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Experimental Study of Leaching Phenomena of Cs-137 From a Cement Matrix Generated at PWR Plant

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=Abstract=

Experimental study for the leaching behavior of Cs-137 was carried out using the simulated evaporator bottom product of PWR plant. The method of leach test proposed by the IAEA was partially modified using ANS method. The effect of various factors, i.e., sampling method, curing temperature, curing time, leachant temperature, vermiculite addition and volume-to-surface ratio, was considered in this experiment.

Diffusion model in semi-infinite slab was in a good agreement with the data obtained from 4-weeks cured specimens. The effective diffusion coefficient of the specimens which were cured at the temperature of 24°C for 4 weeks was found to be $1.20 \sim 1.47 \times 10^{-11} cm^2/sec$. With the experimentally obtained diffusion coefficient($1.47 \times 10^{-11} cm^2/sec$), long-term prediction for the leaching of Cs-137 was carried out using finite-slab approximation.

The estimated fraction of Cs-137 which remains in the environment is found to be less than 0.25 percent of initial amount after 100 years. About 25 years after the beginning of leaching, its fractional amount in the environment reachs the maximum value, 0.66 percent of initial amount.

1. INTRODUCTION

An increasing usage of nuclear energy and radioisotopes generate a variety of radioactive wastes which have diverse physical and chemical characteristics and the wide range of activity level.

The greater part of the radioactive wastes which have been accumulated in Korea is found to be low and intermediate level. More than 95%[1] of these wastes has been generated at nuclear power plant and about 65%[2] of the power plant wastes is the final form of evaporator bottom product.

Solidification processes of the low and intermediate level wastes using several binding agents, i.e., cement, bitumen, polyester polymer[3, 4, 5] has been studied and being employed to convert the waste into less mobile forms. Among these processes, cement solidification has been widely studied and routinely used at nuclear research and power production sites.

In Korea, the cementation process has been used at nuclear power plants from the beginning of commercial generation of nuclear power and the solidified products have been stored at the drum storage area on site of power plant. Generally, the site has a capacity of about 10 years storage[2] and the storage of a plant constructed during 1970's is now almost saturated to its full capacity. Therefore, the final disposal methods to solve this problem have been considered and focused as a national interest.

Under disposal condition, one of the major ways by which the radionuclides may be released from the solidified matrix is a leaching process of ground water during the period of disposal. Although the understanding of the leaching mechanism has been recognized to be important, the leaching mechanism is not completely resolved because of an inhomogeneity of matrix and the complexity of its environment.

The leachability of the radionuclide from a waste matrix has been studied using various methods. To compare the various experimental results obtained using different processes and different materials and to determine the long-term leaching behavior during the geologic disposal, the IAEA has proposed a standard method[6] which was known as a dynamic test performed at ambient temperature. But this method was not accepted as a standard test to be widely used because of several reasons[7, 8]. Subsequent efforts to standardize the leaching test have been followed based on the IAEA proposal. Here, the IAEA and part of the ANS modified test method were followed. Also, models including the various factors to affect the leachability of radionuclide were investigated. The long-term behavior of Cs-137 in the real waste container was predicted using the finite-slab approximation and the appropriate diffusion coefficient of Cs-137.

Ⅱ. THEORY

Cement matrix is considered as an inhomogeneous and porous material. Consequently, the hydration product of cement is more susceptable to the movement of water because of the relatively high diffusive and porous network structure compared to the nonporous body.

For the simplification, the homogeneous porous body and the linear equilibrium relationship between nuclide concentrations in liquid phase and those in i-th solidification agent of solid phase, $G_i = K_i\ C$, are considered, where G_i is the nuclide concentration in the i-th solidification agent, K_i is the equilibrium constant of i-th solidification agent, and C is the nuclide concentration in the liquid phase.

Assuming the diffusion coefficient of the nuclide to be constant, diffusion equation for the nuclide in the homogeneous porous body which is composed of certain binding agents is given as follows;

$$\frac{dC}{dt} = D_{\text{eff}}\nabla^2 C - \lambda C$$

with initial and boundary conditions as;

I.C. $C(\vec{r}, 0) = 0$

B.C. $C(\vec{R}, t) = 0$

where,

$$D_{\text{eff}} = \frac{D_{\text{pore}}}{1 + \frac{1-\varepsilon}{\varepsilon}\rho_s \sum_{i=1}^{n} \eta_i K_i}$$

$$D_{\text{pore}} = \frac{D_{\text{liq}}}{\tau}$$

D_{liq}: the diffusion coefficient of a nuclide in the liquid body

τ: tortuosity

ε: the total porosity of matrix

ρ_s: solid density

η_i: mass fraction of i-th solidification agent

λ: decay constant
∇^2: the Laplacian operator
$D_{\rm pore}$: the diffusion coefficient in the pore network
$D_{\rm eff}$: the effective diffusion coefficient
C: the nuclide concentration in the liquid phase agent
$\vec{r}$: position vector
$\vec{R}$: surface vector

