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AN APPROACH FOR EVALUATING THE GENERAL AND LOCALISED CORROSION OF CARBON STEEL CONTAINERS FOR NUCLEAR WASTE DISPOSAL

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ABSTRACT

The paper considers the long term corrosion of carbon steel containers
for heat generating nuclear waste in a granitic repository. Under such
conditions carbon steel may exhibit general, localised or passive corrosion
behaviour depending on the exact composition and redox potential of the
groundwater contacting the containers; localised corrosion being of most
concern because it has the fastest propagation rate. It is well
established, however, that such localised corrosion is only possible when
the environment is sufficiently oxidising to maintain a positive potential
gradient between the cathodic surface and the corrosion sites, which
requires that species with oxidising potentials greater than water need to
be present. This fact provides a basis for estimating the periods during
which containers may be subject to localised and subsequently to general
corrosion, and hence for making an overall assessment of the metal allowance
required for a specified container life. A model for the diffusion
transport of oxygen has been developed, and a sensitivity analysis has shown
that the period of possible localised attack is strongly dependent on the
passive film leakage current, the radiation dose rate and the oxygen
diffusion coefficient.

INTRODUCTION

Conceptual designs for the disposal of heat generating nuclear waste in
geological formations generally include the provision of a container, which
is intended to isolate the waste for the order of 500 to 1000 years. The use
of carbon steel for the production of such containers has the advantage of
avoiding the difficult task of demonstrating long term corrosion
resistance [1]. It does, however, require knowledge of the type of
corrosion which will occur, and its rate of propagation, so that an
appropriate allowance can be specified. In previous publications the forms
of corrosion considered possible in granitic repository environments were
listed as; general corrosion, localised attack, stress corrosion cracking,
hydrogen embrittlement and microbial induced corrosion [1,2]. On the basis
of more recent work it has been concluded that stress corrosion cracking
should be avoided by stress relieving the containers [3,4], and that
hydrogen embrittlement can also be avoided providing the closure weld
heat-affected- zone is softened by heat treatment [3,5]. Given this
position, attention has focussed on general and localised corrosion with a
combination of mathematical modelling and experimental studies aimed at
evaluating the rates of these processes as a function of disposal time
[6,7].

Because localised corrosion propagates more rapidly than general attack
in granitic environments, it will dominate estimation of the corrosion
allowance. Therefore, if the corrosion allowance is not to be over
conservative, it is important to know for what fraction of the container
life localised corrosion may occur. This paper describes a mechanistically
based approach for making such an estimate, and a sensitivity analysis to
establish which factors control the period of localised attack.

MECHANISM OF LOCALISED CORROSION

First of all it is important to make clear what is meant by localised corrosion because the corrosion literature contains many examples where this term is used to describe the points of maximum penetration on what is essentially an uneven general corrosion front. In the present instance localised corrosion refers to the isolated penetrations which may develop as pits or at crevices on otherwise passive metal surfaces.

This form of corrosion in carbon steel has been studied extensively by Pourbaix [8], who determined the chemical and electrochemical conditions associated with pitting attack using electrochemical polarisation techniques, and depicted these on potential-pH diagrams. We have made similar measurements in Na_2CO_3-$NaHCO_3$-$NaCl$ solutions at 50°C [3,6], and the results from 0.01M $Co_3^=$-HCO_3^- solution are superimposed on the theoretical potential-pH diagram for the Fe-H_2O 0.01M $CO_3^=$ system [9] in figure 1(a). The important points to note are that pitting occurs at a potential somewhat above the passivation potential, that this potential increases with pH, and that pitting does not occur at low pH's because the steel cannot passivate.

The experimentally determined boundary between stability and general corrosion runs parallel to the theoretical equilibrium for the Fe-$FeCO_3$ reaction, but about 50 mV more positive. This represents a reasonably good correlation bearing in mind that the experimental results are for 50°C and the theoretical values for 25°C. Furthermore, the experimental boundary is based on the potentials of zero net current on a mixed reaction surface, which would be expected to be higher than the thermodynamic equilibrium for the anodic reaction alone because of the cathodic current component arising from water reduction. In contrast, there is no clear link between the empirical general corrosion-passivation boundary and any of the thermodynamic equilibria, although it does appear to have a similar pH relationship as the Fe-Fe_3O_4 and Fe_3O_4-Fe_2O_3 equilibria. No analyses have been performed in the present study, but the literature contains evidence that the passive film is Fe_3O_4 [10,11].

