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**The movement of a redox front
downstream from a repository for
nuclear waste**

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THE MOVEMENT OF A REDOX FRONT DOWNSTREAM FROM
A REPOSITORY FOR NUCLEAR WASTE

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THE MOVEMENT OF A REDOX FRONT DOWNSTREAM FROM A
REPOSITORY FOR NUCLEAR WASTE.

- 1 Summary
 - 2 Introduction
 - 3 Conceptual model
 - 4 Mathematical model
 - 5 Application to radiolysis in a nuclear waste repository
 - 6 Discussion and conclusions
Tables and figures
 - 7 Notation
 - 8 References
- Appendix

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1. SUMMARY

In a final repository for nuclear waste it is envisaged that the waste will eventually come in contact with water. When this happens the α -radiation will radiolyse water. The main products will be hydrogen and hydrogen peroxide. The hydrogen is fairly nonreactive and will be transported away by diffusion and convection. The hydrogen peroxide is a strong oxidizing agent and will oxidize any ferrous iron it encounters to ferric iron. This will change the electrochemical potential of the water in the region, from reducing to oxidizing conditions. In oxidizing waters many of the potentially hazardous radionuclides - actinides and technetium - will have a high solubility and low sorption compared to reducing conditions.

The movement of the redox front along the fissures downstream of a repository has been investigated theoretically. The rock is modelled as having parallel fissures where the water flows. The rock matrix has a connected porosity into which the hydrogen peroxide diffuses and quickly reacts with the minerals containing ferrous iron. The movement of the sharp boundary between oxidizing and reducing conditions in the fissures as well as in the matrix of the rock is described mathematically. A calculated example based on expected flowrate of water, rate of α -radiolysis, fissure spacing and measured values of matrix diffusivity and ferrous iron content of the bedrock is made. The results indicate that under some conditions the redox front may move tens of meters downstream during 10^6 years in fairly good rock, poor in ferrous iron.

It is conceivable that the extension of the redox front is larger as well as shorter because many of the important factors governing the front movement may vary considerably.

2. INTRODUCTION

High Level Waste (HLW) and Spent Fuel (SF) from nuclear power plants will have to be emplaced in some kind of final repository. In the Swedish studies the repository is envisaged to be sited in crystalline rock at a depth of about 500 m. It is assumed that the canisters in which the waste is emplaced will be penetrated at some future date. Water flowing in the bedrock will then come in contact with the radioactive material. The radiation, especially the α -particles will radiolyse the water and form reducing species - hydrogen gas and oxidizing species mainly hydrogen peroxide (Christensen 1982). The hydrogen gas is not very reactive and will diffuse away whereas the hydrogen peroxide (H_2O_2) is a strong oxidizing agent. The H_2O_2 will attack the canister material which is copper and lead for SF and HLW respectively in the Swedish cases. The H_2O_2 will also oxidize ferrous iron in the backfill and will ultimately penetrate to the bedrock and the flowing water in the fissures. The bedrock which consists of granite or gneiss also contains ferrous iron of at least 0.1 - 0.3 % by weight (Allard 1982). This will be oxidized by the hydrogen peroxide as it diffuses into the bedrock.

It is of some interest to determine how far and fast the oxidizing front moves because the sorption properties and solubilities are vastly different for many of the escaping radionuclides in oxidizing and reducing environments. Solubilities are many orders of magnitude smaller and sorption capacities many orders of magnitude larger in a reducing (non oxidized) environment than in an environment where oxidizing conditions prevail. (Allard 1978, 1981)

The clay backfill has a limited amount of ferrous iron and the oxidizing front quickly penetrates this to the bedrock. In an initial period the transport into the bedrock adjacent to the backfill is faster than the transport into the flowing water in the fissures of the bedrock. After some time when the bedrock nearest the backfill is oxidized and the distance for diffusion into the bedrock has increased, the H_2O_2 migration into the flowing water in the fissures will dominate.

The present study concerns itself with the transport of the oxidizing agent - H_2O_2 - in the fissures with flowing water and into the matrix of the rock adjacent to the fissures.

The paper is divided into two parts. The first develops the theoretical basis for the movement of a redox front. The main assumptions here concern the geometry of the flow paths. A very simple geometry is chosen because there is very little detailed knowledge on the actual geometry of fissures in crystalline rock.

