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Modeling of nuclide releases from perforated radioactive paraffin waste containers

Ju Youl Kim [a,*], Chang Lak Kim [b], Chang Hyun Chung [a]

[a] *Department of Nuclear Engineering, Seoul National University, San 56-1 Shinrim-dong, Gwanak-gu, Seoul 151-742, South Korea*
[b] *Nuclear Environment Technology Institute, Korea Hydro & Nuclear Power Co., Ltd., 150 Dukjin-dong, Yuseung-gu, Daejeon 305-600, South Korea*

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Abstract

An effect of a pinhole (perforation or pit penetration) that can be formed on the surface of a waste container on the nuclide release is studied. The more realistic pinhole release model is developed from the diffusion-controlled dissolution reaction in consideration of the receding hemispherical concentration front around the pinhole and the relation of the nuclide concentration with its solubility. Except for an initial period, the pinhole release rates rapidly approach to the zero-order kinetics. The steady-state release rates are directly proportional to the diffusion coefficient of the nuclide, the nuclide concentration or the solubility depending on the sub- or super-saturated loading, and the radius of the pinhole not to its area. The predicted results of this theoretical model agree well with experimental results using the leaching test of the paraffin waste form. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

The current design concepts for the potential geological disposal of radioactive wastes usually require the preparation of containers that are used for handling, transport, storage, and disposal of radioactive wastes [1–3]. A container is a permanent part of the waste package which may vary for the different steps in waste management. Radioactive waste packages will play an important role as the ultimate engineered barrier to prevent the releases of radionuclides from various waste forms in the first stage of disposal. Waste containers are likely to fail by localized corrosion, cracks, degradation, or fabrication defects [4–7]. After a failure of a waste container by these processes, the waste form, either low-level or high-level waste, inside the package is assumed to be immediately exposed to the near-field environment, and then radionuclides may be released and transported through the perforation (pinhole or pit penetration) to the edge of the engineered barrier system and biosphere.

Several studies have been performed on the radionuclide release from perforated waste containers [8–14]. But it was not reported that the analytical nuclide release models developed until now had been validated by any laboratory experiment. Besides, the earlier models assumed an idealized transport geometry, e.g., cylindrical or semi-infinite, and the concentration distribution of the nuclide within the waste form was not considered. The purpose of this study is to develop a realistic release model considering the receding hemispherical concentration distribution around the pinhole and to compare the theoretical model with experimental results.

In this paper, a paraffin waste package including boric acid and cobalt is chosen as specimen in order to investigate an effect of a pinhole that can be formed on

* Corresponding author. Tel.: +82-2 880 8331; fax: +82-2 889 2688.
E-mail address: hispeace4u@hanmail.net (J.Y. Kim).

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the surface of the container on nuclide leaching from the waste form. Paraffin waste forms have recently been generated from Korean nuclear power plants by simply mixing the low-level liquid borate concentrate waste with paraffin wax since 1995 [15]. Three cases of the specimen are prepared according to the size of the pinhole. The material properties of the waste containers and mechanistic modeling of the formation of the pinhole are not considered.

2. Theoretical background

2.1. Literature review

Rae [8] was the first to consider the steady-state release rate of the radionuclide through a circular hole in waste container by diffusing through the internal matrix and surrounding permeable medium. The diffusive mass transfer rate through the hole was given by

$$\dot{m}_{ss} = 4\frac{D_i D_o}{D_i + D_o} C_o r_o, \tag{1}$$

where D_i and D_o are the effective diffusion coefficients inside and outside the container, C_o the nuclide concentration within the waste matrix, r_o the radius of the circular hole. If no permeable medium exists outside the container, the steady-state mass transport for a single medium was derived as follows:

$$\dot{m}_{ss} = 4D_i C_o r_o. \tag{2}$$

It was found that the mass transfer rate through a circular hole increased linearly with the hole radius rather than its area, effective diffusion coefficient, and constant surface concentration.