The mechanism whereby localised corrosion develops on passive surfaces of metals such as carbon steel and stainless steel has been the subject of considerable research [12]. Even so there continues to be much debate on the initiation process, although a concensus has been achieved on how the pits remain stable and propagate, the essential features of which are as follows:

(a) At the end of the initiation stage the environment at the localised corrosion site has become sufficiently acidic to cause the metal to corrode actively. This results in the development of a potential gradient between the active sites and the remaining passive surface.

(b) Because the passive surface is more noble than the localised active sites it supplies part of the cathodic current required to sustain dissolution in these sites. As a result anions such as OH^-, Cl^- and $SO_4^=$ migrate into the active sites.

(c) The migration of Cl^- and $SO_4^=$ into the active sites results in the formation of $FeCl_2$ and $FeSO_4$ which hydrolyse to produce HCl and H_2SO_4. This maintains the acidity of the sites, thus sustaining the localised corrosion process.

This mechanism has been supported by direct measurements of the pH in localised corrosion sites, which in the case of carbon steel is in the region of 3-4 [13,14,15]; in good agreement with theoretical estimates based
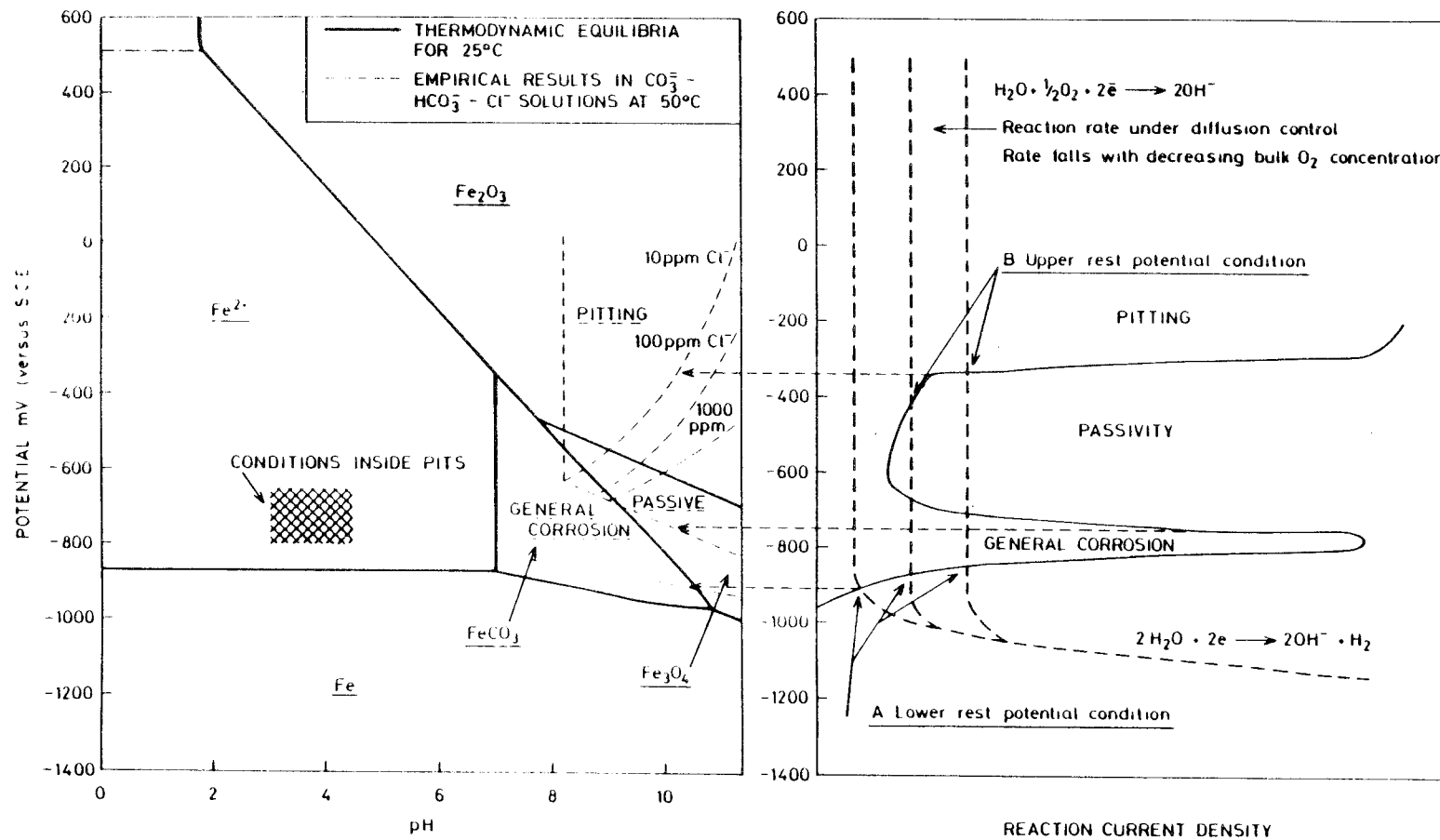
Figure 1   (a)   Potential-pH diagram comparing the thermodynamic equilibria of the $Fe-H_2O$ $0.01M^=CO_3$ system [i.e. for a metal ion concentration of 10^{-6} M litre^{-1}] with experimentally determined conditions for general corrosion, passivity and localised corrosion in $0.01M$ $CO_3^=-HCO_3^-$ solutions.

