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Long-Term Prediction of Radionuclide Leaching from Waste Matrix by Finite-Slab Approximation Method

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유한 격판 근사 방법에 의한 고화체로부터의 방사성 핵종의 용출율 장기 예측

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Abstract

A finite slab approximation method was developed to predict the long-term leachability. It is based on the assumption that the diffusional characteristics of radionuclides in a waste matrix are not dependent on matrix geometry but dependent on volume to surface ratio (V/S) and diffusion coefficient. Consequently it can be expressed as the solution of the equations obtained from a finite slab with an equal V/S ratio (imaginary diffusion length).

The calculational results by the finite slab approximation method have been compared with the results obtained for finite cylinder and sphere with corresponding diffusional analysis. The results of this simple model have showed a good agreement and presented a general applicability for the long-term prediction of the radionuclide leaching behavior.

요 약

장기 용출율을 예측하기 위하여 유한 격판 근사 방법이 개발되었다. 이 방법은 폐기물 고화체에서의 방사성 동위원소 확산 특성이 고화체 형태에 관련되지 않고 체적/면적비 (V/S)와 확산계수에만 의존한다는 가정에 근거하고 있다. 결과적으로 용출율은 동일 체적/면적비를 갖는 유한 격판을 기술하는 방정식의 해로 표시할 수 있다.

유한 격판 근사 방법을 사용한 계산 결과는 유한 원통과 유한 구형에 관한 확산 해석에 관한 해와 비교 되었다.

여기서 도출된 단순 모델은 다른 모델과의 비교 결과 잘 일치하고 있고 방사성 핵종의 용출 현상에 관한 장기 예측에 전반적인 응용이 가능한 것을 보여준다.

1. Introduction

Generally, low and intermediate-level radioactive wastes have been solidified by using several binding agents, i.e., cement, polymer, bitumen, etc.. When those are disposed of in repository area, leaching process by ground water has been found to be one of the major ways by which the radionuclides may be released from the waste container to the environment. To provide engineering data for the design of waste disposal site and to simulate long-term risks to human environment, three quantities of radionuclide leaching characteristics must be identified, i.e., the amount of radionuclide in the waste matrix, the amount in the environment, and the leach rate from the matrix. Here the environment means the just outside the matrix.

Due to the restriction of the test time, many investigators[1-5] have extended their results from short-term test to predict the long-term leaching. Most of the experimental results[2,6,7] on the leaching of radionuclides from the waste matrix have indicated that the dominant mechanism of leaching process is a diffusion, therefore the leachability can be expressed as a function of diffusion coeficient, although the rate controlling step of leaching mechanism remains unidentified.

Numerous papers[3,4,5,8,9] have been published to presdict the long-term leachability of radionuclide from a solidified waste matirx by employing mathematical or empirical models. Empirical models are the simplest and the results of mathematical models are limited to a waste matrix in the form of cylinder or sphere.

In the case when analytic solutions does not exist for the actual geometry of waste matrix, or even though when there exists a solution it requires tedious computational summations of infinite series and/or all of the expression for the three leaching quantities cannot be obtained simultaneously, the simple approximation method by semi-finite slab model has been generally applied to the prediction of long-term leaching. Because this approximation generates a large calculational uncertainty with time, new method with general applicability is required for the long-term prediction.

Although, the amount of radionuclide in the environment was suggested in the general form of equation by Bell[10], other quantities, i.e., the amount in the matrix and the leach rate from it, has not been proposed in the same manner.

So, the object of this study was to develop an approximation method which has a general applicability from the results of short-term test.

General Equations for Leachability

Several studies[2,6,7] have suggested that leaching of radionuclide from a waste matrix mainly depends on the diffusion mechanism. If diffusion coefficient is constant with respect to time and space, the general equation of radionuclide leaching is

$$\frac{\partial C}{\partial t}=D\nabla^2 C-\lambda C \tag{1}$$

$$C(r,0)=C_0 \tag{2}$$

$$C(R,t)=0 \tag{3}$$

where

C: the concentration of radionuclide inside the matrix

D: the diffusion coefficient

λ : decay constant

r: position vector within matrix

R: position vector at the surface of matrix, and

C_0: initial concentration of radionuclide in the matrix

And it may also be subject to another condition, for example, zero flux across a boundary or an axis of symmetry.

Theoretical analyses[11,12,13] have suggested that the equation (1) can be readily solved by taking the transformation

$$C(r,t) = \Psi(r,t)exp(-\lambda t)$$

where, $\Psi(r,t)$ is the solution for the case of stable isotope.