Chambré et al. [9,10,12] developed the analytical solutions for the time-dependent mass flux through the single and multiple holes, and for the associated concentration field at steady-state. The transient mass flux was given using the time-dependent shape factor, $S(t)$.

$$\dot{m}(t) = D_o C_o r_o S(t). \tag{3}$$

For a circular hole, shape factor was approximated by the expression

$$S(t) \sim 4\left[1 + 2r_o\left(\frac{R}{\pi D_o t}\right)^{1/2}\right], \tag{4}$$

where $R = 1 + ((1-\varepsilon)/\varepsilon)\rho K_d$ is the retardation factor of the nuclide in the porous medium, K_d distribution coefficient of the nuclide in the porous medium, and ε, ρ are the porosity and density of the porous medium. The mass transfer rate at steady-state was the same form as Rae's Eq. (2). If the diffusion coefficient of the surrounding porous medium has a comparable value to that of the waste form, the transient mass flux at time t can be expressed as follows:

$$\dot{m}(t) = 4\frac{D_i D_o}{D_i + D_o} C_o r_o \left[1 + 2r_o\left(\frac{R}{\pi D_o t}\right)^{1/2}\right]. \tag{5}$$

Integrating Eq. (5) from 0 to t gives the total released mass from the waste form, $M(t)$.

$$M(t) = \int_0^t \dot{m}(t)\,dt = 4\frac{D_i D_o}{D_i + D_o} C_o r_o \left[t + 4r_o\left(\frac{Rt}{\pi D_o}\right)^{1/2}\right]. \tag{6}$$

It was indicated that the radionuclide transfer from a waste container with numerous small holes could be significant and might approach the mass transfer rate for a bare (uncontained) waste form of the same dimensions. The reason given was that the steady-state diffusive flux from numerous small holes with the uniform radionuclide concentration was extremely large at all hole edges, leading to more intense mass transfer. Chambré et al. also illustrated the case of holes facing a water gap, instead of being in intimate contact with surrounding porous medium. In this case the radionuclide flux from many small holes approached that from a bare waste cylinder. It was then concluded that waste container with many small holes might not be an effective barrier.

Pescatore and Sastre [11] analyzed the steady-state release from penetrations of the finite length and whose concentration fields interact with one another. They performed the analysis by considering a hypothetical cylindrical unit cell for transport calculations. They showed that the steady-state release of radionuclides from multiple holes could not exceed the release from a bare waste form and that the predicted release from these penetrations was lower than the previously calculated release from holes of zero thickness. It was also indicated that for finite length holes the release was proportional to the area of the hole and not to its radius.

DePaoli and Scott [13] developed a numerical solution of the diffusive transport equations for both the waste form and the surrounding media. Both media were treated in the transient case and preferential adsorption was included. The dimensionless contaminant release rate was found to vary over several orders of the magnitude depending on the product of the ratio of the distribution coefficient and the media diffusivities only.

LeNeveu [14] recently developed the analytical solutions for both transient and steady-state mass transfer rates from a pinhole and for two types of source boundary conditions: constant and inventory-limited concentration. Mass transport of radionuclides was developed considering the properties of the interior of the

pinhole, the exterior of the container, and the interior of the container.

2.2. Pinhole release model

The models developed up to now assume an imaginary cylindrical cell or semi-infinite geometry for the analysis of the mass transfer through perforated waste container. Furthermore the nuclide concentration within the waste form is assumed to be constant. However, the actual concentration distribution of the nuclide that would be released from the waste medium to a small hole would take the form of hemisphere inside the container as shown in Figs. 1 and 2. The initial nuclide concentration within the waste form may be below or above solubility. If below (sub-saturation), the nuclide concentration in pore water of the waste form will decrease continuously with time, and if above (super-saturation), the nuclide concentration will be maintained at the solubility limit.