(b)   Schematic anodic and cathodic polarisation curves illustrating how the rest potential of carbon steel is influenced by oxygen supply in $0.01M$ $CO_3^=$ + 10 ppm Cl^- solution.

on the hydrolysis of $FeCl_2$ [15]. The electrode potential within localised corrosion sites has also been measured and has been shown to be more active than the surrounding passive surface [15]. Using the data from these measurements the approximate conditions within a localised corrosion site have been indicated by a cross-hatched zone in figure 1(a). This highlights the potential difference existing between the active areas and the bulk surface when carbon steel is subject to localised attack.

In practice the open surface potential and pH conditions associated with localised corrosion change depending on the relative concentrations of passivating (e.g. HCO_3^-, $CO_3^=$) and aggressive (e.g. Cl^-, $SO_4^=$) ions. This means that the possibility of carbon steel containers being subject to localised attack can only be assessed after the repository environment has been fully characterised. However, the mechanism for the propagation of localised corrosion outlined above indicates one fundamental limitation to the occurrence of such attack, which is that it will cease when the external surface is no longer cathodic (i.e. positive) to the active sites. Without a supply of cathodic current from the external surface all the cathodic reaction required to sustain dissolution will have to occur within the active site. This will result in the consumption of acidity, which will no longer be regenerated by hydrolysis, because there is no potential gradient to cause the migration of Cl^- or $SO_4^=$ ions into the active sites. This forms a basis for estimating the maximum period during which the containers may be subject to localised attack.

The factors which determine the overall potential of a carbon steel surface are illustrated by the reaction current versus potential diagrams in figure 1(b). The solid line indicates the rate of the anodic (i.e. corrosion) reaction which, with increasing potential, goes through transitions from general corrosion to passivity to localised corrosion. The broken line depicting the cathodic reaction rate is based on the assumption that the main cathodic reactants under repository conditions will be oxygen and water. The electrode potential of the metal is determined by the charge neutrality requirement that the algebraic sum of the anodic and cathodic reaction currents should be zero. Figure 1(b) shows that at low and zero oxygen levels this condition is only met at the points marked A where the metal is subject to general attack. However, when oxygen is present the rest potential may be at points A or B depending on the rate of the oxygen reduction reaction. The minimum requirement for maintaining the steel at a passive or possibly pitting potential (i.e. B) is that the oxygen reduction rate is at least equal to the rate of metal dissolution through the passive film.

Comparison of rest potentials A and B with the electrode potential in a localised corrosion sited (figure 1(a)) shows that A is more negative and B more positive. This leads to the not unreasonable conclusion that carbon steel can only be subject to localised corrosion when in the passive state. The above conclusion has been investigated experimentally by a series of tests in 0.1M $NaHCO_3$ + 1000 ppm Cl^- solution (pH 8.3) at 90°C using a carbon steel of composition 0.17% C, 0.75% Mn, 0.06% Si, 0.02% S and 0.02% P. Previous work [3] has shown that this environment causes pitting at 0, -200 and -400 mV (SCE), and these additional experiments were undertaken to determine the lower potential limit for pitting attack. Results for tests lasting 600h are illustrated in figure 2 which indicates a limiting potential of -600 mV, which is approximately 150 mV above the passivation potential for this solution.

It is worth mentioning one important exception to the above analysis which is the localised corrosion of boiler tube steels in highly deoxygenated water. The key difference here is that the high heat flux causes the concentration of corrosive salts in pores and fissures in the protective Fe_3O_4 layer. The heat flux from heat emitting waste containers

is murh lower than for boiler tubes and therefore this mechanism should not be operative under repository conditions.

