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POTENTIAL INCREASES IN NATURAL RADON EMISSIONS DUE TO HEATING OF THE YUCCA MOUNTAIN ROCK MASS*

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ABSTRACT

Heating of the rock mass by the spent fuel in the proposed repository at Yucca Mountain will cause extra amounts of natural radon to diffuse into the fracture system and to migrate faster to the accessible environment. Indeed, free-convection currents due to heating will act to shorten the radon travel times and will cause larger releases than would be possible under undisturbed conditions. To estimate the amount of additional radon released due to heating of the Yucca Mountain rock mass, we obtain an expression for the release enhancement factor, E. This factor is defined as the ratio between the total flux of radon at the surface of the mountain before and after closure of the repository assuming the only cause of disturbance to be the heating of the rock mass. With appropriate approximations and using a heat load representative of that expected at Yucca Mountain, the present calculations indicate that the average enhancement factor over the first 10,000 years will be 4.5 as a minimum. These calculations are based on the assumption that barometric pumping does not significantly influence radon release. The latter assumption will need to be substantiated.

I. INTRODUCTION

Radon is a noble gas and it is not significantly held up during transport with earth geofluids. A recent survey of 41 springs and wells in Sweden[1] showed that ^{222}Rn activity levels are one thousand to ten thousand times those of ^{226}Ra, its immediate progenitor. This thousands fold discrepancy derives from radon's inertness and, hence, high mobility. Radon level anomalies over sources at significant depths (up to 600 meters) in sedimentary rock sequences have also been documented.[2]

As a member of the natural uranium decay series ^{222}Rn is being continually produced within the rock mass at Yucca Mountain which is the proposed site for the nation's high level waste repository. A fraction of the radon is available for release and may reach the accessible environment. A clear indication of radon availability at Yucca Mountain comes from recent measurements[3] which show a ^{222}Rn activity of 670±30 dpm/liter in J-13 well water. Laul also reports that the radon activity is roughly 65,000 times that of its immediate progenitor, ^{226}Ra. The latter finding provides further indication of the ease with which radon may migrate within the fracture system at Yucca Mountain. Additional information is found in a survey[4] of radon and radon daughters concentrations in Rainier Mesa tunnels at the Nevada Test Site (the G and N tunnel, essentially). The survey reports that the natural uranium concentration in the non-welded tuff of the N tunnel and in the welded tuff of the G tunnel is roughly 4 ppm, i.e., 1.5 μCi/g. Equilibrium radon concentrations in the air of the two tunnels were found to be roughly 25 pCi/ℓ and 200 pCi/ℓ, respectively, reflecting the fact that the non-welded tuff is only slightly fractured with respect to the welded tuff. Transient times for reaching these concentration levels, after ventilation of the tunnels, are of the order of a few days, again indicating relatively fast radon migration times.

The in-situ transport of ^{222}Rn in fractured rock has been studied in an abandoned mine.[5] It was determined that *"large scale cracks of surface density of the order of several (6±3) cracks per square meter dominate flow."* It is known that at Yucca Mountain the rock is highly fractured[6] and that several orders of magnitude difference have been reported between laboratory measurements of permeability and the field measurements of in situ permeability[7] with water as the reference fluids. Recent laboratory and large scale measurements of gaseous

* This work was performed under the auspices of the U.S. Department of Energy.

diffusivity at the Nevada Test Site (NTS) confirm that the intrinsic permeabilities of large bodies of fractured volcanic rock are orders of magnitude larger than laboratory samples.[8] At the NTS, typical values of the large-scale permeability-to-porosity ratio are in the range $2\text{-}300 \cdot 10^{-11}\ m^2$.

Heating of the rock mass by the waste will cause extra amounts of radon to diffuse into the fracture system at Yucca Mountain and will cause radon to migrate faster to the accessible environment. Indeed, free-convection currents due to heating will act to shorten the radon travel times and will cause larger releases than would be possible under undisturbed conditions. To estimate the amount of additional radon released due to heating of the Yucca Mountain rock mass, we obtain an expression for the release enhancement factor, E. This factor is defined as the ratio between the total flux of radon at the surface of the mountain after and prior to closure of the repository assuming the only cause of disturbance to be the heating of the rock mass.

Section 2 outlines the approach used to obtain the enhancement factor. The full derivation for this factor can be found in Reference 9.

Section 3 presents the results of this modeling study.

Section 4 presents our conclusions.