Regardless of the geometry of the container, three quantities, i.e., fractional leach rate of radionuclide from the waste matrix [FQ(t)], the fractional amount in the matrix [FR(t)], and the fractional amount in the environment [FE(t], can be obtained simply from the multiplication of $\exp(-\lambda t)$ to the results obtained for stable isotope[9, 10]. In other words, the solution of the mass transport equation including the radioactive decay term is generally expressed as the multiplication of $\exp(-\lambda t)$ term to the solution for the corresponding stable isotope nuclide. (Here, the environment means the just outside the matrix).

$$FQ(t)=-\frac{D_{\rm eff}}{VC_0}e^{-\lambda t}\iint_s\frac{d\phi}{dn}dA$$
$$=e^{-\lambda t}FQ_w(t) \tag{2}$$

$$FE(t)=-\frac{D_{\rm eff}}{VC_0}e^{-\lambda t}\int_0^t\left[\iint_s\frac{d\phi}{dn}dA\right]dt$$
$$=e^{-\lambda t}FE_w(t) \tag{3}$$

$$FR(t)=e^{-\lambda t}\left\{1-\frac{1}{VC_0}\int_0^t\left[-D_{\rm eff}\iint_s\frac{d\phi}{dn}\quad dA\right]\right.$$
$$\left.dt\right\}=e^{-\lambda t}FR_w(t) \tag{4}$$

where $\phi(r,t)$ is the solution to the diffusion equation for stable isotope subject to the same boundary conditions but with no radioactive decay and subscript w means the leaching characteristics for stable isotope.

Dejonghe, et al[12]. employed the solution [13] of the transport equation for a semiinfinite slab to explain the amount of material leaving per unit surface from the asphalt-sludge product. Their results are summarized as;

$$FQ_w(t)=\frac{S}{V}\left(\frac{D_{\rm eff}}{\pi t}\right)^{1/2} \tag{5}$$

$$FE_w(t)=2\frac{V}{S}\left(\frac{D_{\rm eff}t}{\pi}\right)^{1/2} \tag{6}$$

The equations(5) and(6) have been adopted for the analysis of short-term leach tests and the prediction of long-term leaching for relatively short-lived radionuclides.

Doh and Lee[11] developed an approximationmethod with finite-slab diffusion model which can be used in the cases where actual geometry of waste matrix does not permit an analytic solution or where the solution is an exact one of the diffusion model requires tedious numerical summations of infinite series.

$$FQ_w(t)=2\frac{\xi}{t}\sum_{m=1}^{\infty}\exp(-\xi\beta_m^2) \tag{7}$$

$$FE_w(t)=2\sum_{m=1}^{\infty}\frac{1}{\beta_m^2}\left[1-\exp(-\xi\beta_m^2)\right] \tag{8}$$

where,

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

$$\xi=\frac{D_{\rm eff}t}{l^2}$$

$$l=\frac{V}{S}\ \text{(volume to surface ratio)}$$

Eqs. (7) and (8) were utilized later for the long-term prediction of Cs-137 leaching from the matrix

Ⅲ. EXPERIMENT

The specimens used in this experiment were made by combining the desired volume of waste solution with a predetermined weight of Type I Portland cement or cement-vermiculite mixture.

The liquid waste simulating the evaporator bottom concentrate generated at PWR plant

Table 1. Description of Speciments Used for Leaching Study

Sample Name	Dimension	Curing Time (weeks)	Curing Temp. (°C)	Initial Actiuity (μCi)	Nominal Sampling Freq.	Leachant Temp. (°C)
AA—1	a	4	5	372.8	d	24
AA—2	a	4	24	372.8	d	24
AA—3	a	4	24	372.8	d	24
AA—4	a	4	24	372.8	d	24
AA—5	a	4	60	372.8	d	24
AB—1	b	4	24	249.5	d	24
AB—2	b	4	24	249.5	e	24
BB—1	a	12	24	1744.8	d	24
BB—2	a	4	24	1744.8	e	24
BC	c	4	24	167.5	e	24
A—1	a	1	21	20.79	e	12(f)
A—2	a	1	21	20.79	e	23(f)
B	a	1	21	20.89	e	52(f)
C	a	1	21	17.77	e	23
D	a	1	21	17.68	e	23
E	a	1	21	18.24	e	23
F	a	1	23	17.52	e	23

a-height: 5 cm, diameter: 5 cm, only one face exposed
b-height: 7.48 cm‘ diameter: 3.35 cm, only one face exposed
c-height: 3 cm, diameter: 2 cm, all the surface exposed
d-in accordance with the IAEA recommended procedure
e-norminal sampling frequency of once a day
f-retaining the leachant unfilled for 1 week after 18 days from the start of test

was prepared by dissolving boric acid(reagent grade) into deionized water to a content about 12 weight percent. Prior to the addition of the Cs-137(CsCl type), an appropriate quantity of NaOH(reagent grade) was added to the solution to give a final pH of about 12.