In the second part an example is given based on some available data on radiolysis and available ferrous iron in the bedrock.

3. CONCEPTUAL MODEL

The thin fissures in the bedrock are assumed to be of equal width and to have parallel walls. The bedrock is a porous material (Skagius et al 1981) with connected porosity into which the dissolved species in the water may migrate by diffusion but not by flow because of the very low hydraulic conductivity of the bedrock. (Brace 1968, Heard 1979). The rock is assumed to have an initial content of ferrous iron Fe(II) which is the main reducing agent in the system and determines the redox conditions. When ferrous iron is attacked by an oxidizing agent such as H_2O_2 it is oxidized to ferric iron Fe(III) and the redox conditions turn from reducing to oxidizing. The reaction between H_2O_2 and Fe(II) is assumed to be so fast that the two species cannot coexist. When H_2O_2 moves into an area where there is Fe(II), the H_2O_2 will momentarily react with it. As long as there is any Fe(II) in that area no H_2O_2 can penetrate through this region. This leads to the development of a sharp "moving boundary". There will be a sharp line separating the not oxidized region from the oxidized region in the rock. Such sharp lines are frequently seen in fissured rocks exposed to air. The gray (reducing) rock is sharply delineated from a reddish oxidized rock.

The approach taken here is to assume that initially the rock around the fissure is reducing. The water in the portion of the fissure near the backfill with the degraded canister is supplied with H_2O_2 at a known rate. The water flows "downstream" in the fissure and the H_2O_2 moves with the water. Some of the H_2O_2 diffuses into the porous bedrock and reacts with the Fe(II) bound to the rock. Figure 1 shows the movement of the redox front in and near a fissure. The front moving parallel to the rock/backfill interface is due to diffusion from this interface. The front near the fissure is due to diffusion from the fissure surface. The migration due to diffusion from the backfill/rock interface is small compared to the distance the front moves along the fissure after an initial period. See Appendix. In the present treatment only the migration from the fissure surfaces is accounted for.

4. MATHEMATICAL MODEL

Cooper and Liberman (1970) and Neretnieks (1974) treated a similar case. They modelled flow in a fixed bed with spherical particles into which a species moves by diffusion. The species reacts quickly with a stationary component. Neither authors treated dispersion but Neretnieks (1974) included film resistance for transfer from the liquid phase to the particle surface. Neretnieks (1974) also treated slabs into which diffusion occurs from a medium with constant concentration and spheres which are immersed in a bath of finite volume and also a case where a bed of spherical particles moves in countercurrent flow to a moving liquid.

Assuming that the capacities of the pore liquid for holding the diffusing species is negligible compared to the reaction capacity of the solid, the transport can be modelled as a semistationary case with a moving boundary.

For a flat surface we have the rate of transport of the species at the moving boundary

$$\frac{dN}{dt} = - \frac{dc}{dr} \Big|_{r_b} \cdot D_p \epsilon_p \cdot A \quad (1)$$

r_b is the distance from the center of the slab at which the redox front is at a given moment.

The redox front moves with a velocity which depends on the rate of transport of the oxidizing species and the concentration q_o of the stationary reducing agent. The reaction $2Fe^{2+} + H_2O_2 + 2H^+ \rightarrow 2Fe^{3+} + 2H_2O$ gives a stoichiometric factor $f = \frac{1}{2}$.

$$\frac{dr_b}{dt} = \frac{dr_b}{dt} \cdot A \cdot q_o \cdot f \quad (2)$$

combining (1) and (2) gives

$$\frac{dr_b}{dt} = - \frac{dc}{dr} \Big|_{r_b} \cdot \frac{D_p \epsilon_p}{q_o \cdot f} \quad (3)$$

The following initial and boundary conditions are used

$$\text{IC} \quad \left. \begin{array}{l} c = 0 \\ q = q_0 \end{array} \right\} \quad \text{at} \quad t = 0, r < S/2 \quad (4)$$

$$\text{BC 1} \quad c = c_s(t) \quad \text{at} \quad r = S/2 \quad (5)$$

$$\text{BC 2} \quad c = 0 \quad \text{at} \quad r = r_b \quad (6)$$

If the capacity of the pores for holding the adsorbent is neglected, $\frac{dc}{dt} = 0$, then the transport in the pore for $r_b \leq r < S/2$ is described by

$$\frac{\partial^2 c}{\partial r^2} = 0.$$

This can be permitted if the capacity of the solid is much larger than that of the liquid in the pores. This gives

$$\frac{dc}{dr} = \frac{dc}{dr} \Big|_{r_b} = \frac{c_s(t) - 0}{S/2 - r_b} \quad (7)$$