II. EXPRESSION FOR THE ENHANCEMENT FACTOR

A. Modeling Assumptions

Our modeling approach is based on conservation of mass, the ideal gas law, and on the following ancillary assumptions. Namely:

(a) that radon travels as a passive impurity in air,

(b) that, at all locations, air is mostly dry and in thermal equilibrium with the surrounding medium,

(c) that the critical conditions for the onset of free-convection currents are always realized,

(d) that the temperature profile within the mountain is adequately described by the heat conduction equation with a time-dependent source,

(e) that radon moves only due to buoyancy forces. Barometric pumping is assumed to not influence net flow.

Assumptions (a) and (b) are non-conservative, i.e., they do not represent worst-case scenarios. In particular, under assumption (a) we limit our study to the analysis of convection effects neglecting the binary diffusion of radon in air. Under assumption (b) we have less density difference (buoyancy) than if the air temperature were not to decrease while moving upwards in the mountain. Furthermore, dry air is less dense than air saturated with water. Therefore, this assumption leads to a smaller predicted buoyancy flow.

The problem of evaluating the reduction of the radon travel times is made one-dimensional by concentrating on the cylindrical section of the mountain which cuts through the center of the repository. Convective currents due to buoyancy will be largest within a symmetric section passing through the center of the repository because, at all elevations, temperature is highest next to the centerline. The latter condition will also tend to produce a chimney effect whereby lateral flow will occur towards the centerline to join the vertical flow which takes place parallel to it. Effectively, this will increase the amount of radon exhaling within the neighborhood of the repository centerline. By neglecting lateral flow (3-D effects) and in concert with the other assumptions, we obtain a lower bound to the enhancement factor parameter, E.

B. Reference Equations

Our ultimate concern is to estimate the additional amount of radon that will be released from the mountain due to heating. The steady-state radon flux emanating from a section of the mountain of thickness (H - z) can be estimated from the expression:

$$I(z) = \int_z^H J(z)dz = \int_z^H \varepsilon(z)\ P(z)\ \exp[-\lambda t_f(z)]\ dz. \qquad (1)$$

where H represents the elevation of the Yucca Mountain surface above the repository, P(z) is the number of radon particles produced per unit area and unit time at elevation z, $\varepsilon(z)$ is the fraction of those particles that become available for upward migration, λ is the radioactive decay constant, and t_f is the time it takes a particle at elevation z to travel to the surface.

The travel time is a function of the properties of the rock, such as available flow area, hydraulic conductivity, etc., and fluid properties such as density and viscosity. For a homogeneous region the travel time is simply the thickness divided by the average velocity. A discussion of travel times is presented in Reference 9.

Heating of the gas in the pores and fractures causes the gas to become less dense. This in turn causes buoyancy driven flow. At the repository level, the temperature is largest and the density contrast is greatest and therefore, the velocity is highest. The effective velocity at the repository level due to buoyancy driven flow under quasi-hydrostatic conditions is a function of the density contrast and is given by the equation:[10]

$$V_R = -(K/f) \cdot (\rho_R/\rho^* - 1) \qquad (2)$$

where the parameter K is the pneumatic conductivity of Yucca Mountain at the repository level, f is the fraction of the area available for gas flow (flow porosity), ρ_R is the density at the repository elevation, and ρ^* is the reference density. Selection of the appropriate density is discussed later in this section.

The pneumatic conductivity is given by the expression:

$$K = \frac{k\ g\ \rho(z)}{\mu(z)}, \qquad (3)$$

where the parameter k is the bulk intrinsic permeability of the rock mass, and μ the dynamic viscosity of air.

To obtain the velocity at any location in Yucca Mountain, conservation of mass is used. The velocity at any elevation z is:

$$V(z) = \frac{\rho_R\ V_R\ A_R}{\rho(z)\ A(z)}, \qquad (4)$$

where A_R is the effective flow area at the repository level and A(z) is a similar parameter at elevation z. As a first approximation we assume that the flow area is constant throughout the mountain.

To obtain the velocity in Eqn. (4) requires the density as a function of z. Using the ideal gas law, this can be determined once the temperature profile within the mountain is known. An estimate for the temperature profile is presented in Reference 9 and is based on the solution of the heat conduction equation for a time-varying plane source embedded in an infinite medium. The heat source was estimated based on an areal heat load of 55 kW/acre at emplacement with 10 year old PWR spent fuel having a burnup of 33,000 MWd/MTHM. A fixed temperature at the surface of the mountain is achieved using the method of images.

