 I_1(\sqrt{Da}\ R) - A_2 K_1(\sqrt{Da}\ R)]/F,\ 1 < R < R_1$,

$= -R\ A_3\sqrt{\alpha}\ K_1\ (\sqrt{\alpha Da}\ R)/\ (f\ F),\ R > R_1$,

where: $F = A_1 I_1(\sqrt{Da}) - A_2 K_1(\sqrt{Da})$.

The specific parameters used in this analysis are:

	packing material	rock
inner radius; (meters)	r_o = .25m	r_1 = .45m
porosity;	n_o = 0.3	n_1 = 1.E-3
effective diffusion coefficient; (m^2/year)	D_o = 9.5E-3	D_1 = 3.2E-6