Inserting (7) in (3) gives

$$\frac{dr_b}{dt} = - \frac{c_s}{S/2 - r_b} \cdot \frac{D_p \epsilon_p}{q_0 \cdot f} \quad (8)$$

Integration of (8) from $r = S/2$ to $r = r_b$ and with $c = c_s = \text{const}$ gives

$$\frac{D_p \epsilon_p c_s}{(S/2 - r_b)^2 q_0 \cdot f} \cdot t = \frac{1}{2} \quad (9)$$

for $0 \leq r_b < S/2$ and for times less than full penetration.

For penetration to the center of the slab $r_b = 0$ i.e. to a depth of $S/2$, we have

$$t = \frac{1}{2} \cdot \frac{(S/2)^2 \cdot q_0 \cdot f}{D_p \epsilon_p c_s} \quad (10)$$

It is interesting to note that it takes a finite time to fully penetrate a slab of a given thickness.

4.1 Flow in a fissure

In the case of flow in a fissure the surface of the fissure is subject to a concentration which is dependent on the location along the fissure as well as upon time.

A differential mass balance along a fissure neglecting dispersion is given by

$$\frac{\partial c}{\partial t} + v \frac{\partial c}{\partial z} + \frac{1}{m} \frac{\partial q}{\partial t} = 0 \quad (11)$$

This can be transformed into

$$\frac{\partial C}{\partial Z} + \frac{\partial Q}{\partial \theta} = 0 \quad (12)$$

by introducing

$$\theta = \left(t - \frac{z}{v}\right) \frac{D_p \epsilon_p c_o}{(s/2)^2 q_o \cdot f} = \left(t - \frac{z}{v}\right) D^* \quad (13)$$

where

$$D^* = \frac{D_p \epsilon_p c_o}{(s/2)^2 q_o \cdot f} \quad (14)$$

$$Z = \frac{D_p \epsilon_p z}{(s/2)^2 m v} \quad (15)$$

$$C = c/c_o \quad (16)$$

$$Q = q/q_o = 1 - r_b/(s/2) \quad (17)$$

Equation 8 transforms into

$$\frac{\partial Q}{\partial t} = D^* \cdot \frac{c_s}{c_o} \cdot \frac{1}{Q} \quad (18)$$

Introducing the concept of film resistance

$$\frac{dN}{dt} = k_F (c - c_s) \cdot A \quad (19)$$

and eliminating c_s from e.g. (18) and (19) by aid of (2) and (17) we obtain

$$\frac{\partial Q}{\partial t} = \frac{D^*}{Q + 2/Bi} \cdot C \quad (20)$$

$$\text{where } Bi = \frac{k_F S}{D_p \epsilon_p} \quad (21)$$

Equation (20) and the definition of θ equation (13) gives

$$\frac{\partial Q}{\partial \theta} = \frac{C}{Q + 2/Bi} \quad (22)$$

Equations (22) and (12) using the conditions (4), and $C(\theta, 0) = 1$ are solved by the same method as used in Cooper and Liberman (1970) to give

$$Z = -\frac{2}{Bi} \cdot \ln \frac{Q(\theta, Z)}{Q(\theta, 0)} - Q(\theta, Z) + Q(\theta, 0) \quad (23)$$

(23) is valid for $\theta \leq 1/2 + 2/Bi$.

