Transactions of the Korean Nuclear Society Autumn Meeting
Gyeongju, Korea, October 26-27, 2017

Modeling of the Release of a Slowly Diffusing Fission Product from a Spherical Particle under Annealing

Young Min Kim[1] and Chang Keun Jo
Korea Atomic Energy Research Institute
111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, 34057, Republic of Korea
[1] *Corresponding author: nymkim@kaeri.re.kr*

1. Introduction

In a high temperature reactor (HTR), many fission products are released from a coated fuel particle which usually consists of a fuel kernel and four surrounding coating layers, through graphite matrix, finally into a coolant. A numerical method using a finite difference scheme, COPA, has been developed for the estimation of the fission product release [1]. The numerical method has to be verified for the exact estimation of the fission product release. Some simple analytical solutions were already found for a fission product release from a sphere under irradiation and annealing [2-4].

It was, however, found during the verification of the numerical solution that the existing analytical solutions overestimated the release of a radioisotope with low diffusivity such as ^{90}Sr from a kernel under annealing. It was judged that the analytical solutions estimated an excessive release for a radioisotope with low diffusivity from the start.

This study presents a new form of an analytical solution which is appropriate to estimate the release of a slowly diffusing fission product from a sphere under annealing. The new and existing analytical solutions and the COPA solution were applied to the calculation of ^{90}Sr and ^{137}Cs releases from a kernel under annealing, and their results were compared with each other.

2. Diffusion of a Radioisotope in a Sphere

A transport of a radioisotope within a spherical particle can be described by the following Fickian diffusion equation.

$$\frac{\partial C(r,t)}{\partial t} = -\lambda C(r,t) + \frac{D}{r}\frac{\partial^2 (rC)}{\partial r^2}, \quad (1)$$

where C is the concentration of a radioisotope (mol m^{-3}), λ is the decay constant (s^{-1}), D is the diffusion coefficient (m^2 s^{-1}), r is the radial coordinate (m), t is the time (s). The diffusion coefficient is assumed to be dependent on temperature only, and it is volume-averaged. The diffusion current at the particle center is zero on the assumption of radial symmetry. It is assumed that the concentration at the particle surface is equal to zero.

The initial concentration distribution in the particle has been historically assumed to be flat over the radius of the spherical particle.

$$C(r,0) = C_0 , \quad (2)$$

where C_0 = the initial concentration which is constant throughout the radius of a particle (mol m^{-3}). In this study, the following exponential initial condition is also considered.

$$C(r,0) = C_0 \left[1 - \frac{a \sinh\left(\sqrt{\mu}\frac{r}{a}\right)}{r \sinh\sqrt{\mu}} \right], \quad (3)$$

where $\mu = \lambda a^2/D$, and a = the particle radius (m). Fig. 1 shows the initial concentrations according to the flat and exponential initial conditions. The exponential initial condition originally expresses a steady-state distribution for a short-lived fission product during irradiation. It can be applied to a very slowly diffusing radioisotope of which the concentration is zero at the particle surface.

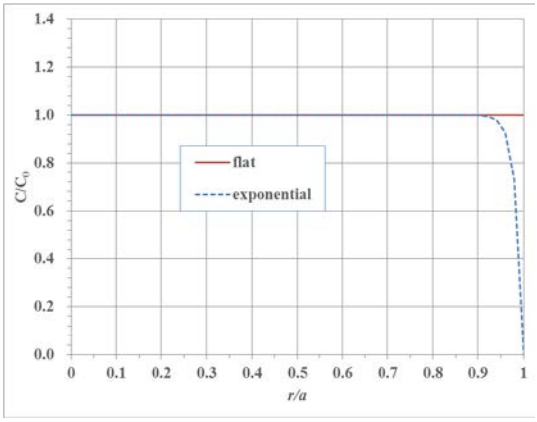

Fig. 1. Flat and exponential types of initial conditions

The analytical solutions can be obtained using a Fourier transformation. The analytical fractional release with a flat initial condition has the following form [4].

