SKB TECHNICAL REPORT 94-13

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October 1993

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Information on SKB technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26), 1981 (TR 81-17), 1982 (TR 82-28), 1983 (TR 83-77), 1984 (TR 85-01), 1985 (TR 85-20), 1986 (TR 86-31), 1987 (TR 87-33), 1988 (TR 88-32), 1989 (TR 89-40), 1990 (TR 90-46), 1991 (TR 91-64) and 1992 (TR 92-46) is available through SKB.

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Keywords: corrosion, stresses, modelling

EXECUTIVE SUMMARY

The Advanced Cold Process Canister (ACPC) is a concept for the encapsulation of spent nuclear fuel for geological disposal, which is being evaluated jointly by SKB of Sweden and TVO of Finland. The basic design of the ACPC consists of an outer oxygen-free copper overpack covering a carbon steel inner container.

In this report, the stresses exerted on the copper overpack as a result of an early breach of the canister, and the subsequent corrosion of the steel, are calculated. It is assumed that there is a circumferential crack in the copper canister. The crack allows water to flow into the annular gap between the steel and copper vessels, which will cause corrosion of the steel. Because the metal oxide produced by the corrosion reaction occupies a greater volume than the metal, the corrosion of the steel will eventually lead to stresses being exerted on the inside of the copper canister. This report assesses the size and location of these stresses.

The hoop stress σ_θ is a typical stress acting on the canister. The calculations in this report suggest that

$$\frac{d\sigma_\theta}{dt} = 3 \times 10^{11} \frac{d\delta}{dt}$$

where $d\delta/dt$ is the rate of increase in thickness of the corrosion residue in the annulus. If the corrosion is aerobic, corresponding to an early canister failure, the thickness of the corrosion residue increases by $1 - 100$ μm/year. If the corrosion is anaerobic, corresponding to canister failure at a later stage when conditions in the repository have become anaerobic, the thickness of the corrosion residue increases by $0.1 - 1$ μm/year. Implications for the corrosion behaviour of the copper canister are discussed.

A diffusion calculation implies that, even after the annular gap between the steel and copper vessels has filled with corrosion residue, under most repository conditions corrosion will continue to take place over a significant fraction of the steel surface, and therefore the crack in the copper canister is unlikely to 'yawn'.

ABSTRACT

The Advanced Cold Process Canister (ACPC) is a concept for the encapsulation of spent nuclear fuel for geological disposal. The basic design of the ACPC consists of an outer oxygen free copper overpack covering a carbon steel inner container.

In this report the stresses exerted on the copper overpack as a result of an early failure of the canister and the subsequent corrosion of the steel are calculated.

Abstract (Swedish)

The Advanced Cold Process Canister (ACPC) är ett system för inkapsling av använt kärnbränsle för geologisk slutförvaring. ACPC:s Grundkonstruktionen består av en ytter kapsel i syrefri koppar öevr en inre kolstålsbehållare.

Denna rapport redovisar beräkningar av de spänningar som utövas på kopparkapseln som ett resultat av ett tidigt brott på kapseln och den därpå följande stålkorrosionen.

1. INTRODUCTION

1.1. Description of concept

The Advanced Cold Process Canister (ACPC) is a concept for the encapsulation of spent nuclear fuel for geological disposal, which is being evaluated jointly by SKB of Sweden and TVO of Finland. The intention is to use these canisters for the disposal of either PWR or BWR fuel in a KBS 3 type repository design.

The basic design of the ACPC consists of an outer oxygen-free copper overpack covering a carbon steel inner container as shown in Figure 1. The copper overpack has a thickness of 50 mm and the carbon steel container has a thickness of 50 mm. There is a 2 mm annular fitting gap between the steel and copper vessels. The copper overpack is intended to be the principal barrier to corrosion degradation by external groundwater, while the role of the carbon steel container is to provide structural support.