With this exception it is clear from the above that the maximum period for localised corrosion can be estimated by calculating the rate of transport of oxygen to the containers as a function of time. The minimum requirement for passivity, and hence possible localised attack, is that the oxygen reduction rate should be at least equal to the passive metal dissolution rate.
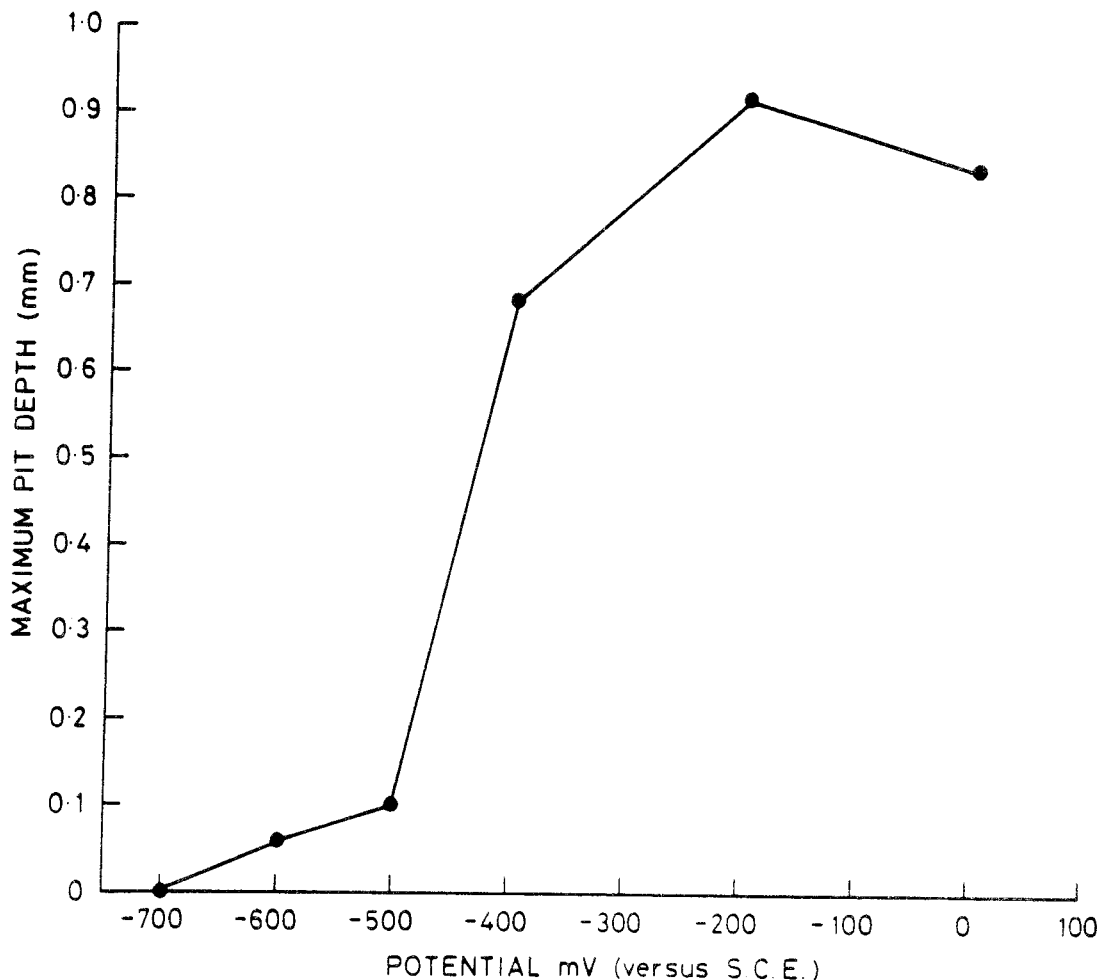

Figure 2   Variation of the maximum depth of pit penetration with electrode potential for experiments in 0.1M $NaHCO_3$ + 1000 ppm Cl^- solution at 90°C.

MODEL FOR OXYGEN TRANSPORT

Designs of repositories for the disposal of heat emitting wastes in granitic rock formations are typified by that developed in Sweden and described in KBS-3 [16]. Essential features are that the containers will be placed in bore-holes drilled in the floors of horizontal tunnels, and that the bore holes and tunnels will be backfilled with a low permeability material such as bentonite. In such a repository there will be three sources of oxygen or similar oxidising agents:

(a)   The air trapped in the pore space of the back-filling material.

(b)   Dissolved oxygen in the groundwater permeating the repository.

(c)   Oxidising species produced by γ-radiolysis of the groundwater.