When the dissolved concentration of the nuclide in pore water of the waste form is at uniform concentration C_o below solubility, the release is mainly governed by diffusion. As shown in Fig. 1, assuming the perfect sink condition at the pinhole ($r = r_o$) and the constant concentration at a certain diffusional distance ($r = r_d$), the hemispherical inward release is described using Fick's law.

$$\frac{\partial C}{\partial t} = \frac{D}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial C}{\partial r}\right) \quad (r_o < r < r_d), \tag{7}$$

where $C = C(r,t)$ is the nuclide concentration (g/cm^3) in pore water of the waste form at position r and at time t,

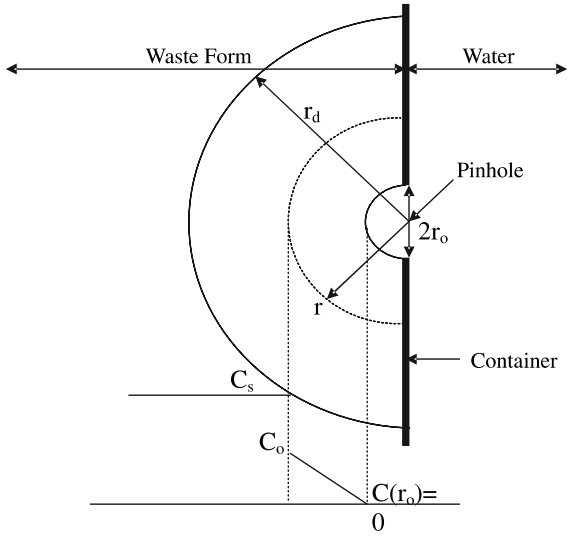

Fig. 1. Conceptual diagram of the pinhole release model for sub-saturation.

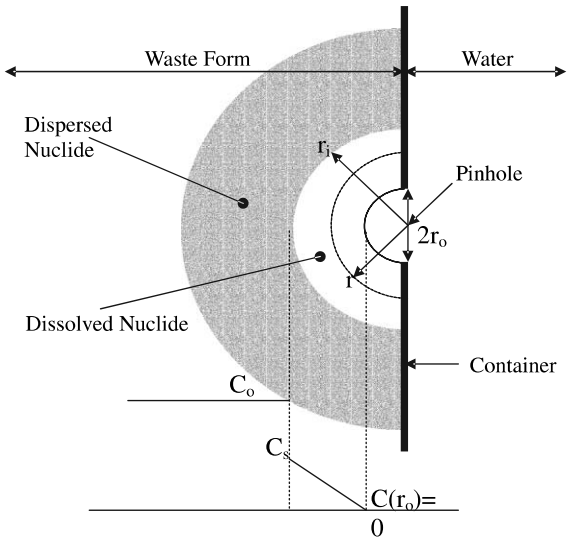

Fig. 2. Conceptual diagram of the pinhole release model for super-saturation.

and D is the nuclide diffusion coefficient (cm^2/s) within the waste matrix. The boundary and initial conditions for the above equation are as follows:

$$C(r_o, t) = 0, \quad t > 0, \tag{8}$$

$$C(r_d, t) = C_o, \quad t > 0, \tag{9}$$

$$C(r, 0) = C_o, \quad r_o < r < r_d. \tag{10}$$

The solution of Eq. (7) is obtained as follows [16,17]:

$$C(r,t) = \frac{r_d(r - r_o)C_o}{r(r_d - r_o)} + \frac{2r_oC_o}{\pi r}\sum_{n=1}^{\infty}\frac{1}{n}\sin\frac{n\pi(r - r_o)}{r_d - r_o} \exp\left(-\frac{Dn^2\pi^2 t}{(r_d - r_o)^2}\right). \tag{11}$$

The release rate out of the pinhole and into the water can be derived from the Fick's law for the flux:

$$\vec{J} = -D\vec{\nabla}C = -D\left(\frac{\partial C}{\partial r}\right)\hat{r}, \tag{12}$$

where the gradient reduces in the third relation to that for radial diffusion in which $\hat{r}$ is the unit normal in accordance with the coordinate convention defined in Fig. 1 (which points from the container interface into the waste form). The nuclide release out of the container will occur from a surface $\vec{A} = \hat{n}S$ where $\hat{n}(= -\hat{r})$ is the outward pointing unit normal from the container and $S = 2\pi r_o^2$ (i.e., from the pinhole geometry, S is simply one-half of the surface area for a sphere of radius r_o). Hence, the release rate out of the pinhole and into the

water, $\dot{m}(t)$, is defined as the dot product between these vector quantities:

$$\dot{m}(t) = \left(\vec{J}\cdot\vec{A}\right)\Big|_{r=r_o} = 2\pi r_o^2\left(\frac{\partial C}{\partial r}\right)_{r=r_o}. \tag{13}$$