It is intended that the canisters should be placed in separate bore-holes drilled into the floor of tunnels cut into granite rock in accordance with the KBS 3 design study. These bore-holes will be backfilled with compacted sodium bentonite. After water saturation of the repository, the external pressure will reach a value of 15 MPa resulting from the hydrostatic pressure of 5 MPa and a bentonite swelling pressure of 10 MPa.

1.2. Description of research

The ACPC has been designed so that it will provide an effective barrier to radionuclide transport for thousands of years. In order to justify this assertion it is necessary to consider a number of possible scenarios for canister failure.

In this report we consider the effect of an early failure of the copper overpack. It will be assumed that there is a circumferential crack in the copper canister. The crack will allow water to flow into the annular gap between the steel and copper vessels, which will cause corrosion of the steel. Because the metal oxide produced by the corrosion reaction occupies a greater volume than the metal, the corrosion of the steel will eventually lead to stresses being exerted on the inside of the copper canister. These stresses could make the copper susceptible to corrosion, and in particular to stress corrosion cracking; this report assesses the size and location of the stresses.

In section 2 some important concepts from the theory of elasticity and the material properties appropriate to our calculations are summarised. Section 3 describes a model of the stresses in the copper canister, and presents two approaches to calculating the stresses. A 'thick cylinder' calculation can be used when, even after the annular gap has filled with corrosion product, the steel continues to corrode over much of its surface area. The stresses on a circular cross-section of the copper canister far from the crack are calculated. Alternatively, a 'thin cylinder' calculation can be used to consider the stresses both near the crack and at the ends of the copper canister. Section 4 is concerned with predicting the location of the corrosion of the steel canister, and therefore the stresses in the copper canister. In particular the results are used to determine whether the crack in the copper canister will 'yawn'. The implications of the above calculations for canister performance are discussed in section 5.

2. ELASTICITY

2.1. Equations of elasticity [1]

In two-dimensional cylindrical polar coordinates the stress tensor has three components as shown in Figure 2. The normal stress component in the radial direction is

$$\sigma_r$$

the normal stress component in the circumferential direction is

$$\sigma_\theta$$

and the shear stress component is

$$\tau_{r\theta}$$

Summing up the forces in the radial direction that act on a small element one derives the equation of equilibrium

$$\frac{\partial \sigma_r}{\partial r} + \frac{1}{r}\frac{\partial \tau_{r\theta}}{\partial \theta} + \frac{\sigma_r - \sigma_\theta}{r} + F = 0 \tag{1}$$

where F is a body force per unit volume in the radial direction. Similarly the equation of equilibrium in the tangential direction is

$$\frac{1}{r}\frac{\partial \sigma_\theta}{\partial \theta} + \frac{\partial \tau_{r\theta}}{\partial r} + \frac{2\tau_{r\theta}}{r} = 0 \tag{2}$$

In considering the displacement of the small element, let u and v denote the components of the displacement in the radial and tangential directions respectively. Then the strain in the radial direction is

$$\epsilon_r = \frac{\partial u}{\partial r} \tag{3}$$

the strain in the tangential direction is

$$\epsilon_\theta = \frac{u}{r} + \frac{1}{r}\frac{\partial v}{\partial \theta} \tag{4}$$

and the shearing strain is

$$\gamma_{r\theta} = \frac{1}{r}\frac{\partial u}{\partial \theta} + \frac{\partial v}{\partial r} - \frac{v}{r} \tag{5}$$

The three strain components are expressed by two functions u and v, and so cannot be independent. There is a differential relation connecting the strain components called the Beltrami-Michell compatibility condition.

Relations between the components of stress and the components of strain have been established experimentally and are known as Hooke's law. In two-dimensional cylindrical polar coordinates these equations take the form

$$\epsilon_r = \frac{1}{E}(\sigma_r - \nu\sigma_\theta) \tag{6}$$

$$\epsilon_\theta = \frac{1}{E}(\sigma_\theta - \nu\sigma_r) \tag{7}$$

$$\gamma_{r\theta} = \frac{1}{G}\tau_{r\theta} \tag{8}$$

where E is the modulus of elasticity in tension, ν is Poisson's ratio and G

$$G = \frac{E}{2(1+\nu)} \tag{9}$$

is the modulus of elasticity in shear.