Preliminary trials showed that the ratio of waste to cement(W/C), 0.362 [cc waste/g cement], had showed a good experimental conditions and no phase separation in the paste. All but three specimens for the study of vermiculite effect were prepared at this W/C ratio.

After sufficient mixing, the resulting pastes were poured into specimen containers and cured for a predetermined number of days and temperature in a closed vessel where the relative humidity was maintained at 100%.

The cured specimens in the container were placed inside a transparent PMMA(polymethyl metaacrylate) cylinder and the leachant was replaced periodically with demineralized water. The results of 17 leach tests are briefly described in Table 1.

Ⅵ. RESULTS AND DISCUSSION

Seventeen tests were carried out to evaluate the leaching phenomena of Cs-137 from the cement matrices with deionized water.

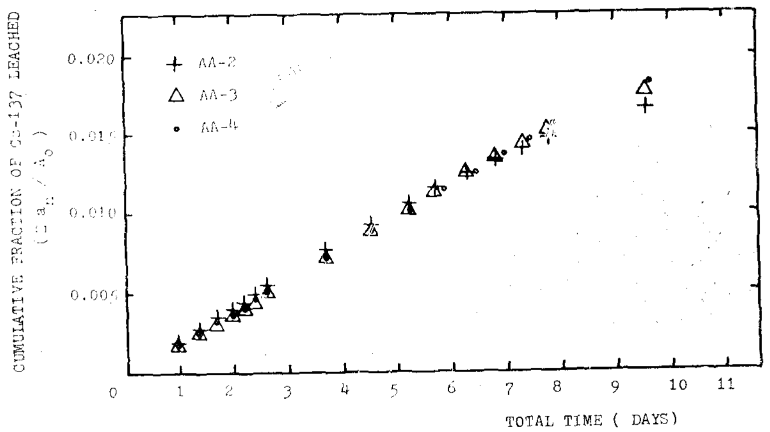

Fig. 1. ($\Sigma a_n/A_0$) Plotted against Square Root of Time for the Specimens AA—2, AA—3 and AA—4. (The specimens were cured for 4 weeks under humid condition and had a V/S of 5 cm).

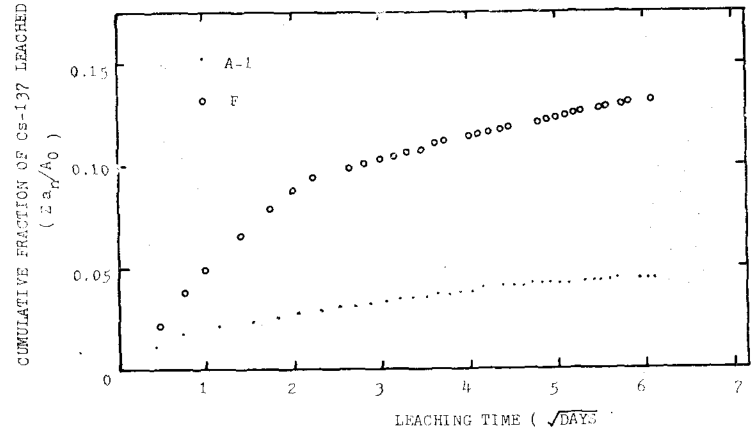

Fig. 2. ($\Sigma a_n/A_0$) Plotted against Square Root of Time for the Specimens A—1 and F. (The specimens were cured for 1 week under humid condition, and had a V/S of 5 cm.)

The curing time under 100% relative humidity prior to the leaching of the specimens ranged from 1 to 12 weeks. During the leaching test, the time interval between replacement of the leachant with deionized water varied from 5 hours to 1 month. The results are plotted in Fig. 1 through Fig. 9.

1. Replication Test

Fig. 1 was obtained from the tests of samples AA-2, AA-3 and AA-4, which were made from the same bath and cured for 4 weeks, in accordance with the IAEA method. The results from the sample number A-1 and F which were made from different batches and cured for 1 week, with changing the leachant normally once a day were plotted in Fig. 2. Fig. 3 shows the results of AA-4 and BB-2 samples(from different batches, cured for 4 weeks).

