

**RELEASE TO PUBLISH UNCLASSIFIED NRC CONTRACTOR
SPEECHES, PAPERS, AND JOURNAL ARTICLES**
(Please type or print)

1. TITLE (State in full as it appears on the speech, paper, or journal article)
Estimated Longevity of Reducing Environment in Grouted Systems for Radioactive Waste Disposal

2. AUTHOR(s)
S. Painter and R. Pabalan

3. NAME OF CONFERENCE, LOCATION, AND DATE(s)
**Nuclear Energy Agency Workshop on Cementitious Materials in Safety Cases for Geological Repositories:
Role, Evolution, and Interactions
Brussels, Belgium
November 17-20, 2009**

4. NAME OF PUBLICATION
N/A

5. NAME AND ADDRESS OF THE PUBLISHER

TELEPHONE NUMBER OF THE PUBLISHER


6. CONTRACTOR NAME AND COMPLETE MAILING ADDRESS (Include ZIP code)
**Center for Nuclear Waste Regulatory Analysis
Southwest Research Institute®
6220 Culebra Road
San Antonio, TX 78228**

TELEPHONE NUMBER OF THE CONTRACTOR
210-522-5252

YES	NO	7. CERTIFICATION (ANSWER ALL QUESTIONS)		
	X	A. COPYRIGHTED MATERIAL	- Does this speech, paper, or journal article contain copyrighted material? If yes, attach a letter of release from the source that holds the copyright.	
	X	B. PATENT CLEARANCE	- Does this speech, paper, or journal article require patent clearance? If yes, the NRC Patent Counsel must signify clearance by signing below.	
		NRC PATENT COUNSEL (Type or Print Name)	SIGNATURE	DATE
X		C. REFERENCE AVAILABILITY	- Is all material referenced in this speech, paper, or journal article available to the public either through a public library, the Government Printing Office, the National Technical Information Service, or the NRC Public Document Room? If no, list below the specific availability of each referenced document.	
		SPECIFIC AVAILABILITY		
X		D. METRIC UNIT CONVERSION	- Does this speech, paper, or journal article contain measurement and weight values? If yes, all must be converted to the International System of Units, followed by the English units in brackets, pursuant to the NRC Policy Statement implementing the Omnibus Trade and Competitiveness Act of 1988, Executive Order 12770, July 25, 1991.	

8. AUTHORIZATION

The signatures of the NRC project manager and the contractor official certify that the NRC contractor speech, paper, or journal article is authorized by NRC, that it has undergone appropriate peer review for technical content and for material that might compromise commercial proprietary rights, and that it does not contain classified, sensitive unclassified, or nonpublic information. (NRC MD 3.9, Part II(A)(1)(d))

A. CONTRACTOR AUTHORIZING OFFICIAL (Type or print name) Budhi Sagar		SIGNATURE 	DATE 6/11/2009
B. NRC RESPONSIBLE PROJECT MANAGER (Type or print name)		OFFICE/DIVISION	MAIL STOP
TELEPHONE NUMBER	E-MAIL I.D.	Did you place the speech, paper, or journal article in ADAMS? If so, provide the ADAMS Accession No. (Use Template OCIO 099)	YES <input type="checkbox"/> NO <input type="checkbox"/>
SIGNATURE			DATE

ESTIMATED LONGEVITY OF REDUCING ENVIRONMENT IN GROUTED SYSTEMS FOR RADIOACTIVE WASTE DISPOSAL

Scott PAINTER (Center for Nuclear Waste Regulatory Analyses)
Roberto PABALAN (Center for Nuclear Waste Regulatory Analyses)

Introduction

A key factor determining the release and transport of redox-sensitive radioelements (e.g., technetium, selenium) from grouted, near-surface, low-activity waste disposal facilities at U.S. Department of Energy (DOE) sites is the redox potential of the cement-based material. Published studies have shown that a reducing chemical environment mitigates the release and transport of redox-sensitive radioelements from grouted systems. At DOE low-activity waste disposal sites, blast furnace slag is added to the grout formulation to impose an initially reducing chemical condition on the grouted system. This effect has been ascribed to the release of sulfide species, predominantly as S^{2-} , into the pore fluid upon hydration of the slag (Atkins and Glasser, 1992), which imposes a strongly reducing redox potential on the system. There is significant uncertainty regarding the long-term persistence of the reducing capabilities of the slag-bearing grout and its long-term effect on radionuclide release and transport. The objective of this study is to estimate how long reducing conditions could persist in near-surface, slag-bearing grouted systems for radioactive waste disposal. The oxidant of most concern is oxygen—in the gas phase or dissolved in infiltrating water—which could react with the blast furnace slag and decrease the grout reductive capacity.