The general equation for the fractional leach rate, FR(t), of radionuclide from waste matrix was derived by Bell[10],

$$FR(t) = -\frac{D}{VC_0}exp(-\lambda t)\iint_S \frac{d\Psi}{dn}dA \quad (4)$$
$$= \exp(-\lambda t)FR_w(t)$$

where, FR_w means the fractional leach rate of stable isotopes in the container and n is a vector normal to the surface of the container.

The fractional amount of radionuclide accumulated in the environment, FE(t), can be expressed from the material balance on the surface of the matrix.

$$\frac{dFE(t)}{dt} = FR(t) - \lambda FE(t) \quad (5)$$

by setting $\zeta = FE(t)exp(\lambda t)$, then equation (5) becomes

$$\frac{d\zeta}{dt} = -\frac{D}{VC_0}\iint_s \frac{d\Psi}{dn}dA$$

$$FE(t) = -\frac{D}{VC_0}\exp(-\lambda t)\int_0^t [\iint_s \frac{d\Psi}{dn}dA]dt$$

$$= \exp(-\lambda t)\int_0^t FR_w(t)dt \quad (6)$$
$$= (-\lambda t)FE_w(t) \quad (7)$$

Total amount of a radionuclide in the matrix at time t normalized by the initial amount of that nuclide, FM(t), was derived from the relationship between the amount of stable isotope in the environment and that in the matrix.

$$\iiint_V \Psi(r,t)dV = VC_0 - \int_0^t [-D\iint_s \frac{d\Psi}{dn}dA]dt \quad (8)$$

The radionuclide fraction within the matrix is given by

$$FM(t) = \frac{1}{VC_0}\iiint_V C(r,t)dV$$
$$= \frac{1}{VC_0}\exp(-\lambda t)\iiint_V \Psi(r,t)dV \quad (9)$$

From equation (8)

$$FM(t) = \frac{1}{VC_0}\exp(-\lambda t)[VC_0$$
$$- \int_0^t (-D\iint_s \frac{d\Psi}{dn}dA)dt] \quad (10)$$

$$= \exp(-\lambda t)FM_w(t) \quad (11)$$

As shown in the equations (4), (7), and (11), the three quantities can be simply described from the multiplication of $exp(-\lambda t)$ by the quantities of stable isotope.

Finite Slab Approximation

The proposed approximation method is based on the hypothesis that the mass transfer within a matrix is not dependent on matrix geometry but dependent on volume to surface ratio (V/S), and it is expressed as the equations obtained from a finite slab model with an equal V/S ratio.

$$l = V/S$$

where l is imaginary diffusion length of a matrix

The three quantities of a leachability are given by the following equations for stable isotopes:

$$FR_w = \frac{2D}{l^2}\sum_{m=1}^{\infty} \exp(-\tau\beta^2{}_m) \quad (12)$$

$$FE_w = 1 - 2\sum_{n=1}^{\infty} \frac{1}{\beta^2{}_m}\exp(-\tau\beta^2{}_m) \quad (13)$$

$$FM_w = 1 - FE_w \quad (14)$$

$$\beta_m = \frac{(2m-1)}{2}\pi$$

where τ is a dimensionless leaching time (Dt/l^2), and t is real time. The equations (12), (13), and (14) indicate that the leaching quantities can be expressed as function of τ. Figure 1 shows the

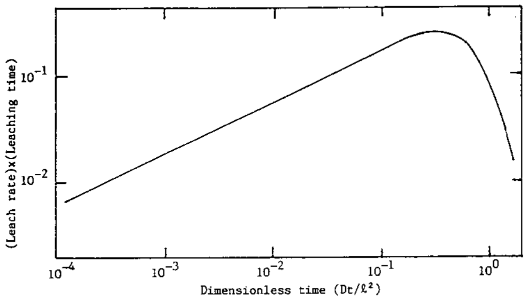

Figure 1. The Multiplication of the Fractional Leach Rate with Total Leaching Time Plotted Against Dimensionless Time, τ.

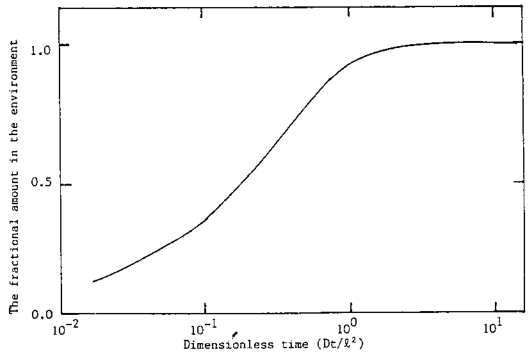

Figure 2. The Total Leaching Fraction in the Environment Plotted Against Dimensionless Time, τ

multiplication of the fractional leach rate with total leaching time plotted against the dimensionless time τ. The total leaching fraction released to the environment was plotted in Figure 2. These figures can be easily used to describe the leaching quantities if the dimension less time and the decay constant of a radionuclide are known.