For $\theta > 1/2 + 2/Bi$ we have

$$\frac{2}{Bi} \ln Q(\theta, Z) + Q(\theta, Z) = \theta - Z + 1/2 \quad (24)$$

$Q(\theta, 0)$ is obtained from equation (9), (17) and (13) for $Z = 0$ giving

$$Q(\theta, 0) = \sqrt{2\theta}/_{Z=0} = 0 = \sqrt{D^*t} \quad (25)$$

When there is no film resistance $Bi \rightarrow \infty$ and the equations simplify to

$$Q(\theta, Z) = \sqrt{D^*t} - Z \quad \text{for } \theta \leq \frac{1}{2} \quad (26)$$

and

$$Q(\theta, Z) = \theta - Z + 1/2 \quad \text{for } \theta > \frac{1}{2} \quad (27)$$

Equations 24 and 27 show the constant pattern case meaning that the shape of the breakthrough front does not change as it moves in the direction of the flow. The velocity of the front is directly obtained from equation (27) for the case of no film resistance by following the movement of the midpoint of the breakthrough curve $Q(\theta, Z) = \frac{1}{2}$.

We then get $\theta = Z$ which after simplification gives

$$V_{\text{front}} = \frac{z}{t} = \frac{v}{1 + \frac{q_o \cdot f}{c_o m}} \quad (28)$$

5. APPLICATION TO RADIOLYSIS IN A NUCLEAR WASTE REPOSITORY

Christensen (1982) calculated the rate of H_2O_2 production from a canister with 1.4 tonnes spent fuel. See table 1. During the period 10^5 years to 10^6 years, $1.58 \cdot 10^4$ moles of H_2O_2 are produced. An average figure of 0.017 moles/yr are produced. The water flow rate which carries this production is 0.45 l/yr in a case calculated by Neretnieks (1982). The water flows in 5 fissures with a velocity of $3 \cdot 10^{-8}$ m/s (~ 1 m/yr). The fissure width $\delta = 0.1$ mm and the fissure spacing $S = 1$ m is used. $q_o = 100$ moles Fe(II)/m³ of rock (Allard 1982). From the above data we obtain $m = \epsilon/(1 - \epsilon) = \delta/(s - \delta) = 10^{-4}$, $c_o = 0.037$ moles H_2O_2 /l.

Skagius et al (1981) measured diffusivities in crystalline rock using tritiated water. $D_p \epsilon_p$ was found to be about 10^{-13} m²/s. This value is taken as the diffusivity for H_2O_2 . The Biot number $Bi \rightarrow \infty$ in the thin fissures because the time to equilibrate the water in the fissure by diffusion is very small compared to the time to equilibrate concentration in the slab of thickness $S/2$.

The above data let us calculate the initial time period to develop a constant pattern $\theta = 1/2$, and the velocity of the moving front.

$$\theta = \frac{1}{2} = \frac{D_p \epsilon_p c_o}{(S/2)^2 q_o \cdot f} t \text{ gives}$$

$$t = 0.34 \cdot 10^{13} \text{ s } (\sim 1.1 \cdot 10^5 \text{ years})$$

The edge of the front has reached $Z_e = 1$ (when $Q = 0$ and $\theta = \frac{1}{2}$) (eq. 26). z_e is obtained from eq. 15 to be

$$z_e = 7.5 \text{ m}$$

After this the midpoint of the front where $C = 0.5$ moves with a velocity

$$V_{\text{front}} = \frac{v}{1 + \frac{q_o \cdot f}{c_o m}} = 2.04 \cdot 10^{-12} \text{ m/s } (6.5 \cdot 10^{-5} \text{ m/yr})$$

After $0.9 \cdot 10^6$ years after canister breakthrough the front has reached 58 m.

Figure 2 shows how the redox front is moving in the fissured rock.

6. DISCUSSION AND CONCLUSIONS

There are at present many factors which may strongly influence the movement of the redox front, which are not well known. Fissure spacing has a strong impact on the movement of the front as it enters the equations with a power of two. The effective diffusivity D_p , ϵ_p and available iron content of the bedrock are also factors which may vary considerably. The water velocity and fissure width influence the results via the product $\epsilon \cdot v = u_0$ where u_0 is the water flux in the bedrock. This may also vary considerably but the individual velocity and porosity variations do not influence the results separately.

The computed rate of α -radiolysis is based on the assumption that all the gas space in the fuel rods is filled with water and that it is evenly distributed around the fuel pellets in the thin layer 0.03 mm thick. If and when the canister and the fuel cladding of the rods have corroded so that water may access the fuel pellets, there will probably be a considerable reduction of the pore space because the corrosion products have a lower density and thus will invade available pore space. This will considerably decrease the radiolysis.