During the leaching test, the cumulative leaching fractions in Fig's. 1 and 3 were limited within 13% differences after 90 days, but the results given in Fig. 2 show that $\Sigma a_n/A_0$ of sample number F is about 3 times greater than that of A-1. This emphasizes

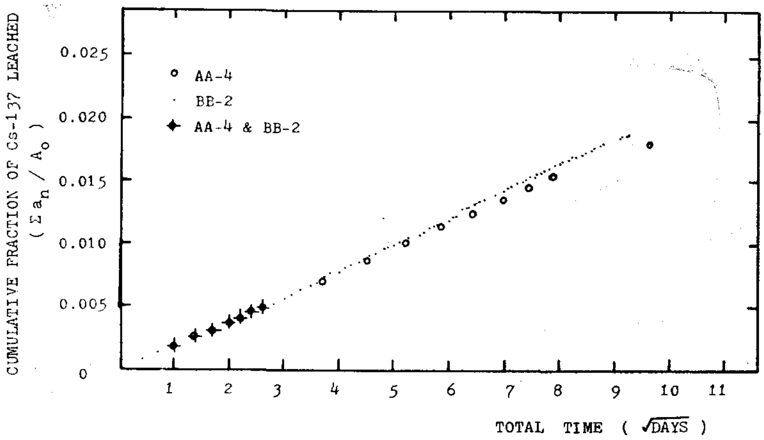

Fig. 3. ($\Sigma a_n/A_0$) Plotted against Square Root of Time for the Specimens AA—4 and BB—2. (The specimens were cured for 4 weeks and from different batches.)

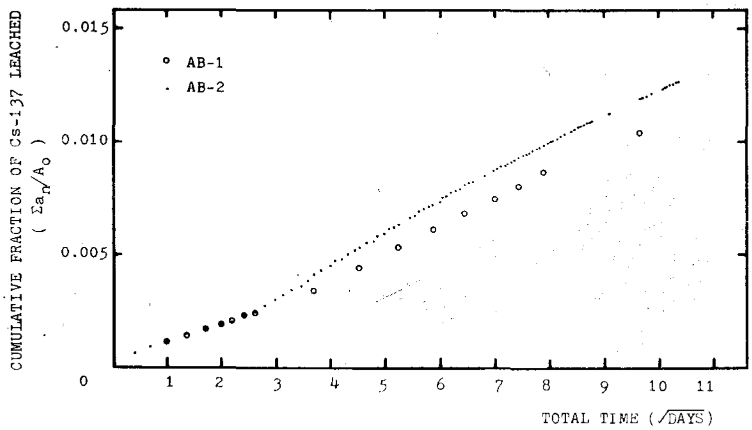

Fig. 4. ($\Sigma a_n/A_0$) Plotted against Square Root of Time for the Specimens AB—1 and AB—2. (The specimens were cured for 4 weeks under humid condition, and had a V/S of 7.48 cm.)

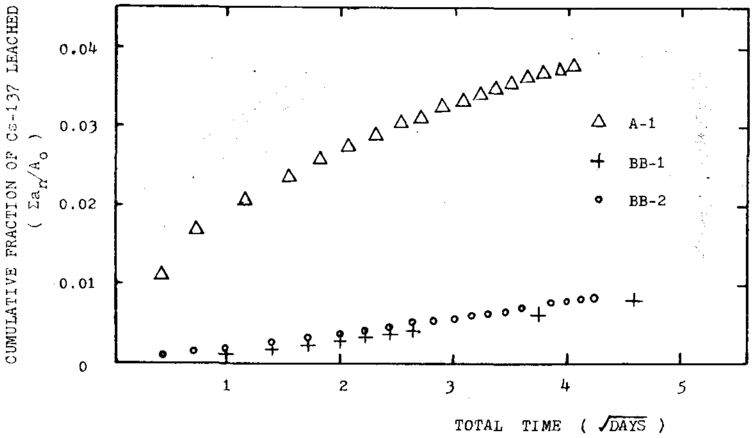

Fig. 5. ($\Sigma a_n/A_0$) Plotted against Square Root of Thime for the Specimens A~1, BB~1 and BB~2. (The specimens were cured for 1,4 and 12 weeks, respectively, before leaching commenced.)

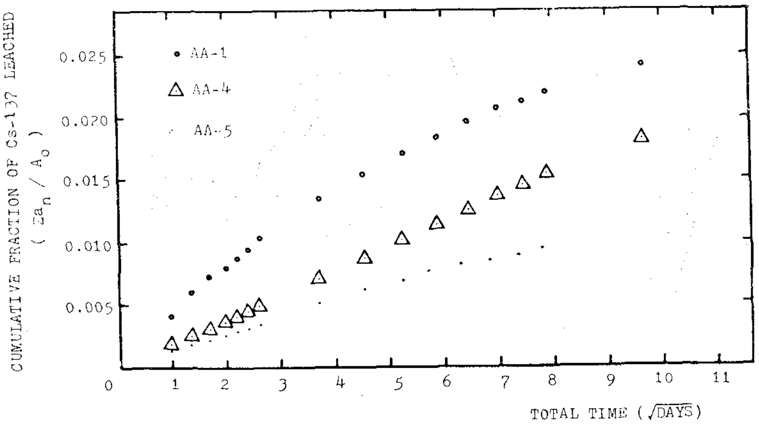

Fig. 6. ($\Sigma a_n/A_0$) Plotted against Square Root of Time for the Specimens AA—1, AA—4 and AA—5. (The specimens were cured for 4 weeks under humid condition, and were setted at 5, 24 and 60°C, respectively.)

the regeneration difficulty frequently encountered in replication test with heterogeneous material.