Comparative Results

1. Finite Cylinder

Imaginary diffusion length of finite cylinder is given by

$$l = \frac{L/a}{1+2L/a} a$$

and dimensionless leach time is expressed as

$$\tau = \frac{Dt}{a^2} \left[\frac{1+2L/a}{L/a} \right]^2$$

where a is the radius of the cylinder and L is the cylinder's half geight.

Table 1 shows that the cumulative leaching fractions calculated by finite slab approximation were compared with those by Nestor[8]. According to the table, this model shows a good agreement with the actual leaching calculation. In the case of $L/a < 5.0$, the largest deviation from the cumulative leaching fractions obtained by Nestor[8] is less than 25 percent when the leach fraction is about 65 percent.

2. Sphere

The leaching quantities for spherical form of waste matrix were obtained by using the equation

$$FR_w = \frac{6D}{R^2} \sum_{n=1}^{\infty} \exp(-Dt\beta_n^2) \quad (15)$$

$$FE_w = 1 - \frac{6}{R^2} \sum_{n=1}^{\infty} \frac{1}{\beta_n^2} \exp(-Dt\beta_n^2) \quad (16)$$

where

$$\beta_n = \frac{n\pi}{R}$$

Imaginary diffusion length of sphere is

$$l = R/3$$

where R is the radius of sphere.

The comparison of the results from the equation (16) with those by finite slab approximation method was plotted against dimensionless time (Dt/l^2) in Figure 3. It shows that the calculations by the suggested approximation method overstimates 20 percent more with respect to the actual value calculated when leach fraction is about 55 percent, and this seems to be the result of difference of the form of the waste matrix container.

Table 1. The Comparison of the Calculations by Finite Slab Approximation Method With That by Nestor [8]

L/a	0.3		0.5		1.0		3.0		5.0	
method / Dt/a^2	a	b	a	b	a	b	a	b	a	b
0.0003	0.10141	0.1042	0.7636	0.0782	0.57570	0.0586	0.04505	0.0456	0.42550	0.0430
0.001	0.18077	0.1903	0.13671	0.1427	0.10353	0.1070	0.08142	0.0833	0.07679	0.0430
0.003	0.30175	0.3296	0.22928	0.2472	0.17494	0.1854	0.13866	0.1442	0.13142	0.1360
0.01	0.51055	0.5981	0.39252	0.4512	0.30400	0.3385	0.24498	0.2633	0.23312	0.2482
0.02	0.67096	0.8008	0.52243	0.6319	0.41050	0.4785	0.33588	0.3723	0.32095	0.3511
0.03	0.77179	0.9013	0.61002	0.7520	0.48490	0.5834	0.40148	0.4559	0.38480	0.4299
0.04	0.84016	0.9511	0.67601	0.8329	0.54284	0.6665	0.45401	0.5257	0.43625	0.4961
0.05	0.88724	0.9757	0.72830	0.8874	0.59036	0.7330	0.49820	0.5857	0.47977	0.5534
0.06	0.92012	0.9880	0.77087	0.9241	0.63055	0.7861	0.53647	0.6379	0.51765	0.6039
0.07	0.94324	0.9940	0.80606	0.9489	0.66524	0.8287	0.57025	0.6835	0.55125	0.6486
0.08	0.95957	0.9970	0.84543	0.9656	0.69561	0.8628	0.60048	0.7233	0.58146	0.6882
0.09	0.97116	0.9985	0.86010	0.9768	0.77249	0.8901	0.62781	0.7581	0.60888	0.7233
0.10	0.97940	0.9993	0.88091	0.9844	0.74648	0.9120	0.65571	0.78885	0.63395	0.7544
0.12	0.98946	0.9998	0.92347	0.9929	0.78745	0.9346	0.9654	0.8383	0.67834	0.8066
0.15	0.99613	1.0000	0.94617	0.9978	0.83567	0.9710	0.75065	0.8919	0.73365	0.8648
0.20	0.99927	1.0000	0.97547	0.9997	0.89196	0.9905	0.81879	0.9448	0.80413	0.9256
0.25	0.99986	1.0000	0.98880	1.0000	0.92865	0.9969	0.86766	0.9718	0.85540	0.9591
0.30	0.99997	1.0000	0.99488	1.0000	0.95280	0.9990	0.90311	0.9856	0.89306	0.9775
0.40	0.99999	1.0000	0.99893	1.0000	0.97933	0.9999	0.94785	0.9962	0.94134	0.9932
0.50	1.00000	1.0000	0.99978	1.0000	0.99094	1.0000	0.97183	0.9990	0.99775	0.9979

a. The calculations by Nestor
b. The calculations by this finite slab approximation method