The rate at which nuclide is inwardly released from the waste form is calculated as

$$\dot{m}(t) = \frac{2\pi r_o r_d DC_o}{(r_d - r_o)} + \frac{4\pi r_o^2 DC_o}{(r_d - r_o)}\sum_{n=1}^{\infty}\exp\left(-\frac{Dn^2\pi^2 t}{(r_d - r_o)^2}\right). \tag{14}$$

Assuming the steady-state ($t \to \infty$) and the radius of the pinhole is too small ($r_d - r_o \approx r_d$), the steady-state pinhole release model for sub-saturation is derived by

$$\dot{m}_{ss} = 2\pi r_o DC_o. \tag{15}$$

Similarly to the previous model (Eq. (2)), the mass release rate through a pinhole increases linearly with the radius of the pinhole, the diffusion coefficient, and the nuclide concentration within the waste form although the concentration distribution inside container is considered. But the initial constant is 2π rather than 4 because the three-dimensional hemispherical diffusion is analyzed in our study. The mass of the nuclide released through the pinhole at time t, $M(t)$, can be obtained from the integrating the above equation.

$$M(t) = \frac{2\pi DC_o r_o r_d}{(r_d - r_o)}t + \frac{4r_o^2(r_d - r_o)C_o}{\pi} \times \sum_{n=1}^{\infty}\frac{1}{n^2}\left[1 - \exp\left(-\frac{Dn^2\pi^2 t}{(r_d - r_o)^2}\right)\right]. \tag{16}$$

When the nuclide is incorporated above solubility into waste form, the sharp interface or receding dissolution front exists as shown in Fig. 2. The moving front separates a region of the completely dissolved nuclide from a region in which nuclide is either in the immobile dispersed state or in the saturated suspension. It was observed from the previous studies that the leaching mechanism of paraffin wastes could be well explained by this receding dissolution front rather than simply by typical diffusion model [18–20]. The governing equation of the pinhole release model for super-saturation is as follows:

$$\frac{\partial C}{\partial t} = \frac{D}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial C}{\partial r}\right), \quad (r_o < r < r_i), \tag{17}$$

$$C(r_o, t) = 0, \quad t > 0, \tag{18}$$

$$C(r_i, t) = C_s, \quad t > 0, \tag{19}$$

$$C(r, 0) = C_o, \quad r_o < r < r_i, \tag{20}$$

where r_i is radial coordinate of the interface between dispersed and dissolved nuclide within the waste form, C_o and C_s is the initial concentration of the nuclide loaded and nuclide solubility, respectively. The solution of Eq. (17) is given by [16,17]

$$C(r,t) = \frac{r_i(r - r_o)C_s}{r(r_i - r_o)} + \frac{2}{\pi r}\sum_{n=1}^{\infty}\frac{r_i(C_o - C_s)(-1)^{n+1} + r_o C_o}{n} \sin\frac{n\pi(r - r_o)}{r_i - r_o}\exp\left(-\frac{Dn^2\pi^2 t}{(r_i - r_o)^2}\right). \tag{21}$$

The release rate of the nuclide is then

$$\dot{m}(t) = \frac{2\pi r_o r_i DC_s}{(r_i - r_o)} + \frac{4\pi r_o D}{(r_i - r_o)}\sum_{n=1}^{\infty}\left(r_i(C_o - C_s)(-1)^{n+1} + r_o C_o\right)\exp\left(-\frac{Dn^2\pi^2 t}{(r_i - r_o)^2}\right). \tag{22}$$