The above constitute a complete set of defining equations, relating the stresses σ_r, σ_θ and $\tau_{r\theta}$ to the displacements u and v.

2.2. Elastic properties

For a fully isotropic material, as has been assumed above, only two physical constants are necessary to characterise the elastic properties.

In the case of carbon steel [2] these constants are

$$E_s = 2.06 \times 10^{11} \text{ Pa}$$

and

$$\nu_s = 0.30$$

For copper [2]

$$E_c = 1.17 \times 10^{11} \text{ Pa}$$

and

$$\nu_c = 0.37$$

Magnetite is a cubic crystal, and so three constants are required to describe its elastic properties [3]

$$C_{xxxx} = 2.70$$

$$C_{xxyy} = 1.08$$

$$C_{xyxy} = 0.987$$

The elastic constants of a polycrystalline material can be related to those of the individual constituents in a number of ways. For a monomineralic aggregate, two methods of averaging the single-crystal constants proposed by Voigt and Reuss [3] yield upper and lower limits for the two elastic constants of the quasi-isotropic aggregate of zero porosity. In the calculations described below, we shall make the simplifying assumption that the magnetite is incompressible.

3. STRESSES ON THE COPPER CANISTER

3.1. Model

It is assumed that there is a circumferential crack in the copper canister. The crack will allow water to flow into the annular gap between the steel and copper vessels, which will cause corrosion of the steel. Because the metal oxide produced by the corrosion reaction occupies a greater volume than the metal, the corrosion of the steel will eventually lead to stresses being exerted on the inside of the copper canister.

Experimental observations suggest that the following assumptions regarding the corrosion reaction can be made:

- the steel undergoes approximately uniform corrosion
- the rate of the corrosion reaction is not sensitive to pressure [4]

The model of the formation of corrosion product in the annular gap is that the steel will corrode uniformly at a constant rate until the gap is full of residue. Thereafter, the corrosion reactants and/or charge carrying species will have to diffuse from the crack in the copper canister through the corrosion residue to the steel. The effect of this diffusion process on the corrosion reaction is modelled by assuming that corrosion will continue to occur at the same rate, but over a restricted area of the steel surface.

Two cases of the aeration conditions outside the canister will be considered:

- oxygen present, which corresponds to an early canister failure
- oxygen absent, since ultimately conditions in the repository become anaerobic

Under aerobic conditions localised corrosion is observed to occur. Pit growth rates have been measured [5] by placing rectangular plates of carbon steel in an electrolyte of 0.1M $NaHCO_3$ and 1000ppm Cl^- designed to simulate Swedish granitic groundwater. The plates were subjected to 'constant current polarisation', an arrangement that corresponds to a constant flux of cathodic reactant reaching the metal surface. The current used was $5\mu A/cm^2$, which is the maximum current that can be supported by the diffusion of oxygen through a 0.5m thickness of backfill from a tunnel flooded with water containing dissolved oxygen. The maximum pit depth was found to fit an equation of the form

$$P = 2.89t^{0.34}\text{mm}$$

where t is the time in years. It was observed that the pits spread laterally faster than they increased in depth and that the area of metal surface subjected to corrosion increased with time. After two years, which is about the time it will take the annular gap in the ACPC to fill with corrosion residue, it is not unreasonable to assume that the corrosion is uniform with a corrosion rate in the range $1 - 100\mu$m/year. (Note that a corrosion current of $5\mu A/cm^2$ would correspond to a uniform corrosion rate of 58μm/year.)

Under anaerobic conditions uniform corrosion is observed to occur, and the corrosion rate of steel has been measured as $0.1 - 1\mu$m/year [4].

Some analytic calculations of the stresses experienced by a cylindrical copper canister subject to the above deformations are described below.

3.2. Thick cylinder calculations

This first calculation calculates the stresses acting on a circular cross-section of the copper canister, at a distance from the circumferential crack.