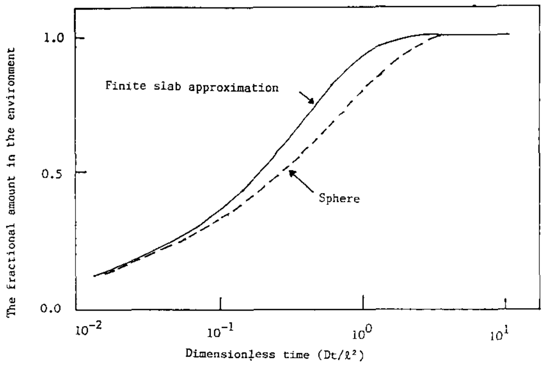

Figure 3. The Comparison of The Results From Equation (16) with Those from Finite Slab Approximation Plotted Against Dimensionless Time, τ.

Conclusion

The prediction of long-term leaching behavior from short-term test will experience a limitation due to physical and chemical properties of the environment and waste matrix during leaching time such as dispersion coefficient, porosity, and chemical reactions. Several experiments show that the leachability of radionuclide from waste matrix is subsequently decreased with time by other factors such as surface resistance, therefore the prediction from short-term test is considered to be conservative.

The comparisons were performed for cylindrical and spherical waste form with our suggested method. According to this comparison, the results from finite slab approximation are also conservative and more practical with respect to the rigorous results.

Finite slab approximation has 3 merits;

1) It represents the fractional amount in the environment for practical shapes of waste mat-

rix form as a function of dimensionless time (Dt/l^2) which is important factor in the three leaching quantities.

2) It provides an easy calculational application to the long-term prediction.

3) It can express the leachability by using simple table or graph as a function of dimensionless time (Dt/l^2).

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References

1. E. D. Hespe, Ed., Leach "Testing of Immobilized Radioactive waste solid, A Proposal for a Standard Method." Atomic Energy Review **9,** 195 (1971).
2. J. G. Moore, H. W. Godbee, A.H. Kibbey, and D. S. Joy, "Development of Cementitious Grouts for the Incorporation of Radioactive Wastes." Part I ORNL-4962, (1975).
3. H. W. Godbee and D. S. Joy. "Assessment of the Loss of Radioactive Isotopes from Waste Solids to the Environment." Part I: Background and Theory. ORNL-TM-4333, February (1974).
4. E. L. Compere and H. W. Godbee, "The Interrelation of Short-Term Leach Tests and Long-Term Leaching." American Nuclear society 1979 Winter Meeting. CONF-791103-98.
5. Cheng Y. Hung, "Prediction of Long-term Leachability of a Solidified Radioactive Waste from a Short-Term Leachability Test by a Similitude Law for Leaching Systems." Nuclear and Chemical Waste Management **3,** 235 (1982).
6. R. W. Crawford, F. P. Glasser, A. A. Rahman, and M. J. Angus, "Diffusion Mechanism and Factors Affecting Leaching of Cesium-134 from Cement-Based Waste Matrices." Radioactive Waste management and The Nuclear Fuel Cycle **6,** 177 (1985).
7. H. Matsuzuru, N. Moriyama, Y. Wadachi, and A. Ito, Leaching Behavior of Cesium-137 in Cement-Based Composites. Health Physics **32,** 529-534 (1977).
8. C. W. Nestor, "Diffusion from Solid Cylinders." ORNL/CSD/TM-84 (1980).
9. G. L. Richardson, "The Phantom Dissolution Leach Model." Nuclear and Chemical Waste Management **2,** 237 (1981).
10. M. J. Bell, "An Analysis of the Diffusion of Radioactivity from Encapsulated Wastes." ORNL-TM-3232 (1971).
11. H. S. Carslaw and J. C. Jaeger, "Conduction of Heat in Solids." 2nd Ed. Oxford University, Oxford (1959).
12. J. Crank, "The Mathematics of Diffusion." 2nd Ed. Clarendon Press, Oxford (1975).
13. M. Necati Ozisic, "Heat Conduction." John Wiley and Sons, New York (1980).