The specimen cured for more than 4 weeks has been adopted because test method must be based on the replicability and reality.

2. Effect of Sampling Frequency

In the experiment with the different sampling frequencies, it became apparent that the longer sampling periods(the IAEA method) had significant effect on the leach results. Fig. 4 shows the data obtained from AB-1 and AB-2(from same batch of paste).

On Fig's 3 and 4, the leachant for the case of AA-4 and AB-1 was changed by following the IAEA method, but that for AB-2 and BB-2 was changed norminally once a day.

It can be seen from the Fig's 3 and 4 that the slope of leach curve changes due to sampling method after about 7 days from the beginning of leaching. Normally, it is well-known that the mass transport flux between the two different phase(here, solid and liquid) is expressed as the product of mass transport coefficient and the concentration difference between the two phase. Here, the mass transport coefficient is strongly dependent on the solid surface resistance, and the sampling method significantly influence the concentration difference between the two phase. Therefore, these results suggest that the leach rate had been influenced by the concentration difference of the nuclide between leachant body and the surface resistance of waste matrix. Since the ground water in burial site is nearly static, the surface resistance will play an important role on the leachability of radionuclide from waste maxtrix.

3. Effect of Curing Time

To examine the effect of curing period, the leach tests were carried out using the specimens cured for 1 week(A-1), 4 weeks (BB-2) and 12 weeks(BB-1). Fig. 5 shows the results obtained from these specimens. This leads to the conclusion that longer curing times cause the decrease of the amount leached from the matrix, and curing time of longer than 4 weeks has little effect on the

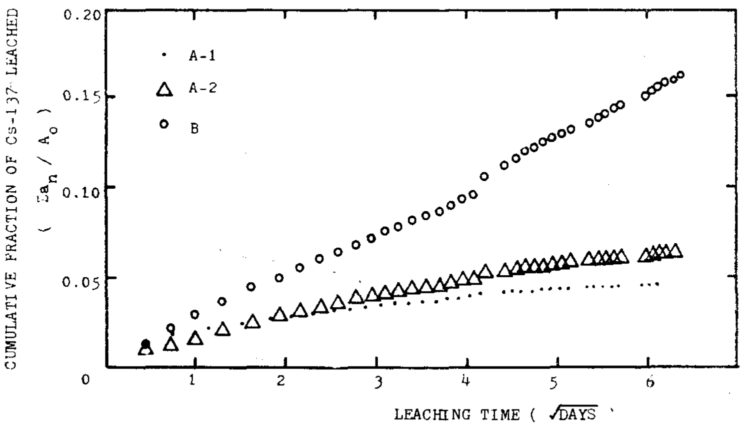

Fig. 7. ($\Sigma a_n/A_0$) Plotted against Square Root of Time for the Specimens A—1, A—2 and B.(The specimens were cured for 1 week under humid condition, and those leachant temperature were 12, 23 and 52°C, respectively.)

leach rate. This is due to the fact that the rate of hydration decreases exponentially with time at ambient temperature[14]. The total porosity decreases and the hydraulic radii of the entire pore systems decrease with processing hydration[15]. Hence, the decrease of porosity and the longer tortous path result in reducing the leachability of nuclide.

4. Effect of Curing Temperature

To test the effect of curing temperature, the leach tests were conducted for the 3 specimens which were cured at 5°C(AA-1), 24°C(AA-4) and 60°C(AA-5) for 4 weeks. After curing at different temperatures, leach tests were carried out at the same temperature of 24°C.

Fig. 6 shows that the higher curing temperature leads to the reduced leachability. Because more hydration products were made at elevated curing temperature than at lowes temperature[14], the corresponding leachability decreases.

5. Effect of Leachant Temperature

To find out the effect of leachant temperature on the leachability, leaching tests were done at the several temperatures. Fig. 7 shows that leach rate increases by increasing the leachant temperature(A-1 at 12°C, A-2 at 23°C and B at 52°C).