Although the flow of water through granite may be variable because of channelling effects through cracks and fissures, the bentonite backfill will have the effect of smoothing the flow of water to the containers. It is therefore reasonable to assess the significance of oxygen transport by water flow (i.e. (b)) using an average value for water movement through granite which has been estimated to be $\sim$ 0.2 litres m^{-2} yr^{-1} [17]. Assuming this groundwater is equilibrated with air at 1 atmosphere pressure (i.e. dissolved O_2 $\sim$ 8 ppm) the rate of supply of oxygen to a container of 4 m length and 1 m diameter will be 6400 ppm per year. This is equivalent to a cathodic current of about 2 x 10^{-7} Amps m^{-2} which is small compared to the passive leakage current from carbon steel (see later). Furthermore, because of the low permeability of the backfill, convective transport should not occur within the bore-hole, despite the heating effect of the waste [17]. Therefore calculations of the rate of transport of oxygen to the container surface need only take account of (a) and (c).

We have developed a one-dimensional model for oxygen transport under such repository conditions in which the diffusion path length 'a' is taken as the distance between the top of the container and the floor of the repository tunnel. In the model the concentration of oxygen c at any distance x from the container at time t is governed by

$$\alpha \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} + \alpha \, G \, R_o \exp \left[\frac{-t}{\tau} \right] \exp \left[\frac{-x}{L} \right] \quad \text{for } 0 \leq x \leq a \quad (1)$$

where D is the intrinsic diffusion coefficient of oxygen in the backfill, R_o the initial γ-radiation dose at the surface of the containers, τ the average disintegration constant equivalent to a mean half-life of 30 years, L the relaxation length of the backfill (0.16 m) and G the number of molecules of oxidising species produced by a 100 eV radiation dose to the groundwater. Experimental studies [3] of the effect of radiation on the rate of corrosion of carbon steel have yielded a cumulative G value, representing the maximum rate of production of all radiolytic species which are oxidising towards carbon steel, of 2.8, and this value has been used in the calculations. Finally α is a capacity factor which reflects the physical retention of the diffusing species in the porosity of the backfill and bulk chemical retention by equilibrium sorbtion. In this case α is taken to be equal to the porosity of the backfill (i.e. 0.33) [17].

The boundary conditions for the solution of (1) were taken as follows:

(a)   At time zero, which is taken as the time at which the repository becomes flooded, an even distribution of oxygen exists throughout the backfill pore structure. This initial concentration Co is taken to be 8.9 x 10^{-3} moles dm^{-3}, which assumes all the oxygen originally present in the pores as air, dissolves in the water which subsequently saturates the backfill.

(b)   At the container surface the flux of oxygen remains equal to the passive dissolution current (i.e. leakage current) of the carbon steel, until the concentration gradient is no longer sufficient to sustain this situation. This is described mathematically by

$$\frac{I}{4F} \leq D \frac{\partial C(o,t)}{\partial x} \quad (2)$$

(c)   The concentration of oxygen at floor of the repository tunnel (i.e.

distance 'a' from the containers) remains constant at Co. This
recognises that there is a much greater volume of backfilling in the
shaft, and that this may have a greater permeability permitting
convective transport.

(d) The model assumes constant temperature conditions through the backfill,
and at the present stage of development the cooling of the repository
is not taken into account.

Equation (1) has been solved with the above boundary conditions using a
Laplace transform method. The transform is inverted numerically using
Talbot's method, which involves integrating the inversion integral along the
steepest descent contour of the function $g(t) = {}^1/t$ [18].