Computational Approach

The study used a numerical model representing oxygen transport in fractures and porous grout to assess the lifetime of reducing conditions for a range of hydrological conditions, fracturing scenarios, and grout parameter values. The evolution of the oxygen concentration in the grout is represented by coupling one-dimensional advection-dispersion in the fracture with one-dimensional diffusion in the perpendicular direction representing diffusion into the porous grout. Rather than model the detailed oxidation-reduction reactions, the approach of Kaplan, et al. (2005, 2007) was used. In this approach, the oxidation-reduction reaction is written as an effective reaction that consumes oxygen (O_2) and grout-reducing equivalents (R) and produces oxidized grout (RO_2):



The oxygen conservation equation in the fractures is given by

$$\phi_f S_l \frac{\partial C_l}{\partial t} + \phi_f S_g \frac{\partial C_g}{\partial t} + \frac{\partial}{\partial z} (\Omega_l + \Omega_g) = -\psi_{F \rightarrow M} \quad (2)$$

where

ϕ_f	—	fracture internal porosity (with a value of 1 for an open fracture containing no sediment)
S_l and S_g	—	liquid and gas saturation in the fracture
C_l and C_g	—	liquid-phase and gas-phase oxygen concentration
Ω_l and Ω_g	—	liquid-phase and gas-phase fluxes in the fracture

$$\psi_{f \rightarrow m} = \frac{\phi}{b} D_e \left. \frac{\partial C'}{\partial x} \right|_{x=0} \quad \text{--- fracture-to-matrix oxygen transfer rate}$$

t — time
 z — vertical distance from the surface of the grouted system
 b — fracture half-aperture

The oxygen conservation equation in the matrix is given by

$$\phi \frac{\partial C'}{\partial t} - \phi D_e \frac{\partial^2 C'}{\partial x^2} = -kC'R\rho_g \quad (3a)$$

$$\frac{\partial R}{\partial t} = -kC'R \quad (3b)$$

where

ϕ — matrix porosity
 ρ_g — bulk density of the grout
 D_e — effective diffusion coefficient in the matrix
 C' — oxygen concentration in the matrix pore water
 k — matrix saturated hydraulic conductivity
 R — grout reductive capacity
 x — coordinate in the direction perpendicular to the fracture

Results

The results of example calculations are shown in Figure 1, which illustrate the calculated oxygen concentrations versus depth along the fracture at different times for several different combinations of fracture aperture, fracture spacing, and fracture hydraulic conductivity (parameter values listed in Table 1). These modeling cases include both open fractures and wide fractures filled with sediment. The results indicate that for a wide range of hydrological conditions, oxygen concentrations in through-going vertical fractures in grouted subsurface systems for radioactive waste disposal are maintained at or near the levels of the surrounding soil by a combination of gas-phase diffusion and liquid-phase advection. This is because gas-phase oxygen diffusion in the fracture is fast compared with the rates of diffusive loss to the porous grout matrix. In other words, grout oxidation is controlled by the diffusion rate in the grout. For the long time frames of interest in performance assessments, transport processes in the fractures can easily resupply the fractures with oxygen from the surrounding soil. Because diffusion in the pore space of intact grout material is the rate-limiting process for grout oxidation, detailed process-level models coupling fracture transport processes with matrix diffusion and chemical reactions can be avoided. Simpler models coupling oxygen diffusion and grout oxidation reaction within the matrix can be used instead, with fractures representing internal boundary conditions with specified oxygen concentrations. The conference poster will compare the results of these simpler models with those of the numerical model and also present sensitivity analyses to assess the relative importance of the diffusion coefficient, grout reductive capacity, and fracture spacing to rate of grout oxidation.

References

Atkins, M. and Glasser, F. P. 1992. Application of Portland Cement-Based Materials to Radioactive Waste Immobilization. Waste Management 12, 105-131.

Kaplan, D. I. and Hang, T. 2007. Estimated Duration of the Subsurface Reduction Environment Produced by the Saltstone Disposal Facility on the Savannah River Site. WSRC-STI-2007-00046, Revision 0. Westinghouse Savannah River Company, Aiken, SC.

Kaplan, D. I., Hang, T., and Aleman, S. E. 2005. Estimated Duration of the Reduction Capacity Within a High-Level Waste Tank (U). WSRC-RP-2005-01674, Revision 0. Westinghouse Savannah River Company, Aiken, SC.

This abstract is an independent product of the Center for Nuclear Waste Regulatory Analyses and does not necessarily reflect the view or regulatory position of the U.S. Nuclear Regulatory Commission.

Table 1. Model cases considered when assessing oxygen content in fractures (see Figure 1)					
Modeling Case	Aperture (mm)	Spacing (m)	Fracture Hydraulic Conductivity Model	Fracture Porosity (-)	Calculated Saturation in Fracture
A	0.3	0.3	Cubic law $\eta=0.1$	1.0	0.15
B	0.1	0.3	Cubic law $\eta=0.1$	1.0	0.28
C	0.1	1.0	Cubic law $\eta=0.1$	1.0	0.43
D	0.1	2.0	Cubic law $\eta=0.1$	1.0	0.57
E	0.1	5.0	Cubic law $\eta=0.1$	1.0	0.87
F	10	5.0	Sediment filled, $k_f = 10^{-4}$ m/s	0.25	0.31
G	10	20	Sediment filled, $k_f = 10^{-4}$ m/s	0.25	0.58
H	10	5	Sediment filled, $k_f = 10^{-5}$ m/s	0.25	0.80
The following grout parameters taken from Kaplan, et al. (2005) are also used in the model: $D_e = 1.58 \times 10^{-5}$ m ² /yr; $R_0 = 49.8$ meq e ⁻ /kg; $\rho_g = 1,700$ kg/m ³ ; $\phi = 0.46$; and $C_0 = 1.0 \times 10^3$ meq e ⁻ /m ³ .					

