Failure Mechanism of Concrete Barriers in Low- and Intermediate-Level Waste (LILW) Disposal Facility

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1. Introduction

Concrete barriers have been used as the structural components of low- and intermediate-level waste (LILW) disposal facilities[1]. They have played major roles to inhibit water infiltration into a LILW disposal facility and the release of radionuclides from a LILW disposal facility.

However, the permeability of the concrete barriers slowly increases over a period of time because of various degradation mechanisms[2]. After a long period of time in the future, the concrete barriers would lose their effectiveness as barriers against groundwater infiltration and the release of radionuclides. Therefore, the concrete barriers of a LILW disposal need to maintain their physico-chemical soundness for certain period of time based on the results of safety assessment.

This paper briefly reviews the chemical processes which affect concrete degradation along with mathematical models. In addition, the main failure mechanism of the concrete barriers of a LILW disposal facility is investigated by applying those models.

2. Concrete Degradation Processes

A number of processes are responsible for concrete degradation in the subsurface environment. In general, the major degradation processes include acid attack, sulfate attack, the corrosion of reinforcement steel induced by chloride attack, calcium hydroxide leaching, alkali-aggregate reaction, and carbonation. Of those, sulfate attack, the corrosion of reinforcement steel induced by chloride attack, and calcium hydroxide leaching are reviewed and evaluated in this paper, because the disposal facility is located below over 80 m of ground surface and saturated with neutral groundwater.

Sulfate attack. Through a chemical reaction, sulfate ion penetrates into the surface of concrete and weakens the concrete wall. The model for sulfate attack was already proposed by Atkinson and Hearne[3]. The degradation rate of concrete from sulfate attack is estimated by:

$$R = \frac{EB^2 c_o C_E D_o}{\alpha \gamma (1-\nu)} \qquad (1)$$

where, B is linear strain caused by one mole of sulfate reacted in 1 m^3 (1.8×10^{-6} m^3/mol), c_o is sulfate concentration in water (mol/m^3), C_E is concentration of

reacted sulfate as ettringite (mol/m^3), D_o is intrinsic diffusion coefficient (m^2/s), E is Young's modulus (20 GPa), α is roughness factor for fracture path (assumed to be 1.0), γ is fracture surface energy of concrete (10 J/m^2), υ is Poisson's ratio (0.3), and the rate of degradation, R, is in (m/yr).

Calcium hydroxide leaching Leaching of concrete by groundwater removes soluble compounds such as $Ca(OH)_2$ from the hardened concrete. With these compounds removed, the compressive strength of the concrete gradually decreases. A shrinking core model proposed by Atkinson and Hearne[3] is adopted here. The model assumes the calcium removal from the concrete exterior is rapid compared to the movement of calcium through the concrete. Thus, the transport of calcium in the concrete is controlled by diffusion. The depth into the concrete affected by Ca removal is then given by :

$$X = \left(2D\tau\phi \frac{C_i - C_{gw}}{C_b} t \right)^{1/2} \qquad (2)$$

where, D is the diffusion coefficient of Ca^{2+} ions in concrete (m^2/s), τ is the tortuosity factor (-), ϕ is the porosity (-), C_i is the concentration of Ca^{2+} ions in the pore space (mmol/kg), C_{gw} is the concentration of Ca^{2+} in groundwater (mmol/kg), and C_b is the bulk concentration of Ca^{2+} ions in solid concrete (mmol/kg).

Corrosion of reinforcement steel When steels corrode, the corrosion products have a greater volume than the initial metal. The expansion can cause the concrete to crack and simultaneously increase the permeability. The concrete pore water tends to passivate the steel surface due to hyper alkaline condition. The passivated condition must be overcome before corrosion starts. The depassivation process is the result of diffusion of chloride ions, through the concrete, to the surface of the reinforcement steel. The diffusion of chloride ion is described by Fick's 2nd law[4]:

$$C(x,t) = C_i + (C_S - C_i) \cdot erf\left(1 - \frac{x}{2\sqrt{D_{Cl}t}} \right) \qquad (3)$$

where, $C(x,t)$ is chloride concentration at depth x at time t, C_i is initial chloride concentration (kg/m^3), C_s is surface chloride content (kg/m^3), D_{Cl} is apparent Cl diffusion coefficient (m^2/s), and *erf* is error function.

Because of limited data for corrosion within the concrete, a conservative rate of 10^{-5} m/yr is assumed[5].

3. Results and Discussion

Groundwater samples were collected from the site of the disposal facility to estimate the amounts of sulfate, chloride, dissolved oxygen (DO), dissolved organic carbon (DOC), pH, and other ions present in the samples.

In sulfate attack, concrete degradation rate is calculated to be 1.03×10^{-3} cm/yr. For the concrete thickness of 60 cm in the disposal facility, this rate is negligible over 5,000 years, and therefore sulfate attack is assumed to be negligible for this analysis. In the model of calcium hydroxide leaching, the Ca concentration in the concrete is calculated to be 15.7 mmol/kg using PHREEQC[6]. After 1,000 yr, the depth of calcium hydroxide leaching is estimated to be approximately 2 cm. Hence, it is concluded that this mechanism is negligible over 5,000 years for the disposal facility.

In the evaluation of Eq. (3), it is unveiled that the degradation rate of the concrete due to chloride attack is much faster than the rates estimated from sulfate attack and calcium hydroxide leaching. Therefore, the predominant failure mechanism would be chloride attack on the reinforcement steel with the volume expansion of steel corrosion products creating internal stresses in the concrete.

4. Conclusion

Chemical processes responsible for the degradation of the concrete barriers are briefly reviewed for the subsurface LILW disposal facility which is saturated with neutral groundwater. Service life of the concrete barriers is estimated with the mathematical models for sulfate attack, the corrosion of reinforcement steel, and calcium hydroxide leaching. Sulfate attack is negligible over 5,000 yr, and the depth of calcium hydroxide leaching is approximately 2 cm after 1,000 yr. Hence, chloride attack is evaluated as the main failure mechanism.

REFERENCES

[1] K. A. Snyder, and J. R. Clifton, 1995, "4SIGHT manual: a computer program for modelling degradation of underground low level waste concrete vaults", NISTIR 5612, National Institute of Standards and Technology.

[2] NCRP Report 152, 2005, "Performance assessment of low-level waste disposal facilities", Report of the national council on radiation protection and measurements.

[3] A. Atkinson and J. A. Hearne, 1990, "Mechanistic models for the durability of concrete barriers exposed to sulfate-bearing groundwaters," in Scientific Basis for Nuclear Waste Management XIII, V.M. Oversby and P.W. Brown eds., MRS Symposium Proceedings, Vol. 176, p. 149-156.

[4] S. Lay, 2003, "Lifecon deliverable D 3.2, Service life model", Technical research centrer of Finland (VTT).

[5] US NRC, 2006, U. S. Nuclear regulatory commission technical evaluation report for the U.S. DoE Idaho national laboratory site draft section 3116, waste determination for Idaho nuclear technology and engineering center tank farm facility.

[6] D. L. Parkhurst and C.A.J. Appelo, 1999, "User's guide to PHREEQC – A computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculation", Water-resources investigations report 99-4259, U.S. Geological survey.