In the calculated example the redox front is assumed to move in a thin band downstream without transverse dispersion. The latter will in reality widen and shorten the front. The amount of rock oxidized is directly dependent on the amount of ferrous iron in the bedrock. A very low value 0.2 % was used in the example. Furthermore the canister material, if of metal, will undoubtedly be oxidized by parts of the hydrogen peroxide formed and thus further limit the extent of the radiolysis front.

The redox front may under some circumstances move outside of the backfill and some way downstream from the canister. The actual distance will depend on many factors such as actual rate of radiolysis and ferrous iron content of the bedrock. These factors are being investigated further.

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Time years	Production of H_2O_2 mol/yr	Time interval yrs	Produced H_2O_2 in time interval moles
40	5.1		
		60	290
100	4.6		
		200	780
300	3.2		
		300	750
600	1.8		
		400	620
1000	1.31		
		9000	7400
10^4	0.34		
		$9 \cdot 10^4$	16200
10^5	0.025		
		$9 \cdot 10^5$	15800
10^6	0.009		
		Totally produced during 10^6 yrs	41400

Table 1. Production of H_2O_2 by α -radiolysis from 1.4 tonnes of spent fuel of different ages. Christensen (1982).

The computations are based on the following assumptions. The zirkalloy cladding of all 498 fuel rods is penetrated. All fuel pellets with a total surface area of about 144 m^2 are wetted with a 0.03 mm thick water film. This is the penetration depth of the α -particles. More than 40 different reactions including reactions with solvated ferrous and ferric species are accounted for in the computations of the radiolysis. The computations have been performed at Riso Laboratory in Denmark with a program which previously has been able to well model radiolysis experiments.