Here, r_i is a function of the time and an expression for the variation of r_i with time can be derived from the case of a moving boundary diffusion problem [16]. In order that the front can advance a distance δr_i we need to supply an amount $(C_o - C_s)\delta r_i$ of diffusant. The amount arriving at r_i in a time interval δt is $|-D\delta t(\partial C/\partial r)_{r=r_i}|$. Conservation at the moving boundary yields

$$(C_o - C_s)\frac{dr_i(t)}{dt} = D\left(\frac{\partial C}{\partial r}\right)_{r=r_i}, \tag{23}$$

$$(C_o - C_s)\frac{dr_i(t)}{dt} = \frac{r_o DC_s}{r_i(r_i - r_o)} + \frac{2D}{r_i(r_i - r_o)}\sum_{n=1}^{\infty}\left(r_o C_o(-1)^n - r_i(C_o - C_s)\right)\exp\left(-\frac{Dn^2\pi^2 t}{(r_i - r_o)^2}\right) \tag{24}$$

with initial condition

$$r_i(0) = r_o. \tag{25}$$

The above equation cannot be solved analytically but numerically. Thus we perform the pseudo-steady-state (PSS) approximation in order to understand the asymptotic behavior of the above system and to obtain the insight into the mechanism. PSS approximation assumes that at any instant in time, the steady-state concentration distributions exist in the time-dependent partially released region. The dissolution rate at the moving front can be expressed as follows:

$$\dot{m}_{1,pss} = (C_o - C_s)(2\pi r_i^2)\frac{dr_i}{dt}. \tag{26}$$

The diffusion rate, $\dot{m}_{2,\mathrm{pss}}$, through the reaction layer between r_i and r_o is obtained from the steady-state Fick's law.

$$\frac{\mathrm{d}}{\mathrm{d}r}\left(r^2\frac{\mathrm{d}C}{\mathrm{d}r}\right)=0, \quad (r_\mathrm{o}<r<r_\mathrm{i}), \tag{27}$$

$$C=C_\mathrm{s}, \quad r=r_\mathrm{i}, \tag{28}$$

$$C=0, \quad r=r_\mathrm{o}, \tag{29}$$

$$\dot{m}_{2,\mathrm{pss}}=2\pi r_\mathrm{o}^2 D\left(\frac{\mathrm{d}C}{\mathrm{d}r}\right)_{r=r_\mathrm{o}}=2\pi DC_\mathrm{s}\frac{r_\mathrm{i}r_\mathrm{o}}{r_\mathrm{i}-r_\mathrm{o}}. \tag{30}$$

At steady-state ($t\to\infty$), the dissolved nuclide interface has progressed far enough to assume that $r_\mathrm{i}\gg r_\mathrm{o}$. Therefore, the release behavior of the nuclide at long times is explained as nearly linear or zero-order.

$$\dot{m}_{2,\mathrm{pss}}=2\pi DC_\mathrm{s}r_\mathrm{o}. \tag{31}$$

Note that the nuclide solubility is important in case of the super-saturation while the nuclide concentration in pore water of the waste form is the controlling factor for sub-saturation. Equating Eqs. (26) and (30) and integrating the resultant equation from $t=0$ to t and $r_\mathrm{i}=r_\mathrm{o}$ to r_i gives

$$\frac{r_\mathrm{o}^2}{6}+r_\mathrm{i}^2\left(\frac{r_\mathrm{i}}{3r_\mathrm{o}}-\frac{1}{2}\right)=\frac{DC_\mathrm{s}}{C_\mathrm{o}-C_\mathrm{s}}t. \tag{32}$$

3. Experimental

Low-level liquid borate wastes generated from the operation of Korean nuclear power plants have been immobilized with paraffin wax using the concentrate waste drying system (CWDS) since 1995 [15]. CWDS has the advantages of the high volume reduction (about 1/8 compared with that of the cement waste form), decrease in radiation expose dose to worker (1/9), low cost, and simple manufacturing process. The CWDS has produced the paraffin waste form with the mixing ratio of 78:22 (%) between borate concentrate and paraffin wax.

