

Effect of Oxidants from Radiolysis of Groundwater on the Corrosion of a High-Level Radioactive Waste Canister

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1. Introduction

The spent fuels from the nuclear power plants will be directly disposed of in the repository. It is planned to store them in the interim storage facility more than 40 years before the final disposal, during which we expect most of short-lived radionuclides might decay. However, there are still lots of long-lived nuclides in the spent fuels. The radiations from these radionuclides make it difficult to deal with the high-level wastes. Also, they cause the radiolysis of groundwater, and the oxidants from the groundwater radiolysis corrode the canister.

Several kinds of canisters for the high-level radioactive wastes have been developed by KAERI. The canisters consist of two layers, one for an outer shell for the corrosion resistance and another for an insert. This kind of double layered canister was introduced in the countries such as Sweden and Finland in which the lifetime of the canister was asked to be more than 100,000 years. It is thought that the radioactivity in the spent fuel will decay to the level of uranium mine. According to the design requirements for the canister prepared by KAERI, the lifetime of the canister should be longer than 1,000 years like Japan. If the lifetime of the canister is 1,000 years, the introduction of outer shell for the corrosion resistance should be evaluated in detail. In this case the effect of oxidants from the radiolysis of groundwater on the carbon steel canister should be assessed.

Marsh et al.[1] modeled the effect of oxidants generated from the radiolysis of groundwater on the carbon steel around high-level radioactive wastes. They used the G-value to calculate the amount of oxidants generated from the radiolysis of groundwater. The transport of oxidants around the waste canister was modeled by diffusion only due to too low groundwater velocity. They used one-dimensional mass transport equation in rectangular coordinate and obtained an analytical solution. Japanese H12 report [2] followed the same approach to analyze the effects of radiolysis on the corrosion of the canister and determine the thickness of the canister, but unfortunately they misunderstood the exact meaning of the apparent diffusion coefficient in the porous media, which underestimated their calculation.

The main purpose of this study is to develop a new model for the calculation of oxidant concentration distribution around the canister. We introduced the cylindrical coordinate to calculate the oxidant concentration around the canister since it seemed to be more realistic than the rectangular coordinate for the

modeling of canister. A simple approach to calculate the cathodic current density from the governing equation was developed. We compared the new solution with that of Marsh et al.[1].

2. Mathematical Model

Many kinds of oxidizing chemical species were generated by the radiolysis of the groundwater in the pore of buffer due to gamma radiation and neutron emitted from the spent fuels in the canister. The number of production rates of the oxidizing species was expressed with G-value (molecules produced per 100eV of radiation). The oxidizing species were assumed not to be absorbed by the buffer material and to migrate in the buffer material by diffusion due to the low groundwater velocity. The diffusion equation in the cylindrical geometry was given as follows:

$$\frac{\partial C}{\partial t} = D \frac{1}{r} \frac{\partial}{\partial r} r \frac{\partial C}{\partial r} + R_0 e^{-k|r-r_0|} \quad (1)$$

$$R_0 = \frac{PGE}{100A} \quad (2)$$

The equation (2) was the source term for oxidizing species generated by the radiolysis of groundwater. E in equation (2) means the absorbed dose, and A means the Avogadro number.

The boundary and initial conditions are as follows:

$$C(t=0, r) = 0 \quad (3)$$

$$C(t, r=r_0) = 0 \quad (4)$$

For the corrosion to happen the flux of oxidizing species should support a cathodic current that at least balances the anodic dissolution current flowing through the protective layer. The cathodic current density was obtained using the equation (1) as follows:

$$I_{cath} = -nFD \left. \frac{dC}{dr} \right|_{r=r_0} \quad (5)$$

3. Calculation of Absorbed Dose

The absorbed doses given in Table 1 were calculated with MCNP4c2 computer program for the calculation of the equation (2). MCNPLIB02 and ENDF-VI were used for the calculation of the absorbed doses for photons and neutrons, respectively. Figure 1 showed the canister for PWR spent fuels and a borehole with the canister and buffer material. The specifications of the borehole were cited from the SKI report [3]. The calculation showed that the absorbed doses from photons were much larger than those from the neutrons and secondary photons.

Radius(cm)	Total Absorbed Dose Rate (Gy/hr)		Total Absorbed Dose Rate (eV/m ³ s)	
	4.0w/o	4.5w/o	4.0w/o	4.5w/o
	40.0	1.998E+00	2.445E+00	3.464E+18
42.5	4.911E-01	6.024E-01	8.515E+17	1.044E+18
45.0	1.290E-01	1.591E-01	2.237E+17	2.759E+17
47.5	3.369E-02	4.199E-02	5.841E+16	7.280E+16
50.0	9.461E-03	1.192E-02	1.640E+16	2.067E+16
52.5	2.848E-03	3.667E-03	4.938E+15	6.358E+15
55.0	9.565E-04	1.278E-03	1.658E+15	2.216E+15
Reference	1.198E-03	1.575E-03	2.077E+15	2.731E+15

Table 1. Total absorbed dose rate in radial direction

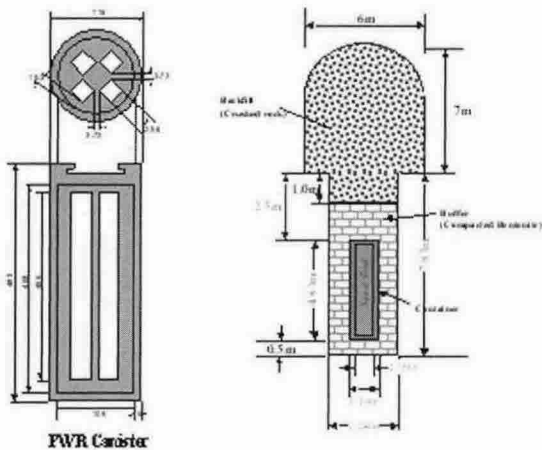


Figure 1. schematic of the canister for PWR fuels.

4. Calculation of Cathodic Current Density

The cathodic current density given in the equation (4) needs not the concentration of oxidizing species but the first derivative of the concentration at the surface of the canister. The first derivative especially at steady state was simply obtained by integrating the right hand side term of the equation (1). The sample calculation was conducted to show the advantage of introducing a cylindrical geometry over the rectangular geometry that Marsh et al. used with the parameters in Table 2. The cathodic current density by Marsh et al. model was

$6.63 \times 10^{-3} \text{ A/m}^2$, and the model developed in this study gave $4.86 \times 10^{-3} \text{ A/m}^2$.

Parameter	Value
P, porosity	0.1
G-value	2.13
E, absorbed dose, eV/m ³ s	6.07×10^{19}
A, Avogadro number	6.022×10^{23}
n, valance of oxidant	2
F, Faraday constant	9.65×10^4
K, absorption coefficient, m ⁻¹	6.25
r ₀ , radius of canister, m	0.6

Table 2. Parameters for the model comparison

5. Conclusion

The oxidizing species were generated from the radiolysis of groundwater in the pore of buffer material around the canister used for the disposal of spent fuels. A new model was introduced to calculate the cathodic current density around the canister, which determines the corrosion of carbon steel. We developed a simple method to get the cathodic current density in the cylindrical coordinate. Using the solution, the cathodic current densities from both the rectangular coordinate and cylindrical coordinate were compared. The results showed that the new model improved the calculation remarkably. The thickness of the canister was calculated with the new model.

Acknowledgement

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