Upon examining Eqns. (2-3), it is readily realized that the travel times from the various hydrogeologic units to the surface of Yucca Mountain are highly sensitive to the values of the ratios (k/f). Recent large-scale measurements of the bulk pneumatic diffusivity of air at the Nevada Test Site indicate that typical values of the ratio (k/f) for large bodies of volcanic rocks (tuff) are in the range $2\text{-}300 \cdot 10^{-11}\ m^2$. However, there is much uncertainty involved in the flow parameters at Yucca Mountain. In order to estimate the additional amount of radon released due to heating without accurate knowledge of these parameters, we define an enhancement factor for radon release, E, which is the ratio of the radon flux from the surface of the mountain after and prior to heating. Namely:

$$E = \frac{I_2(z)}{I_1(z)} \qquad (5)$$

Where the subscripts 1 and 2 refer to prior to and after heating respectively and I is the integrated flux out of the mountain given by Eqn. (1). This parameter will be shown to be largely insensitive to the hydraulic properties of the rock mass.

As an approximation, we treat the Yucca Mountain system as a homogeneous rock mass in determining the enhancement factor, i.e., P(z) is constant and the travel time is found from the expression:

$$t_f(z) = (H-z)/V_{av}(z) \qquad (6)$$

Where $V_{av}(z)$ represents the average effective velocity of an air particle traveling from elevation z to the surface of the mountain. Namely:

$$V_{av}(z) = \frac{\int_z^H V(z)\ dz}{\int_z^H dz} \qquad (7)$$

where V(z) is evaluated using Eqn. (4). From Eqn. (4) it is determined that the spatial dependence of V(z) is caused by and inversely proportional to changes in density. To obtain a lower bound on the enhancement factor, E, we assume a constant value for V(z) determined from the highest density. The highest density occurs at the lowest temperature which is at the surface of the mountain. In this case,

$$V_{av} = V_H = \frac{\rho_R\ V_R}{\rho_H} \qquad (8)$$

The assumption that V(z) is constant is not exact. However, applying conservation of mass, Eqn. (4) and recognizing that the density changes slowly with temperature, it can be shown that the velocity will decrease by less than 20% from the repository horizon to the surface of the mountain. Further, in deriving Eqn. (9) we have taken the value for the velocity to be the surface velocity. This is the lowest value and. therefore, gives the smallest value for the enhancement factor.

Using Eqns. (2, 3, 6, and 8) in Eqn. (5) gives:

$$E = \frac{\varepsilon_2}{\varepsilon_1} \cdot \frac{\mu_{R,1}}{\mu_{R,2}} \cdot \frac{\rho_{R,2}}{\rho_{R,1}} \cdot \frac{\rho_{R,2} - \rho^*_2}{\rho_{R,1} - \rho^*_1} \qquad (9)$$

where (a) the subscripts 1 and 2 refer to physical conditions before and after heating of the mountain, respectively; (b) the subscript R indicates repository-level conditions; (c) the parameter ε represents the ^{222}Rn fraction which, on average, becomes available for migration after its generation in the rock units extending from the repository horizon to the accessible environment; (d) the parameter μ represents the dynamic viscosity of air; (e) the parameter ρ represents the density of air; and (f) the parameter ρ^* is the reference density of air against which the density contrast (buoyancy) is calculated.

The reference density against which the buoyancy driven flow is calculated can be taken as the average density in the mountain prior to heating, or the average density at the surface of the mountain. Our calculations have shown that the enhancement factor is insensitive to the choice of reference density. Therefore, as a first approximation, we take the reference density as air saturated with water-vapor at 20°C, (the surface temperature), i.e., $\rho^* = \rho_{atm} = 0.00107$ g/cm^3. This reference density applies to both before and after heating.

The ratio $\varepsilon_2/\varepsilon_1$ is a number greater than one because, after heating, more radon is able to diffuse from the rock pores into the fracture system. However, since ε is proportional to the square root of the diffusion coefficient and since the binary diffusion coefficient of a gas depends on temperature as $T^{1.75}$, where T is expressed in degrees Kelvin, the ratio $\varepsilon_2/\varepsilon_1$ is very close to unity. On the other hand, the ratio $\mu_{R,1}/\mu_{R,2}$ is smaller than unity and varies between 0.8 and 1, roughly. Thus an approximate reference expression for E is as follows:

$$E = \frac{\rho_{R,2}}{\rho_{R,1}} \cdot \frac{\rho_{R,2} - \rho_{atm}}{\rho_{R,1} - \rho_{atm}} \qquad (10)$$

The latter expression does not depend on the flow parameters of the geologic system.