Boric acid and paraffin wax are two major constituents of the paraffin waste form. The properties of the paraffin wax are specific gravity of 0.933, melting temperature of 72 °C, insolubility in water, and thermoplastic material. Boric acid has the specific gravity of 1.44 and the melting point of 171 °C. The waste form is prepared under the conditions on mixing temperature of 120–140 °C, stirrer speed of 600 rpm, and the operation time of about 15 min. The mixture is poured into cylindrical PVC mold with a thickness of 2 mm and hardened at a room temperature for a week. Cylindrical waste package with a inner diameter of 5 cm, a outer diameter of 5.2 cm, and a height of 10 cm is prepared. In order to simulate the pinhole of the waste package, the lateral side of the cylindrical mold is perforated before the mixture is put into mold. The circular openings have three kinds of the diameter, e.g., 2 mm, 5 mm and 10 mm as shown in Table 1. The specimens of CASE-1, CASE-2 and CASE-3 have a single hole on their vertical surfaces and the cross-sectional area of the pinhole increases by 6.25 and 25 times. The corresponding fractional surface area of CASE-1, CASE-2 and CASE-3 is also about 1.53×10^{-4}, 9.54×10^{-4} and 3.82×10^{-3}, respectively. The cases for multiple holes are not considered here in order to avoid the complex interactions among them.

Table 1
Specimens used in the test

	Diameter of the pinhole (mm)	Fractional surface area
CASE-1	2	1.53×10^{-4}
CASE-2	5	9.54×10^{-4}
CASE-3	10	3.82×10^{-3}

The mixing ratio of boric acid to paraffin is very important in order to make a homogeneous waste form because it makes a difference between boric acid and paraffin in specific gravity. If the paraffin content is lower than 20%, it is very difficult to make a waste form due to a low fluidity. While the stratification begins to occur if the paraffin content exceeds a certain limit of the mixing ratio. Therefore the waste forms whose paraffin contents are within the range of 20–24 are easily manufactured owing to good workability, and the corresponding compressive strengths exhibit the maximum value of 622–673 psi (4.23–4.58 MPa).

The leaching test is performed according to ANSI/ANS-16.1 procedure [21]. The test is developed by an American Nuclear Society (ANS) Standards Committee for the characterization of solidified low-level radioactive waste forms. This procedure uses the demineralized water as leachant and is conducted at a temperature of (22.5 ± 5) °C. The ratio of leachant volume to external surface area of the waste form is (10 ± 0.2) cm. The leachant is sampled and replaced at the following frequency: 2, 7 and 24 h from the initiation of the test, then at 24 h intervals for the next 4 days, then at 14, 28 and 43 days intervals to extend the entire test to 90 days. Nonradioactive soluble chemical of the cobalt(II) chloride hexahydrate is substitutively used in order to simulate the leaching behavior of radioactive species immobilized within the paraffin waste form. The concentration of the cobalt included in leachate is measured by means of the inductive coupled plasma-mass spectroscopy and that of the boric acid within the leachate is analyzed by the titration.

4. Results and discussion

Fig. 3 shows the comparison between model prediction and test results for cobalt. As given in Table 2, the nuclide concentration of the cobalt is so lower than its solubility by 566 times that the developed pinhole release models for sub-saturation, Eqs. (14) and (15), agree well with experimental data. The used diffusion coefficient is obtained from the previous leaching test for bare waste form [18–20]. The constant concentration distance, r_d, is fixed to $10r_o$ according to the results of earlier studies where it was reported that the concentration distribution had fallen to the negligible level about 10 hole radii away [8–10,12].

It is observed that the pinhole release rates rapidly approach to the zero-order kinetics except for an initial period. This behavior is different from the release rate of the monolithic waste form whose external surface is entirely exposed to the water. In case of the monolithic waste form, the release rate is proportional to the reciprocal of the square root of time. This is because the release rate is inversely proportional to the distance where the nuclide must travel from within the matrix to the surface of the waste form. Since this diffusional distance increases with time as the nuclide is released, the release rate decreases. However, in the present pinhole situation, it is assumed that the increasing hemispherical dissolution surface compensates for this increasing diffusional distance and zero-order release kinetics can be achieved.

The steady-state release rates are directly proportional to the diffusion coefficient of the nuclide, the nuclide concentration or the solubility depending on the sub- or super-saturated loading, and the radius of the pinhole not to its area. The steady-state release rates of CASE-1, CASE-2 and CASE-3 are calculated as 1.84×10^{-6}, 4.59×10^{-6} and 9.18×10^{-6} g/day, respectively. As expected intuitively, the CASE-3 with the largest hole arrives at the steady-state release rate the most late than the others.