SENSITIVITY ANALYSIS

This model has been used to calculate the time at which the oxygen flux
falls below the minimum needed to maintain passivity as a function of three
key variables; the intrinsic oxygen diffusion coefficient D, the initial
radiation dose rate R_o and the passive leakage current I_{12} The values of D
investigated were 1.2×10^{-10}, 1.2×10^{-11} and 1.2×10^{-12} m^2 sec^{-1}, the
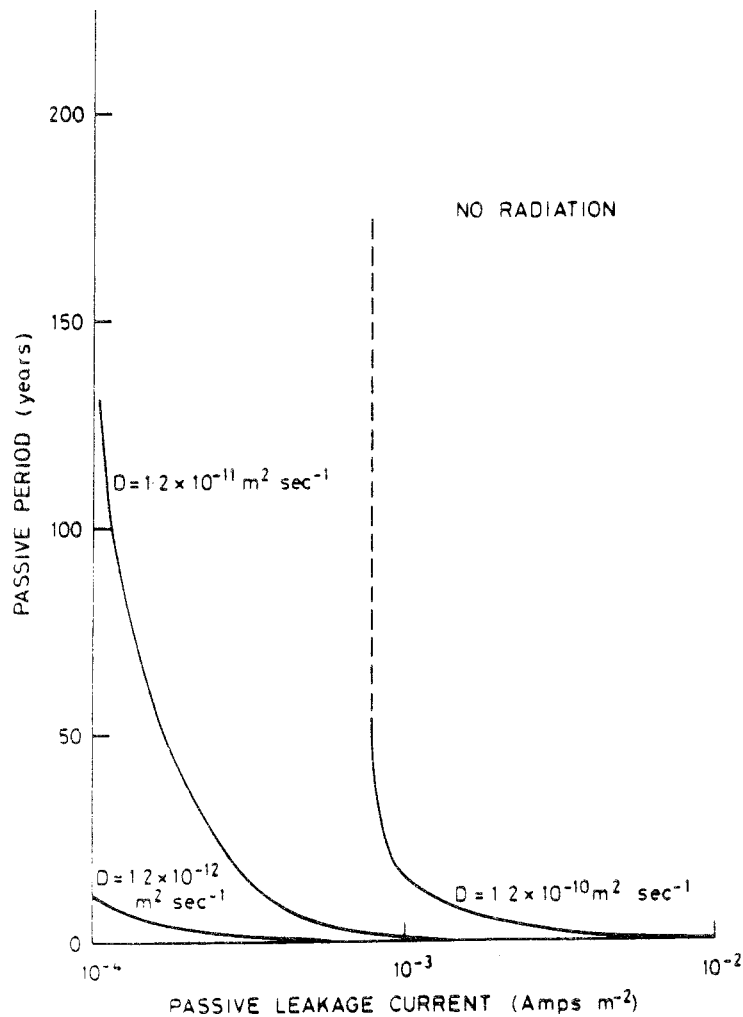

Figure 3   Passivation period as a function of leakage current and oxygen
diffusion coefficient without γ-radiation.

former two being representative of O_2 diffusion coefficients through wet bentonite at 90°C and 25°C respectively. R_o levels investigated were zero, 10^2 and 3.5×10^3 R h^{-1}; the latter two values representing the initial surface dose rates from carbon steel containers with wall thicknesses of approximately 250 and 100 mm, filled with 50 year old vitrified waste. These levels were selected to investigate the behaviour of containers with 180 and 30 mm corrosion allowances, since it has been estimated that a wall thickness of 70 mm will be required from considerations of mechanical strength [19]. A backfill thickness of 0.5 m was used in all the calculations.

Results for the case of a zero radiation dose indicate, as would be expected, that the passivation period of the containers falls with increasing leakage current. This period is also sensitive to the oxygen diffusion coefficient, and it is particularly significant that the calculations indicate that the containers will remain passive indefinitely at leakage currents below $\sim 9 \times 10^{-4}$ A m^{-2} with a D of 1.2×10^{-10} m^2 sec^{-1}. This arises because, under these particular conditions, the steady state oxygen flux is sufficient to balance the passive leakage current. A similar result would have been obtained with the other D values if lower leakage currents had been investigated. Figure 4 shows that the effect of an initial radiation dose rate (i.e. R_o) of 10^2 R h^{-1} is to extend the passive period, particularly under conditions of slow diffusion.
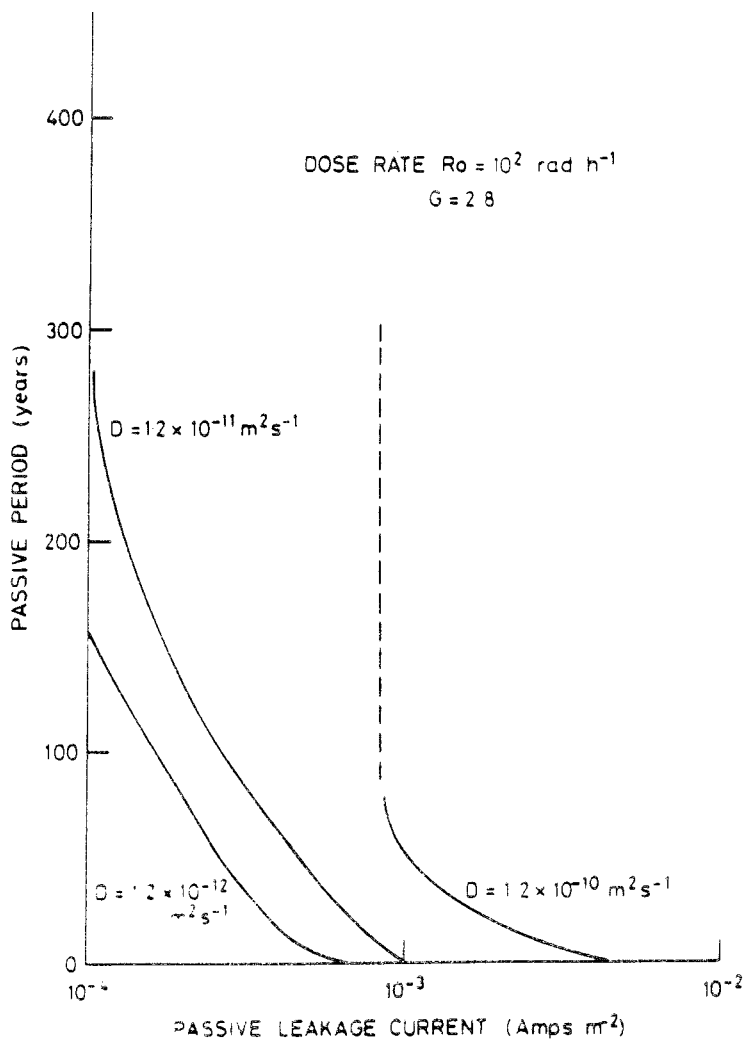