III. PREDICTIONS OF THE ENHANCEMENT FACTOR

To evaluate the terms in Eqn. (10) requires an estimate for the reference densities and the density of air in the repository as a function of time. As previously discussed, the reference density is taken to be the atmospheric density of air saturated with water-vapor at 20°C

The density at the repository horizon prior to heating is taken to be air saturated with water-vapor at 300°K, approximately the expected pre-emplacement temperature. The density at the repository horizon after emplacement requires knowledge of the average temperature at this location as a function of time. This has been estimated by solution of the transient heat conduction equation with a time-dependent heat source representative of 10 year old PWR spent fuel with a burnup of 33,000 MWd/MTHM at emplacement.[9] The initial area heat loading was chosen to be 55 kW/acre. The results of the calculation are presented in Figure 1 for times between 200 and 10,000 years. This calculation provides the **average** temperature at the repository horizon. Temperatures at the bore hole wall and near to the waste package will be higher.

The predicted temperatures presented in Fig. 1 were fit to a second order polynomial using the method of least squares. The resulting expression for the average repository temperature (°C) is:

$$T(t) = 97.01 - 7.25 \cdot 10^{-3} \cdot t + 3.37 \cdot 10^{-7} \cdot t^2 \qquad (11)$$

where t is the time in years. This correlation is only valid for $200 \le t \le 10{,}000$.

Equation (11) was used to provide the temperature needed in the ideal gas law to evaluate the air density at the repository horizon as a function of time.

With the reference and repository air densities known, the minimum enhancement factor was calculated and is presented in Figure 2. Examining Fig. 2 it can be seen that heating of the repository will increase radon release from the mountain for the entire 10,000 year period. At 200 years, the enhancement factor is approximately 5.6. By 10,000 years it has decreased to 3.9. This analysis implies that heating of the rock without any release from the waste itself will cause a factor of four to five increase in the amount of radon exhalating from Yucca Mountain.

The EPA regulations pertaining to HLW are written in terms of cumulative release in a 10,000 year period. Therefore, the average enhancement factor is an important parameter. The average enhancement factor is:

$$E_{avg} = \frac{\int E(t)dt}{\int dt} \qquad (12)$$

Performing this integration numerically using the data in Fig. 2 from 200 to 10,000 years gives an average enhancement factor of 4.5 during this period. *In other words, if 1 curie were to be released during 10,000 years without a heat source, 4.5 curies would be released if the heat source were present.*