Diffusion coefficients of electrolytes can be predicted very accurately at infinite dilution using the equation[16].

$$D_{AB}=\frac{2RT}{(1/\lambda_+^0+1/\lambda_-^0)\bar{F}}$$

where,

D; diffusion coefficient based on the concentration of A in B

R; the gas constant

T; absolute temperature

$\bar{F}$; Faraday's constant

λ_+^0, λ_-^0; the limiting ionic conductances

Due to such a temperature dependency of the diffusion coefficient, the leachability of nuclide increases with increasing temperature.

6. Effect of Dimension

The relationship between the cumulative fraction of nuclide leached and V/S(volume to exposed surface) ratio has been examined by using the specimens with various V/S

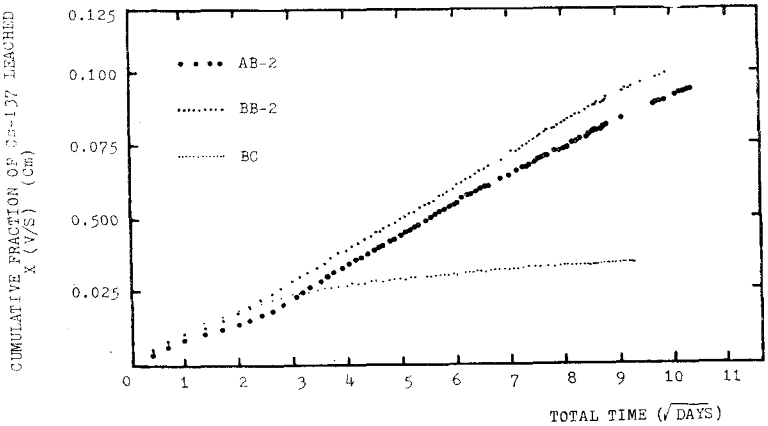

Fig. 8. $(\Sigma a_n/A_0)\times(V/S)$ Plotted against Square Root of Time for the Specimens AB—2, BB—2 and BC. (The specimens were cured for 4 weeks under humid condition, and a V/S of 7.48, 5 and 0.375 cm, respectively)

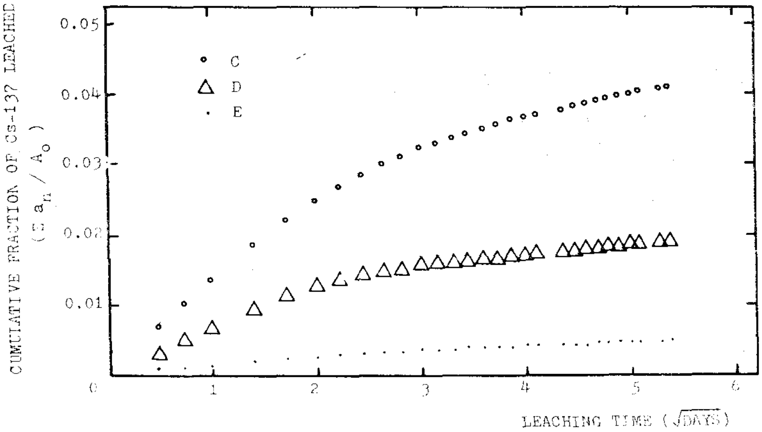

Fig. 9. $(\Sigma a_n/A_0)$ Plotted against Square Root of Time for the the Specimens C, D and E. (The specimens were cured for 1 week under humid condition, and had a vermiculite content of 3.2, 6.2 and 20 percent, respectively.)

ratios, BB-2(V/S: 5 cm) AB-2(V/S: 7.48 cm) and BC(V/S: 0.375 cm). On Fig. 8, the specimens(AB-2 and BB-2) which have only one exposed surface, appear to have similar leachability, but BC with all the face exposed show entirely different property from AB-2 or BB-2. Therefore, it is not appropriate to deduce a general law for various V/S ratiosas described by Matszuru et al.[17].

The results of AB-2 and BB-2 reveal that, at the early stage of leaching, the leach rate decreases with respect to the value of theoretical trends and then, after aoout 7 days, the data fit well with the square root of time. This can be explained by analyzing the characteristics of pore in the matrix. Because the volume of the hydration product formed is less than the sum of the volumes of cement and water which react to form it, the hydration product does not fill completely the volume avaiable for it[15].