For a problem for which

- two-dimensional cylindrical polar coordinates are applicable
- the stress distribution is symmetrical with respect to an axis through the origin and perpindicular to the xy-plane
- the body force is zero

the stresses can be calculated completely generally as

$$\sigma_r = \frac{A}{r^2} + B \tag{10}$$

and

$$\sigma_\theta = -\frac{A}{r^2} + B \tag{11}$$

These equations are known as the Lamé equations [1]. The constants A and B are determined by the boundary conditions.

This solution can be adapted to represent the stress distribution in a hollow cylinder submitted to a uniform pressure on both its inner

$$\sigma_r(r = R_i) = -P_i \tag{12}$$

and outer surfaces

$$\sigma_r(r = R_o) = -P_o \tag{13}$$

In particular we find that

$$A = \frac{R_i^2 R_o^2 (P_o - P_i)}{R_o^2 - R_i^2} \tag{14}$$

$$B = \frac{P_i R_i^2 - P_o R_o^2}{R_o^2 - R_i^2} \tag{15}$$

Note that the sum $\sigma_r + \sigma_\theta$ is constant through the thickness of the wall of the cylinder. Thus the stresses σ_r and σ_θ produce a uniform extension or contraction in the direction of the axis of the cylinder; cross-sections perpindicular to the axis remain plane, and it is justifiable to consider the element to be in a condition of plane stress.

The displacement of the wall of the cylinder can be calculated as

$$u = \frac{1}{E}\left(-\frac{(1+\nu)A}{r} + B(1-\nu)r\right) + C \tag{16}$$

In the case of the ACPC, the above equations can be applied to a scenario in which, even after the annular gap has filled with corrosion product, the steel continues to corrode over much of its surface area. The stresses on a circular cross-section of the copper canister far from the crack can be calculated.

The constant C is determined from the boundary condition that the inner surface of the copper canister should be undeformed when the pressure on the inner and outer surfaces is equal to 1 atmosphere

$$C = \frac{1.013 \times 10^5}{E}(1-\nu)R_i \tag{17}$$

The constants A and B are determined from the boundary conditions that the inner surface of the copper canister

$$R_i = 0.39 \text{ m}$$

has undergone a radial displacement

$$u(r = R_i) = \delta \tag{18}$$

and the outer surface

$$R_o = 0.44 \text{ m}$$

is submitted to a uniform pressure

$$\sigma_r(r = R_o) = -P \tag{19}$$

Thus

$$\begin{aligned} A &= \frac{-\delta E - (P - 1.013 \times 10^5)(1-\nu)R_i}{(1+\nu)R_i/R_i^2 + (1-\nu)R_i/R_o^2} \\ &= -2.4 \times 10^{10}\delta - 5.1 \times 10^{-2}(P - 1.013 \times 10^5) \end{aligned} \tag{20}$$

and

$$
\begin{aligned}
B &= -\frac{A}{R_o^2} - P \\
&= 1.3 \times 10^{11}\delta + 2.7 \times 10^{-1}(P - 1.013 \times 10^5) - P
\end{aligned} \tag{21}
$$

Four cases can be distinguished, depending on whether the copper canister is undergoing deformation before saturation of the repository with water, when the external pressure is

$$P = 1.013 \times 10^5 \text{ Pa}$$

or after saturation, when

$$P = 1.5 \times 10^7 \text{ Pa}$$

and whether the corrosion is aerobic

$$\frac{d\delta}{dt} = 1 - 100 \ \mu\text{m/year}$$

or anaerobic

$$\frac{d\delta}{dt} = 0.1 - 1 \ \mu\text{m/year}$$

The hoop stress σ_θ exerted on the inner surface of the copper canister after a time t years is shown in Table 1, and plotted in Figure 3

	P (Pa)	$d\delta/dt$(μm/year)	σ_θ(Pa)
unsaturated/anaerobic	1.013×10^5	0.1	$2.9 \times 10^4 t - 1.013 \times 10^5$
saturated/anaerobic	1.5×10^7	0.1	$2.9 \times 10^4 t - 6.0 \times 10^6$
unsaturated/aerobic	1.013×10^5	10	$2.9 \times 10^6 t - 1.013 \times 10^5$
saturated/aerobic	1.5×10^7	10	$2.9 \times 10^6 t - 6.0 \times 10^6$

Table 1 Hoop stress exerted on the inner surface of the copper canister.