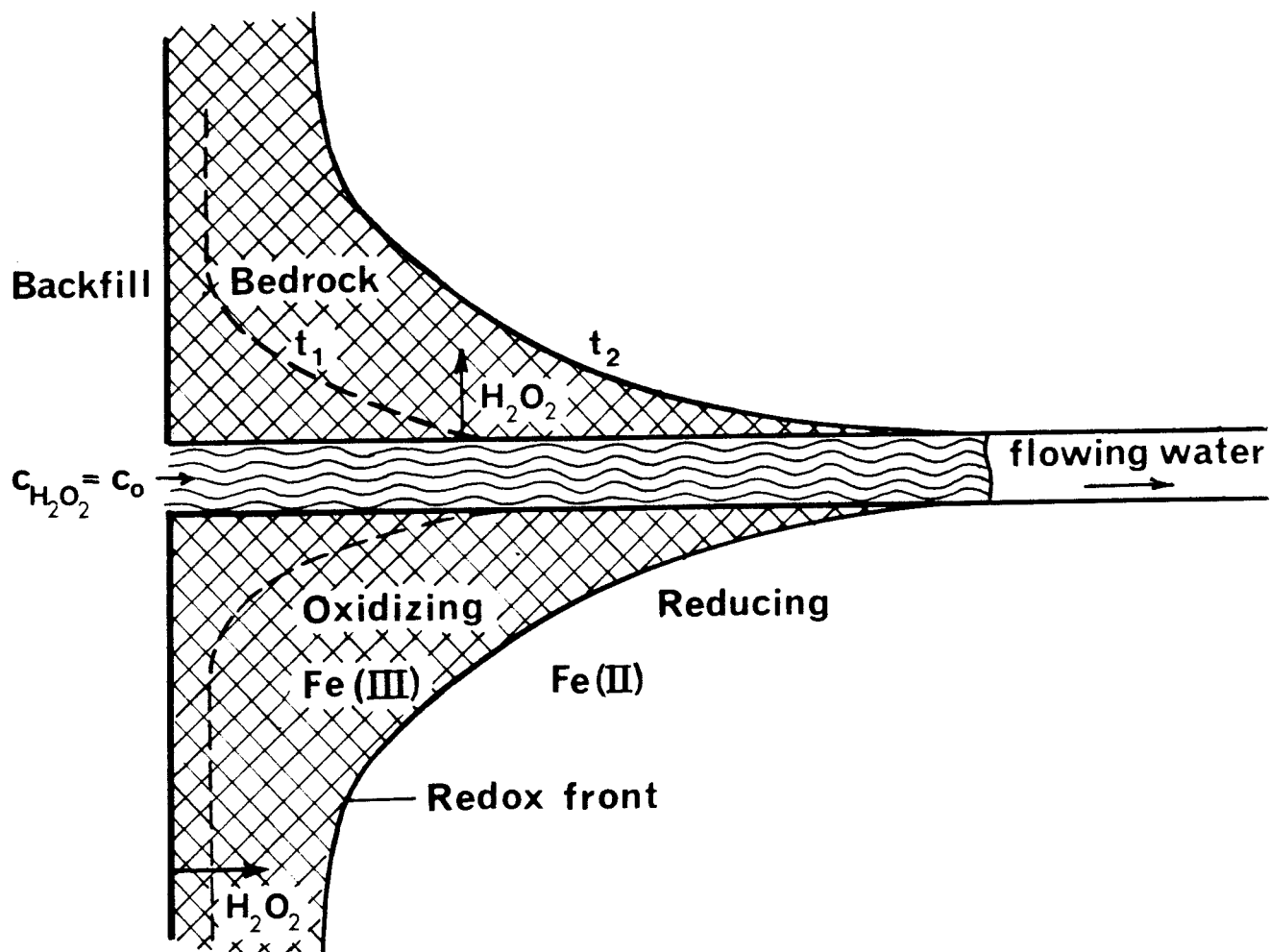


Figure 1. The movement of a redox front along a fissure. Dashed line shows position of the front at time t_1 and full line shows position at time t_2 .