When the cement matrix is in contact with water, more water will be drawn into the paste and radionuclides in relatively dry

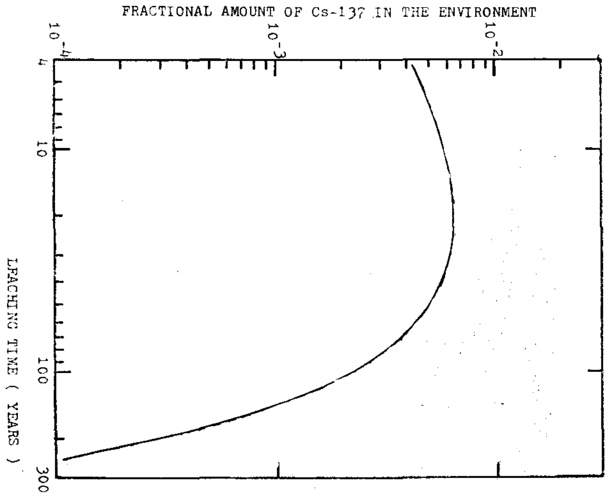

Fig. 10. Fractional Amount of Cs-137 in the Environment Plotted against Total Leaching Time(D_{eff}= $1.47\times10^{-11}cm^2/sec$, 55 Gal drum)

pores will be less mobile than those in water-filled pores. Therefore, water intrusion and diffusion to the pores are the dominant factors which decrease the leach rate at early stage of leaching from matrix cured for 4 weeks or longer.

7. Effect of Vermiculite

Various additives have been used to the cementification systems to improve the sorption and immobilization of radionuclides.

The effect on the leachability due to the variation of the fraction of vermiculite in the matrix was studied by using the three specimens C, D and E. Fig. 9 shows the results of the leach tests on three cement/ Vermiculite mixture containing 3.2(C), 6.2 (D) and 20 weight percent(E) of vermiculite.

Due to the ionic selectivity of vermiculite on electrolyte, the leach rate is decreased for cement/vermiculite matrix with increasing vermiculite content. In other words, increasing the vermiculite interfere the leachability of cesium, but the workability of unhardened paste decreases concurrently. This is because vermiculite absorb water extensively, which results in a paste with insufficient water.

V. EFFECTIVE DIFFUSION COEFFICIENT AND LONG-TERM LEACHING

The prime objective in the analysis of the leach data is to determine the parameters which are able to describe the leaching characteristics of radionuclide from waste matrix for the long time.

The results for 17 specimens used in this paper have a property that the cumulative leaching fractions are directly proportional to the square root of time. Among them, the results from 1 week-cured specimens and all surface exposed specimen present a enormous discrepancy to a theoretical trend because of the shape effect of the specimen or unstable state of matrix. In these specimens, the leach data can be analyzed by two parameter method[17]. In the case of analysis of the result from one face exposed specimen, expression by two parameter method is to be meaningless because the aim of such a test is in obtaining one constant. As shown in Figs 3 and

4, the leachability of a matrix cured for 4 weeks, can be expressed as 3 stages.

1 st stage: decrease of leach rate by water intrusion into pores and diffusion through pores(up to about 7 days)

2 nd stage: normal diffusion(from 7 to about 60 days)

3 rd stage: decrease of leach rate by surface resistance(thereafter)

Therefore, the second stage can be considered as critical stage to obtain effective diffusion coefficient. The data from the specimens AA-4, BB-2, AB-1 and AB-2 were fitted to equation(6) to give D_{eff}'s of 1.22×10^{-11}, 1.47×10^{-11}, 1.2×10^{-11} and 1.47×10^{-11} cm^2/sec, respectively. As the specimens AB-1 and AB-2 are made from the same batch of paste, sampling period is the only factor which can explain the difference on diffusion coefficient.

Equation(6) appears to have provide a reasonably good explanation of the leach behavior of Cs-137 from 4 weeks cured specimens. Test that is carried out for short period (about 30 40 days) is sufficient to obtain the effective diffusion coefficient. With the obtained diffusion coefficient(1.47×10^{-11}cm^2/sec), long-term leaching of Cs-137 was predicted by finite slab approximbtion [Equation (3) and (8)]. Fig. 10 represents the fractional amount of Cs-137 which remains in the environment outside of the container. The estimated fraction of Cs-137 in the environment is found to be about 0.25 percent of initial amount after 100 years. After about 25 years, the value reach to maximum value of 0.66 percent.

VI. CONCLUSION

The following conclusions have been derived from this study.

1. From the replication tests, it is understood that the specimen used in leach test must be cured longer than 4 weeks and that the cumulative fraction leached in the environment may differ by as much as a factor of 1.3.
2. With respect to the test of curing time, longer curing period decreases the leach rate but curing for longer than 4 weeks has no significant effect on the amonut of Cs-137 leached from cement matrix.
3. The leach rate was lowered by retaining high curing temperature, but flash setting is turned out to be important in such conditions.
4. The leachability of cesium increases with increasing temperature due to temperature dependency of diffusion coefficient in water body.
5. The leachability of Cs-137 was lowered by the addition of vermiculite which has a good selectivity to cesium compared to other nuclides.
6. The solution of semi-infinite slab model gives good agreement with the data obtained in tests using 4 weeks cured specimens.
7. The effective diffusion coefficient obtained using the IAEA test method is lower than that obtained from the experiment changing leachant once a day. Test period about 30~40 days is sufficient to obtain D_{eff}.
8. The estimated fraction of Cs-137 leached from a cement matrix is less than about 0.25% of initial amount after 100 years. After about 25 years from the beginning of leaching, the fractional amount in the environment reach to maximum value(about 0.66% of initial amount).