3.3. Thin cylinder calculations [6]

An alternative approach to calculating the stresses on a cylinder is provided by the theory of thin cylindrical shells. The advantage of the 'thin cylinder' calculation, is that it can be used to consider the stresses both near the crack and at the ends of the copper canister.

This theory assumes that the thickness of the cylindrical shell h is much smaller than its radius, which is only partially justified in the case of the ACPC (nevertheless meaningful numbers can be calculated using this approach). The surface that bisects the thickness of the shell is called the 'middle surface'. It is usual to define a system of coordinate axes in which the axes x and y are tangent at O to the lines of principal curvature, and the axis z is normal to the middle surface.

In the case of the copper canister, the forces acting on the cylindrical shell are distributed symmetrically with respect to the axis of the cylinder. To derive the equations required for the solution of this problem, we consider a small element as shown in Figure 4. It can be concluded from symmetry that three of the six equations of equilibrium of the element are identically satisfied. The remaining equations are obtained by projecting the forces on the x and z axes, and by taking the moment of the forces about the y axis. Assuming that the external force consists only of a pressure normal to the surface, these three equations of equilibrium are

$$\frac{dN_x}{dx} = 0 \tag{22}$$

$$\frac{dQ_x}{dx} + \frac{N_\phi}{a} = -F \tag{23}$$

$$\frac{dM_x}{dx} - Q_x = 0 \tag{24}$$

where

$$N_x = \int_{-h/2}^{+h/2} \sigma_x \left(1 - \frac{z}{a}\right) dz \tag{25}$$

$$N_\phi = \int_{-h/2}^{+h/2} \sigma_\phi \left(1 - \frac{z}{a}\right) dz \tag{26}$$

$$Q_x = \int_{-h/2}^{+h/2} \tau_{xz} \left(1 - \frac{z}{a}\right) dz \tag{27}$$

$$M_x = \int_{-h/2}^{+h/2} z\sigma_x \left(1 - \frac{z}{a}\right) dz \tag{28}$$

The first equation indicates that the force N_x is constant, and it shall be taken to be zero. The remaining two equations define the stresses produced by a lateral load. These two equations contain three unknown quantities N_ϕ, Q_x and M_x.

Next we consider the displacements of points in the middle surface of the shell. From symmetry, the component v of the displacement in the circumferential direction vanishes. We therefore have only to consider the components u and w in the x and z directions. The expressions for the strain components are

$$\epsilon_x = \frac{du}{dx} \tag{29}$$

$$\epsilon_\phi = -\frac{w}{a} \tag{30}$$

Applying Hooke's law we obtain

$$N_x = \frac{Eh}{1-\nu^2}(\epsilon_x + \nu\epsilon_\phi) \tag{31}$$

$$N_\phi = \frac{Eh}{1-\nu^2}(\epsilon_\phi + \nu\epsilon_x) \tag{32}$$

The above constitute a complete set of defining equations for the forces acting on, and the displacements of, a cylindrical shell.

3.3.1. *Cylindrical shell submitted to a uniform internal pressure*

The first problem that we consider is a cylindrical shell submitted to a uniform internal pressure P. P produces only a hoop stress

$$\sigma_\theta = \frac{Pa}{h} \tag{33}$$

and the radius of the cylinder increases by an amount

$$\delta = \frac{a\sigma_\theta}{E} \tag{34}$$

Thus

$$\begin{aligned} \sigma_\theta &= \frac{E}{a}\delta \\ &= 2.8 \times 10^{11}\delta \text{ Pa} \end{aligned} \tag{35}$$

which suggests that if the corrosion is aerobic with corrosion rate 10 μm/year, the hoop stress σ_θ exerted on the surface of the copper canister after a time t years is

$$\sigma_\theta = 2.8 \times 10^6 t \text{ Pa} \tag{36}$$

and if the corrosion is anaerobic with corrosion rate 0.1 μm/year

$$\sigma_\theta = 2.8 \times 10^4 t \text{ Pa} \tag{37}$$

These results are in good agreement with the results of the 'thick cylinder' calculation.