Figure 4   Passivation period as a function of leakage current and oxygen diffusion coefficient with a γ-radiation dose of 10^2 R h^{-1}

More complex behaviour is indicated by the results for a dose rate of 3.5×10^3 R h^{-1} (figure 5). Here lower diffusion coefficients yield longer passivation periods, at least until the steady state oxygen flux with the highest diffusion coefficient approaches the passive leakage current below $\sim 9 \times 10^{-4}$ A m^{-2}. This anomalous behaviour would seem to be attributable to the high rate of production of oxidising radiolysis products, which are able to balance the passive leakage current, because they are slow to diffuse from the container surface. Eventually the rate of radiolysis will decrease so that the system returns to control by the diffusion of oxygen from the repository shaft. Figure 5 shows this change to normal diffusion control for $D = 1.2 \times 10^{-10}$ m^2 sec^{-1} at a passive leakage current of about 9×10^{-4} A m^{-2}, but this change does not occur with the lower D values over the range of currents and times investigated.

The analysis shows that the passivation time is susceptible to all three of the parameters investigated. At a leakage current of 10^{-3} A m^{-2} and D of 1.2×10^{-10} m^2 sec^{-1} the passivation time increases from 30 years at zero radiation dose to 125 years at 3.5×10^3 R h^{-1}. This suggests that extended storage would be beneficial in reducing the period during which localised attack is possible.

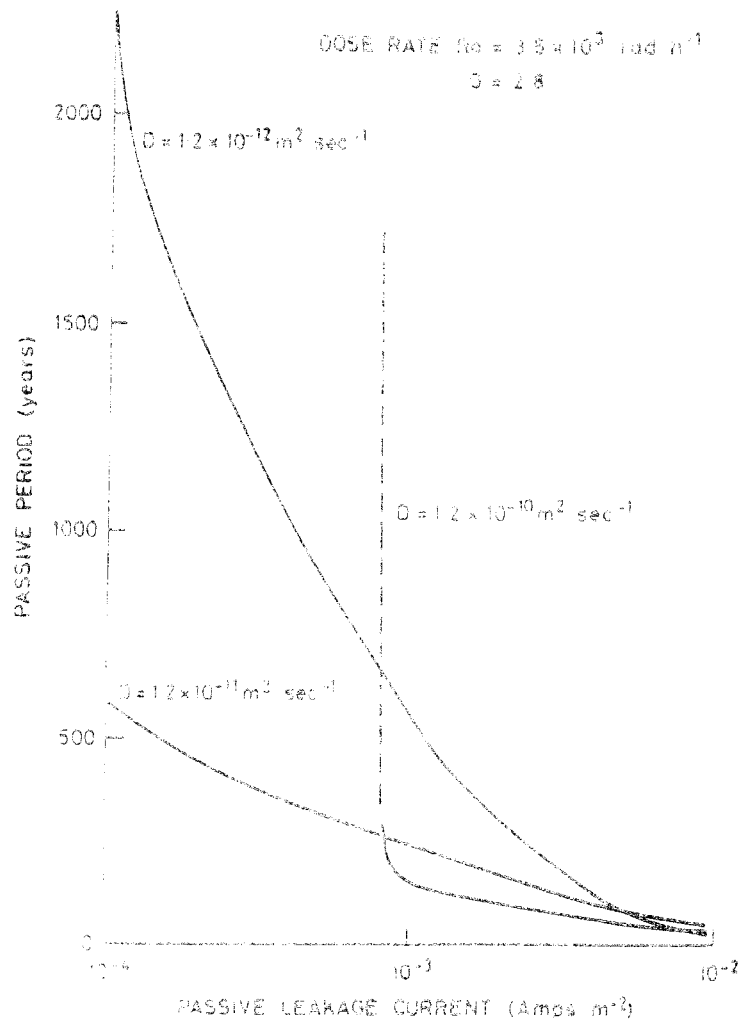Figure 5   Passivation period as a function of leakage current and oxygen diffusion coefficient with a γ-radiation dose of 3.5×10^3 R h^{-1}.