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REFERENCES

1) 저준위 방사성 폐기물 처분방안(Ⅰ), KRC-84N-S19 한국전력공사 기술연구원(1985).

2) Final Safety Analysis Report of Korea Nuclear Unit 7 & 8, Vol. 10.

3) A.A. Moghissi, H.W. Godbee, M.S. Ozker and M.W. Carter Ed., *Nuclear Power Waste Technology*, ASME, New York(1978).

4) W.R. Gilmore, *Radioactive Waste Disposal*, Noyes Data Corporation, Park Ridge(1977).

5) International Atomic Energy Agency, *Conditioning of Low-and Intermediate-Level Radioactive Wastes*, Tech. Rep. Series No. 222, IAEA, Vienna(1983).

6) E.D. Hespe, Ed., "Leach Testing of Immobilized Radioative Waste Solids, A Proposal for a Standard Method," *Atomic Energy Review* Vol. 9(1971).

7) A.E. Ogard and E.A. Bryant, "The Misused and Misleading IAEA Leach Test," *Nuclear and Chemical Waste Management* Vol. 3(1982).

8) J.G. Moore, *et al.*, *Development of Cementitious Grouts for the Incorporation of Radioactive Wastes. Part I: Leach Studies*, Oak Ridge National Laboratory Report ORNL-4962(1975).

9) M.J. Bell, *An Analysis of the Diffusion of Radioactivity from Encapsulated Wastes*, Oak Ridge National Laboratory Report ORNL-TM-3232(1971).

10) J.Y. Doh "Leaching Study of Cs-137 from a Cement Matrix Generated at Nuclear Power Plants," Master Thesis, Department of Nuclear Engineering, KAIST(1986).

11) J.Y. Doh and K.J. Lee, "Long Term Prediction of Radionuclide Leaching from Waste Matrix by Finite Slab Approximation Method," Paper Presented at Int. Topical Meeting on Waste Management, Decontamination and Decommissioning, Sep. 14-18, Niagara Falls, New York(1986).

12) H.W. Godbee and D.S. Joy, *Assessmet of the Loss of Radioactive Isotopes from Waste Solids to the Environment. Part I: Background and Theory*, Oak Ridge National Laboratory Report ORNL-TM-4333(1974).

13) J. Crank, *The Mathematics of Diffusion*, 2nd Ed., Clarendon Press, Oxford(1975).

14) H.F.W. Taylor, Ed., *The Chemistry of Cement*, Academic Press, New York(1964).

15) Ivan Odler *et al.*, "Pore Structure Analysis by Water Vapor Adsorption," *Journal of Colloid and Interface Science*, **38**(1), (1972).

16) J.R. Welty, C.E. Wicks and R.E. Wilson, *Fundamentals of Momentum, Heat and Mass Transfer*, 3rd Ed., John Wiley & Sons, New York(1984).

17) H. Matsuzuru and A. Ito, "Effect of Dimension of Specimen on Amounts of Cesium-137, Strontium-90 and Cobalt-60 Leached from Matrix of Hardened Cement Grout," *J. Nucl. Sci. Technol.*, **15**(1978).

가압 경수로에서 생성된 시멘트 고화체로부터 Cs-137의 용출 현상의 실험적 연구

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=요약=

가압 경수로형 원자로에서 발생하는 증발기 저부 폐액에서의 Cs-137의 용출에 대한 실험을 수행하였다. IAEA에 의해 제안된 용출실험 방법에 근거를 두고, ANS 방법의 일부를 채용하였다.

용출에 영향을 미치는 여러 인자들로서, 시료채취방법, 양생온도, 양생기간, 용출액 온도, Vermiculite첨가와 체적 대 표면적비 등이 고려되었다.

준 무한 격판(Semi-infinite Slab)에 대한 확산 모델은 4주간 경화된 시료의 실험치와 좋은 일치를 보이고 있다. 4주간 25°C에서 양생된 시료의 표면적 확산 계수는 $1.20 \sim 1.47 \times 10^{-11} cm^2/sec$가 됨을 확인했으며, 이 계수에 의해 Cs-137의 장기 용출을 예측을 유한 격판 근사(Finite-slab Approximation) 방법을 이용하여 수행하였다.

또한, 계산 결과로부터 Cs-137은 용출 개시후 약 25년이 되면 초기량의 0.66%인 최대치가 되며 100년 후에는 약 0.25%가 잔류한다.