3.3.2. *Long cylindrical shell submitted to a load uniformly distributed at one end*

Next we consider a long cylindrical shell submitted to the action of bending moments M_o and shearing forces Q_o distributed uniformly along the edge $x = 0$ (Figure 4). The expression for the deflection of the surface in the z direction is

$$w = \frac{e^{-\beta x}}{2\beta^3 D}(\beta M_o(\sin\beta \mathrm{x} - \cos\beta \mathrm{x}) - \mathrm{Q_o}\cos\beta \mathrm{x}) \tag{38}$$

where

$$D = \frac{Eh^3}{12(1-\nu^3)} \tag{39}$$

$$\beta^4 = \frac{3(1-\nu^2)}{a^2h^2} \tag{40}$$

The maximum deflection occurs at the loaded end where

$$w(x=0) = -\frac{1}{2\beta^3 D}(\beta M_o + Q_o) \tag{41}$$

The negative sign for this deflection results from the fact that w is taken positive toward the axis of the cylinder. This result suggests that, near the crack in the copper canister, the forces are given by

$$8.8M_o + Q_o = 1.8 \times 10^{12}\delta \tag{42}$$

These forces can be related to stresses through equations (27) and (28). These stresses are of similar magnitude to the hoop stress discussed earlier.

3.3.3. *Cylindrical shell with fixed ends submitted to a uniform internal pressure*

Finally, we consider a cylindrical shell submitted to a uniform internal pressure P, and assume that the ends of the shell are fixed. If the length of the shell is large, the solution

$$w = \frac{e^{-\beta x}}{2\beta^3 D}\left(\beta M_o(\sin\beta x - \cos\beta x) - Q_o\cos\beta x\right) \tag{43}$$

can also be used to investigate the bending that occurs at the edges. The bending moment M_o and the shearing force Q_o are determined from the boundary conditions

$$\begin{aligned} w(x=0) &= \delta && (44)\\ \frac{dw}{dx}(x=0) &= 0 && (45) \end{aligned}$$

Solving for M_o and Q_o

$$\begin{aligned} M_o &= 2\beta^2 D\delta && (46)\\ &= 2.0 \times 10^{11}\delta \\ Q_o &= -4\beta^3 D\delta && (47)\\ &= -3.5 \times 10^{12}\delta \end{aligned}$$

This result suggests that a positive bending moment and a negative shearing force act at the ends of the copper canister. These forces can be related to stresses through equations (27) and (28). These stresses are of similar magnitude to the hoop stress discussed earlier.

4. LOCATION OF STRESSES AND CRACK YAWNING

4.1. Model

The final question that we must address is at what position in the annular gap between the steel and copper vessels corrosion will take place.

It is assumed that there is a circumferential crack in the copper canister. The physical picture is that corrosion in the vicinity of the crack will be controlled by the diffusion of reactants/charge-carrying species through the corrosion residue. If the diffusion process is such that corrosion takes place over a significant fraction of the steel surface, then the copper vessel will be uniformly distorted. Alternatively, if corrosion takes place over an area of the steel surface that is comparable in size to the area of the crack, then the crack will 'yawn'.

4.2. Diffusion calculation

In the model described here the annular gap is taken to be infinitely long, with inner radius R_i and outer radius R_o. There is a circumferential crack with half-width w in the outer surface of the annulus at

$$r = R_o, z = 0$$

and the concentration of 'reactants' at the crack is given by

$$C_o$$

In the case of aerobic corrosion, the reactant is likely to be oxygen, though if a differential aeration cell were to be set up the rate controlling process might be the transport of 'the ions in the groundwater which carry the electrical charge'. In the case of anaerobic corrosion, the reactant is water.