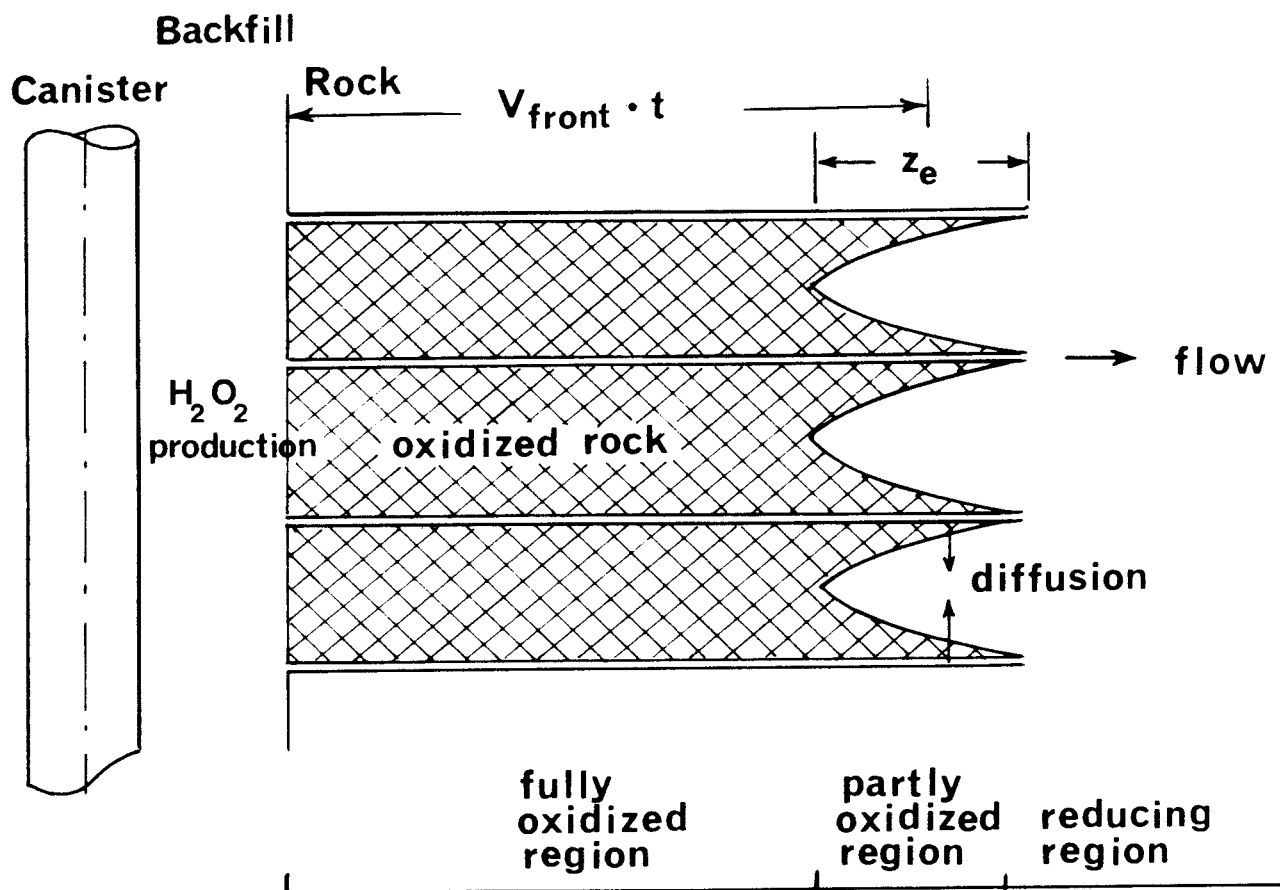


Figure 2 shows the region influenced by the oxidizing species and the movement of the redox front.

7. NOTATION

A	surface for mass transfer	m^2
Bi	Biot number $k_F S / D_p \epsilon_p$	-
c	concentration of oxidant in liquid	mol/m^3
c_o	concentration at inlet of fissure	mol/m^3
c_s	concentration at surface of fissure	mol/m^3
C	dimensionless concentration c/c_o	-
D_p	pore diffusivity	m^2/s
D^*	$D_p \epsilon_p c_o / q_o (S/2)^2$	s^{-1}
k_F	mass transfer coefficient	m/s
f	stoichiometric factor for oxidation reaction	-
m	$\epsilon / (1 - \epsilon)$	-
N	mass of species	mol
q	concentration of reducing species in solid	mol/m^3
q_o	original concentration in solid	mol/m^3
Q	dimensionless concentration q/q_o	-
r	distance from center of slab	m
r_b	location of redox front in slab	m
S	thickness of slab	m
t	time	s
u_o	flux of water in bedrock	$m^3/m^2 \cdot s$
U_e	equivalent flowrate of water to transport oxidant	$m^3/s(1/yr)$
v	velocity of water in fissure	m/s
V_{front}	velocity of redox front	m/s
z	distance along fissure	m
z_e	distance along fissure of leading edge of redox front	m
Z	dimensionless distance	-
δ	fissure width	m
ϵ	flow porosity of bedrock	-
ϵ_p	diffusional porosity of rock	-
θ	dimensionless time	-

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APPENDIX

A simplified comparison of the transport rate of the oxidant by convection with the flowing water in the fissures and the transport due to diffusion into the rock adjacent to the backfill is made in the following way: For a given driving force (concentration difference) the transport of a species into the bedrock can be approximated by

$$\frac{dN}{dt} = - D_p \epsilon_p \cdot A \cdot \frac{\Delta c}{\Delta x} \quad \text{A 1.1}$$

where Δx is the thickness of the oxidized front of the bedrock surrounding the backfill. The transport to the flowing water can be expressed in terms of an equivalent water flow rate past the canister U_{eq} and its increase in concentration (Neretnieks 1982)

$$\frac{dN}{dt} = U_{eq} \cdot \Delta c \quad \text{A 1.2}$$

The thickness of the oxidized front Δx when the transport rate by the two mechanisms is equal is obtained from the above equations by eliminating dN/dt .

$$\Delta x = \frac{D_p \epsilon_p A}{U_{eq}}$$

The following data are used in evaluating Δx

$$D_{p,p} \varepsilon = 10^{-13} \text{ m}^2/\text{s} \quad (\text{Skagius et al 1982})$$

$$U_{eq} = 0.45 \text{ 1/yr} \quad (\text{Neretnieks 1982})$$

A is approximated by the cylindrical surface of the emplacement hole. It has a diameter of 1.5 m and an effective length of about 6 m. $A = 28 \text{ m}^2$. With these data we obtain $\Delta x = 0.19 \text{ m}$.

This means that when the oxidizing front has penetrated more than 0.19 m into the rock from the backfill/rock interface the transport to the flowing water in the fissures will dominate. The amount of H_2O_2 needed to oxidize this thickness of rock is $A \cdot \Delta x \cdot q_0 \cdot f = 28 \cdot 0.19 = 100 \cdot \frac{1}{2} = 266 \text{ moles}$. This is a small fraction of the H_2O_2 produced over the time interval of interest.

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