As shown in Figs. 4 and 5, the pinhole release of the super-saturated boric acid is also satisfactorily explained by Eqs. (30) and (32). The release behavior is similar to that of the cobalt, but the magnitude is greater than 2000 times because of the highly loaded boric acid. The used diffusion coefficient is also obtained from the previous leaching test for the whole surface of the bare waste form [18–20]. It should be noted that the

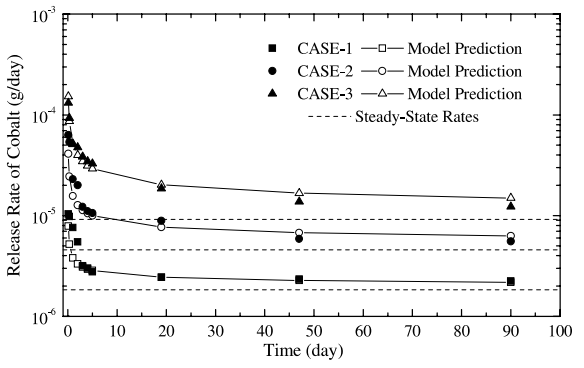

Fig. 3. Comparison between model prediction and test data for cobalt.

Table 2
Parameters of the pinhole release model

	Sub-saturation (cobalt)	Super-saturation (boric acid)
r_o (cm)	0.1, 0.25, 0.5	0.1, 0.25, 0.5
r_d (cm)	$10r_o$	–
r_i (cm)	–	Eq. (30)
D (cm^2/sec)	5.71×10^{-8}	1.57×10^{-6}
C_o (g/cm^3)	5.92×10^{-4}	0.9918
C_s (g/cm^3)	0.335 at 20 °C	0.05 at 20 °C

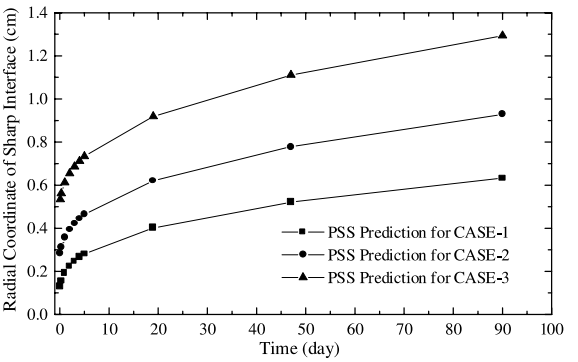

Fig. 4. Radial coordinates of sharp interface for boric acid.

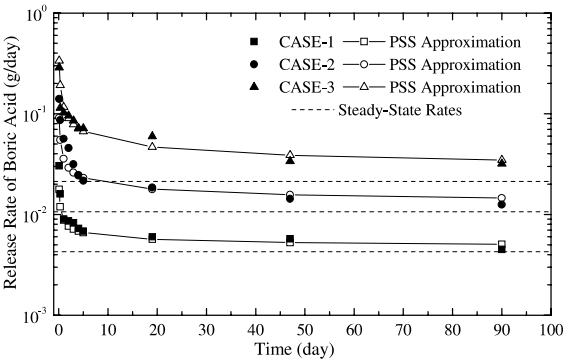

Fig. 5. Comparison between model prediction and test data for boric acid.

steady-state release rate of the boric acid is linear with the nuclide solubility, and that solute diffusion is the rate-controlling step rather than nuclide dissolution.

5. Conclusions

In order to model the nuclide release from a perforated radioactive waste container, the more realistic pinhole release model considering the ratio of the nuclide concentration to solubility and the moving dissolution front is developed. It is found that the pinhole release rates are nearly linear or zero-order except for a brief period initially. The steady-state release rates are slightly greater than that of the earlier developed model by $\pi/2$ owing to the inwardly releasing hemisphere. The developed pinhole release model predicts well the experimental data using the leaching test of the paraffin waste form.

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