$$F = e^{-\lambda t}\left(1 - 6\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2}\right), \quad (4)$$

where $\tau = \int_0^t \frac{D}{a^2}dx$. And the analytical fractional release with an exponential initial condition is as follows.

$$F = e^{-\lambda t}\left[1-\frac{6\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2}-6\sum_{n=1}^{\infty}\frac{e^{-n^2\pi^2\tau}}{n^2\pi^2+\mu}}{1-\frac{3}{\sqrt{\mu}}\left(\coth\sqrt{\mu}-\frac{1}{\sqrt{\mu}}\right)}\right]. \quad (5)$$

For $\tau \le 0.1$, Eqs. (4) and (5) approximate to Eqs. (6) and (7), respectively.

$$F \approx e^{-\lambda t}\left(6\sqrt{\tau/\pi}-3\tau\right), \quad (6)$$

$$F \approx e^{-\lambda t}\left[1-\frac{1-6\sqrt{\tau/\pi}+3\tau-\frac{3e^{\mu\tau}}{\sqrt{\mu}}\left(\coth\sqrt{\mu}-erf\sqrt{\mu\tau}\right)+\frac{3}{\mu}}{1-\frac{3}{\sqrt{\mu}}\left(\coth\sqrt{\mu}-\frac{1}{\sqrt{\mu}}\right)}\right]. \quad (7)$$

3. Calculation of ^{90}Sr and ^{137}Cs Releases

The case number one of the accident benchmark problems of a coordinated research program (CRP) of the International Atomic Energy Agency (IAEA), IAEA CRP-6, is the ^{137}Cs releases from a bare UO_2 kernel under heating at constant temperatures of 1200 and 1600 °C for two hundred hours [1]. ^{90}Sr is included in this study because its diffusivity in a UO_2 kernel is much less than that of ^{137}Cs.

Figs. 2 and 3 show the fractional releases of ^{90}Sr from a bare UO_2 kernel under heating at temperatures of 1200 and 1600 °C, respectively. The numerical solutions are in good agreement with the analytical solutions with an exponential initial condition at 1200 °C, but they move toward the analytical solutions with a flat initial condition at 1600 °C.

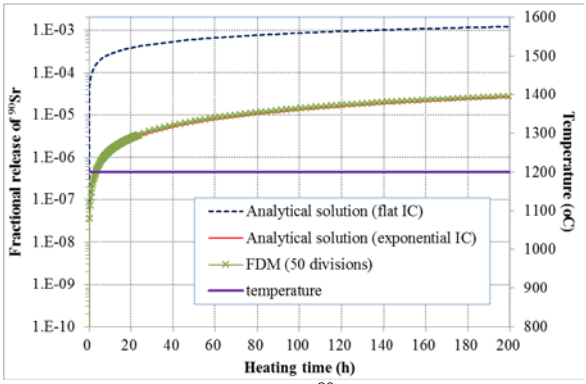

Fig. 2. Fractional release of ^{90}Sr during annealing at 1200 °C.

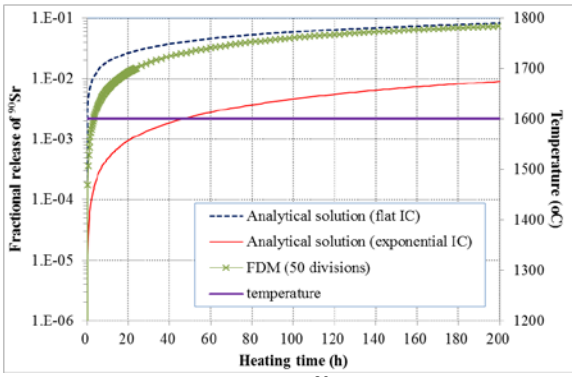

Fig. 3. Fractional release of ^{90}Sr during annealing at 1600 °C.

Figs. 4 and 5 display the fractional releases of ^{137}Cs from a bare UO_2 kernel under heating at temperatures of 1200 and 1600 °C, respectively. The numerical solutions are in good agreement with the analytical solutions with a flat initial condition even though some minor differences exist at the early stage of heating.