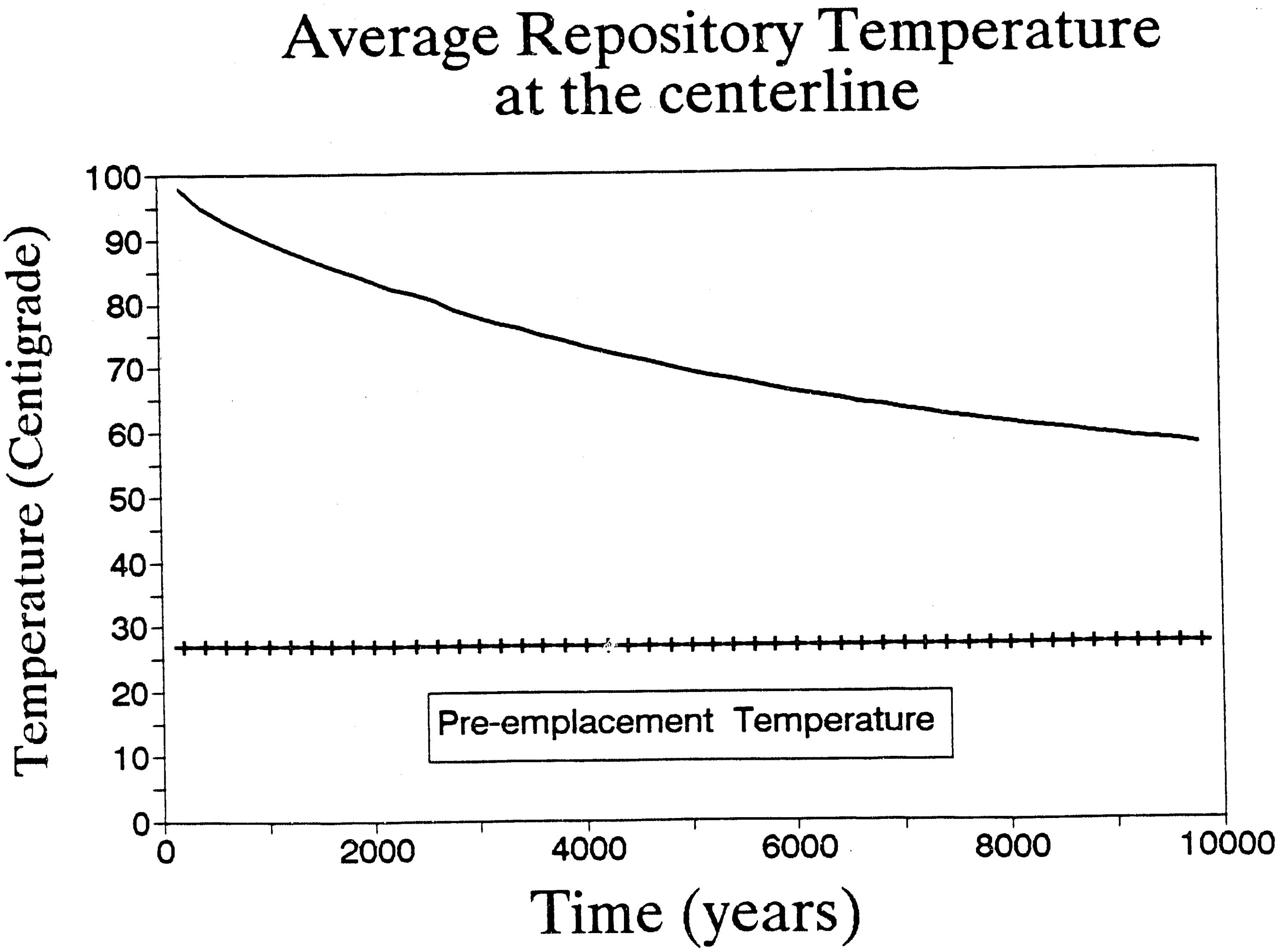

Figure 1 Average repository temperature as a function of time. Heat source is 10 years old PWR spent fuel with a burnup of 33,000 Mwd/MTHM.