Interpretation of the effect of the diffusion coefficient is difficult because of the complicated behaviour predicted for the highest radiation dose rate, which was discussed above. At this stage, in what is an on-going project, the only additional observation which can be made is that the results in figure 5 may under-estimate the effect of radiation. This is because the calculations were made for constant D, whereas in practice the cooling of the repository will cause D to fall with time. This will tend to increase the passivation periods beyond those given in figure 5.

Finally the results show that if this analysis is to be applied successfully it is essential to have reliable data on the passive leakage current. This is particularly the case if D is the region of 1.2×10^{-10} m^2 sec^{-1} since a change in the leakage current from 10^{-3} to 9×10^{-4} A m^{-2} may change the passivation time from 30-125 years to ∞.

ESTIMATING THE CORROSION ALLOWANCE

An indication of the benefit of estimating the passivation period can be gained using the data in figure 5. In this example the oxygen diffusion coefficient is taken to be 1.2×10^{-10} m^2 sec^{-1} and the leakage current to be 10^{-3} A m^{-2}. This leakage current is considered to be reasonably conservative since results in the literature for HCO_3^- and $CO_3^=$ solutions indicate currents of ∿ 10^{-2} A m^{-2} [11,20]. Furthermore the literature results are for 25°C rather than the 90°C temperature likely at least initially in a granite repository and inferred by the use of a D value of 1.2×10^{-10} m^2 sec^{-1}. On the other hand, however, the literature data were obtained from short term electrochemical tests, and it would be expected that the leakage current would have fallen to some extent with time. In fact one important requirement for applying this method for evaluating corrosion allowances is the provision of reliable data on the passive leakage currents of carbon steel under relevant environmental conditions.

Using the values specified above, figure 5 indicates a passivation period of 125 years. Assuming that localised corrosion occurs during this period the maximum depth of penetration can be estimated from the expression [3]

$$P = 8.35 \ t^{0.46}$$
(3)

where P is in mm and t in years. This predicts a maximum localised corrosion penetration of 77 mm.

Beyond the first 125 years the containers will be subject to general corrosion, and in this case experimental measurements have indicated that the maximum rate of attack will be ∿ 10 μm/yr [3]. This infers an additional corrosion allowance for a 1000 year container life of ∿ 9 mm, so that the total allowance is 86 mm. This should be compared with an allowance of 200 mm if it had been assumed that localised corrosion could occur throughout the container life.

It must be emphasised that the above has only been presented by way of an example of the potential benefit to be gained by estimating the period during which localised corrosion may occur. A reliable estimate must await refinement of the model to take account of the change in repository temperature with time, and better estimates of the passive leakage currents under repository conditions. However, the example illustrates that this approach may reduce the corrosion allowance by the order of 50%, which would be beneficial not only in reducing material costs but also in simplifying container manufacture and seal welding. It will also reduce the weight of the containers which has implications on handling and transportation problems underground.

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CONCLUSION

1. A mechanistically based approach has been developed for estimating the maximum period during which carbon steel containers for HLW may be subject to localised corrosion in a geological repository.

2. A mathematical model is under development for estimating the maximum period for localised corrosion based on the KBS design for a granite repository using bentonite as a backfill material.

3. At present the model works for constant temperature conditions but further development is underway to include the temperature transients which will occur under repository conditions.

4. A sensitivity analysis using the existing constant temperature model has shown that the estimated localised corrosion period increases significantly with radiation dose rate.

5. The localised corrosion period is also strongly dependent on the oxygen diffusion coefficient. Without radiation the localised corrosion period falls as the diffusion coefficient is reduced, but at a dose rate of 3.5×10^3 R h^{-1} this trend is reversed. This has been attributed to the retention of oxidising radiolytic specimens at the container surface under low diffusivity conditions.

6. The localised corrosion period is also critically dependent on the passive film leakage current, and more accurate long term data on this parameter are needed before reliable estimates can be made.

7. A demonstration calculation using existing data has shown that the corrosion allowance specified for carbon steel containers may be reduced by the order of 50% if the above model is used to estimate the localised corrosion period rather than assuming such attack will continue throughout the container life. This reduction has potential benefits for the manufacture, seal welding and underground handling of carbon steel containers.

ACKNOWLEDGEMENTS

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