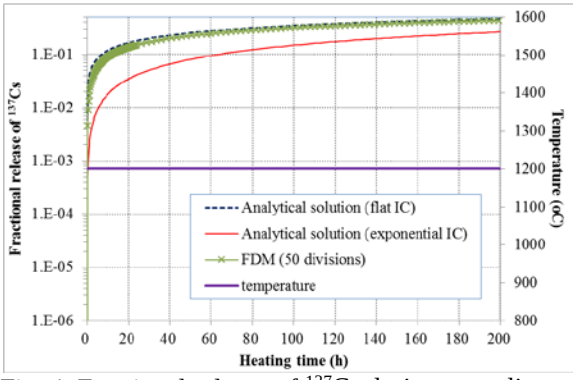

Fig. 4. Fractional release of ^{137}Cs during annealing at 1200 °C.

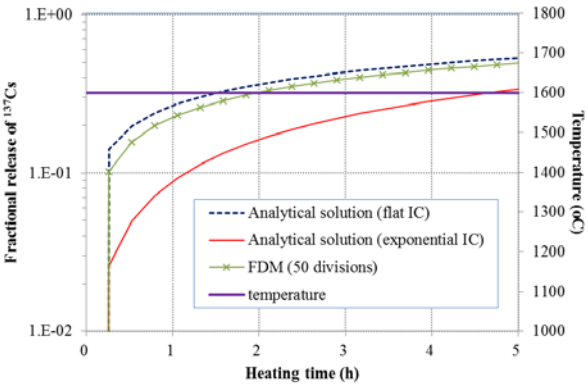

Fig. 5. Fractional release of ^{137}Cs during annealing at 1600 °C.

Fig. 6 shows the μ values of ^{90}Sr and ^{137}Cs according to temperature. The μ value of ^{90}Sr is very large at 1200 °C. That's because the diffusivity of ^{90}Sr in a UO_2 kernel is much less than that of ^{137}Cs, and the decay constants of the two radioisotopes are nearly the same.

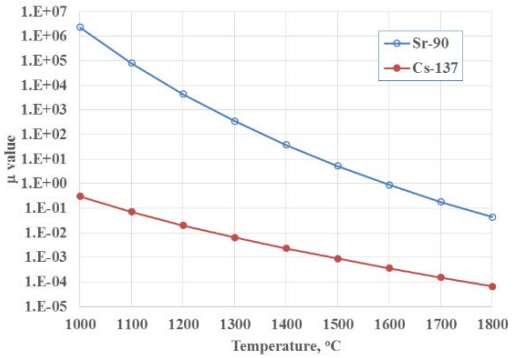

Fig. 6. μ values of ^{90}Sr and ^{137}Cs during annealing.

4. Summary

It is highly recommended to use an analytical solution with an exponential type of an initial condition to verify numerical solutions for a release of a radioisotope with low diffusivity such as ^{90}Sr. Numerical solutions are between the analytical solutions using the flat and exponential initial conditions. As the μ value becomes smaller, the numerical solutions approach to the analytical solutions with a flat initial condition from the analytical ones with an exponential initial condition. Even for a radioisotope with a high diffusivity, ^{137}Cs, some minor difference exists between the analytical and numerical solutions at the early stage of heating.

ACKNOWLEDGEMENTS

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIP) (No. 2017M2A8A1014757).

REFERENCES

[1] IAEA, “Advances in High Temperature Gas Cooled Reactor Fuel Technology,” IAEA-TECDOC-1674, 2012.
[2] Beck, S.D., “The Diffusion of Radioactive Fission Products from Porous Fuel Elements,” BMI-1433, 1960.
[3] Lewis, B.J. and Iglesias, F.C., A Model for Calculating Fission Gas Diffusion in Annealing Experiments, Journal of Nuclear Materials 154, pp. 228-232, 1988.
[4] Lewis, B.J., Evens, D., Iglesias, F.C., and Liu, Y., Modelling of Short-Lived Fission Product Release Behaviour during Annealing Conditions. Journal of Nuclear Materials 238, pp. 183-188, 1996.