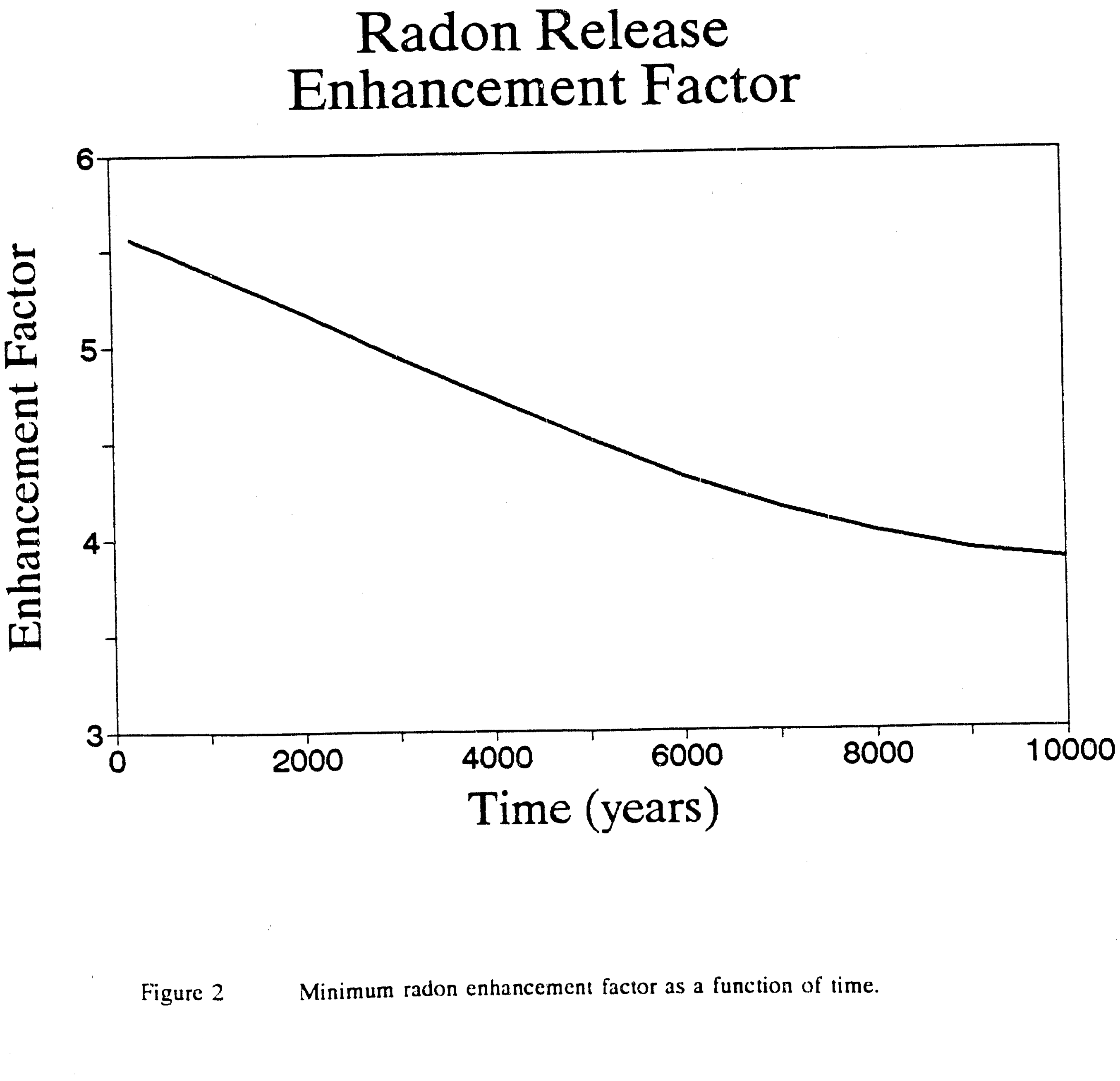

Figure 2 Minimum radon enhancement factor as a function of time.

Discussion

The enhancement factor is defined as the ratio of the radon flux at the surface of the mountain after and before heating. Normalization of the release rate to the value prior to emplacement has the advantage of being largely insensitive to the hydraulic flow properties of the mountain. If the radon flux levels at Yucca Mountain were measured now, an estimate for the radon release after heating could be obtained without detailed knowledge of the flow properties.

As defined by Eqn. (10) the enhancement factor depends most prominently on the average repository temperature. From this, we conclude that the area thermal load is the key parameter in this model.

Scoping calculations aimed at estimating the minimum increase in release due to heating indicate that radon release will increase by a minimum of 4 to 5 times due to heating of the mountain by the HLW.

The preliminary calculations are based on a number of assumptions that permit the minimum increase to be obtained. These assumptions include:

a) use of a constant velocity equal to the exit velocity at the surface of the mountain.

b) neglecting 3-D flow which would tend to have higher flow rates above the center of the repository where the temperatures are highest, i.e., a chimney effect.

c) treating the mountain as a homogeneous medium. Yucca Mountain is a fractured system. It is possible that much of the gas flow will be carried within just a few fractures. This may lead to local hot spots which show substantial radon release.

d) ignoring the contributions of radon from below the repository level and above the water table.

e) treating the air in the Mountain as mostly dry and, therefore, heavier than air saturated with water vapor.

Further refinements to the calculations which would relax any of the above assumptions could lead to a prediction of a larger enhancement factor. Other refinements such as examining the effects of different areal heat loadings (e.g. older spent fuel, different burn up spent fuel, different waste container spacing, etc.) and the choice of the reference density should be examined.

A major assumption of this work is that barometric pumping does not play a significant role in determining radon transport. It is known that barometric pressure changes will cause pressure waves to move several hundreds of meters within fractured rock systems. The role that these pressure waves have in transporting radon at Yucca Mountain requires further study.

IV. CONCLUSIONS

We have developed an estimate of the minimum expected increase in radon release from Yucca Mountain due to buoyancy driven flow caused by the repository heat load. This estimate is based on conservation of mass and the ideal gas law.

Our scoping calculations support the conclusion that the heating of the rock mass at Yucca Mountain may cause significant radon levels anomalies at the surface of the mountain. On the average, these anomalies will be at least four or five times greater than the current background. Further refinements, planned as part of future studies, to our calculations are likely to predict higher releases.

One area that requires particular attention is the role of barometric pumping on radon release. If barometric pumping is an efficient mechanism for removing radon from Yucca Mountain, then heating of the repository may only cause minimal increases in release. The role barometric pumping plays will be examined in future studies.

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