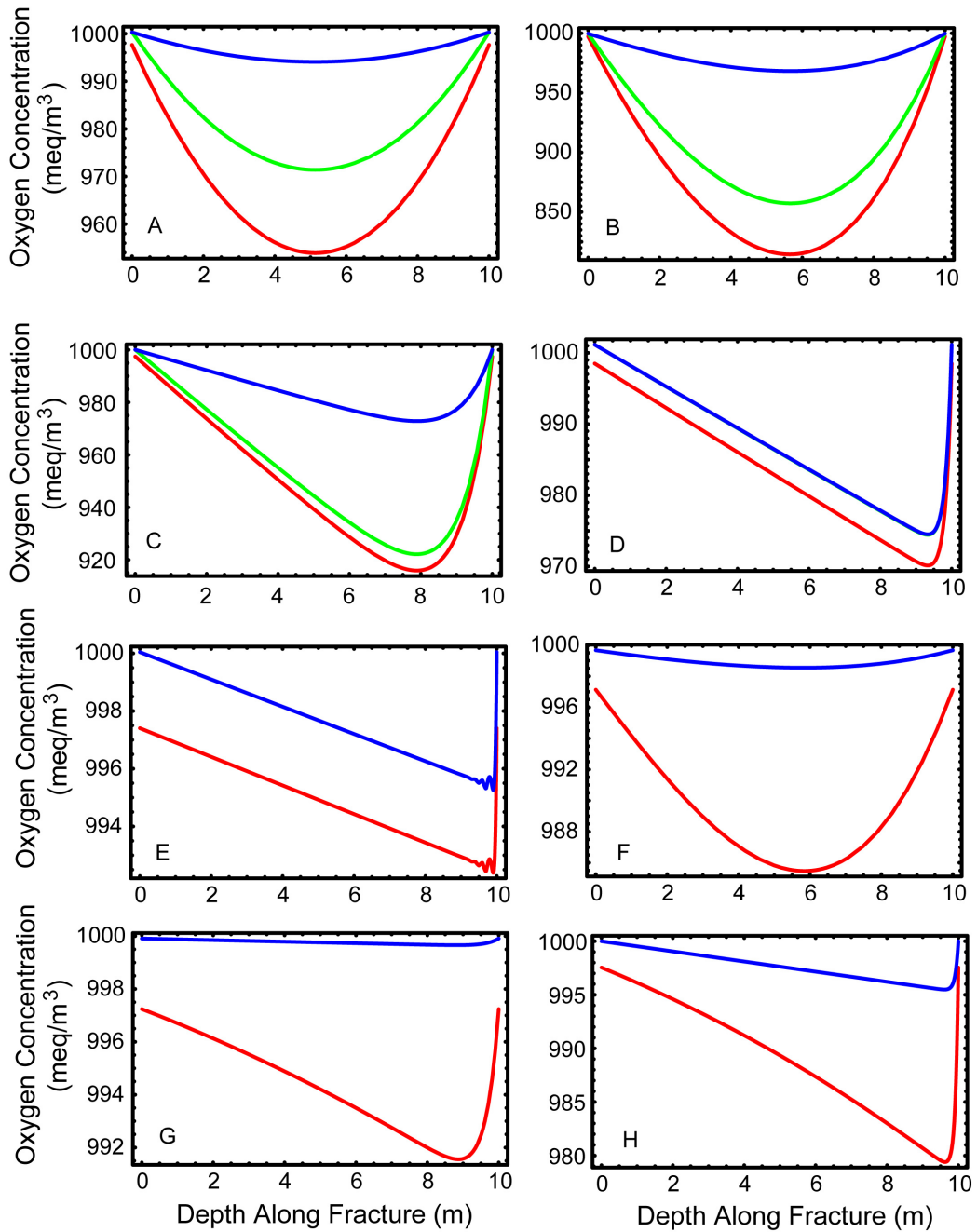


Figure 1. Calculated oxygen concentration in through-going vertical fractures versus depth assuming a coupled advection/dispersion and matrix diffusion model. The red, green, and blue curves are the results after 5, 10, and 100 years, respectively. The green curves are hidden behind the blue curves in Figures 1(d) and 1(e). The key point is that the oxygen concentration in the fracture stays very close to that of the surrounding soil ($1.0 \times 10^3 \text{ meq e}^-/\text{m}^3$). Table 1 lists the parameters for the different cases.