In the absence of detailed knowledge of the corrosion process, we shall assume that the inner surface of the annulus has a constant flux of reactants/charge carrying species

$$\frac{i}{nF} \tag{48}$$

where i is the corrosion current, n is a constant of order unity depending on the stoichiometry of the corrosion reaction, and F is Faraday's constant. This assumption about the corrosion current is of limited validity for aerobic corrosion, but is correct for anaerobic corrosion.

Since the surface of the steel canister moves on a much longer timescale than the characteristic time of the diffusion process, it is legitimate to use the steady-state diffusion equation in two-dimensional cylindrical planar coordinates to describe the diffusion of reactants through the corrosion residue.

The equation that we have to solve is

$$D\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial c}{\partial r}\right) + D\frac{\partial^2 c}{\partial z^2} = 0 \tag{49}$$

subject to the boundary conditions

$$c = C_o \qquad r = R_o, 0 < z < w \tag{50}$$

$$\frac{\partial c}{\partial r} = 0 \qquad r = R_o, w < z < \infty \tag{51}$$

$$\frac{\partial c}{\partial z} = 0 \qquad R_i < r < R_o, z = 0 \tag{52}$$

$$D\frac{\partial c}{\partial r} = \frac{i}{nF}H(c) \qquad r = R_i, 0 < z < \infty \tag{53}$$

where H is the Heaviside step function.

We can non-dimensionalize the above equation, giving

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial c}{\partial r}\right) + \frac{\partial^2 c}{\partial z^2} = 0 \tag{54}$$

subject to the boundary conditions

$$c = 1 \qquad r = \frac{R_o}{w}, 0 < z < 1 \tag{55}$$

$$\frac{\partial c}{\partial r} = 0 \qquad r = \frac{R_o}{w}, 1 < z < \infty \tag{56}$$

$$\frac{\partial c}{\partial z} = 0 \qquad \frac{R_i}{w} < r < \frac{R_o}{w}, z = 0 \tag{57}$$

$$\frac{\partial c}{\partial r} = \frac{i}{nFC_oD}wH(c) \qquad r = \frac{R_i}{w}, 0 < z < \infty \tag{58}$$

Solutions of this differential equation were found numerically as a function of the parameters

$$\Lambda = \frac{i}{nFC_oD} \tag{59}$$

and

$$w$$

The corrosion rate of steel has been measured in the range

$$0.1 - 100\ \mu\text{m/year} \tag{60}$$

and so

$$\frac{i}{nF} = 5 \times 10^{-10} - 5 \times 10^{-7}\ \text{mole/m}^2\text{/s} \tag{61}$$

The details of the diffusion process are not known. However, for gaseous diffusion it would be reasonable to assume

$$D = 10^{-5}\ \text{m}^2\text{/s} \tag{62}$$

and

$$C_o < 10\ \text{mole/m}^3 \tag{63}$$

If diffusion takes place either in the liquid phase or through the pore space of a saturated solid, then typically

$$D = 10^{-10}\ \text{m}^2\text{/s} \tag{64}$$

and

$$C_o < 5 \times 10^4 \text{ mole/m}^3 \tag{65}$$

On the basis of the above considerations, values of Λ in the range

$$10^{-6} - 1 \text{ m}^{-1} \tag{66}$$

were considered. For most combinations of aeration and saturation conditions in the repository, Λ will lie in the lower part of this range. However, if we consider aerobic corrosion in a repository saturated with groundwater then Λ will be large, and perhaps larger than 1 m^{-1}.

The size of the active surface of the steel, measured as the length of the inner surface of the annulus at which c is positive, is shown in Table 2

	$w = 10^{-4}$ m	$w = 10^{-3}$ m	$w = 10^{-2}$ m
$\Lambda = 10^{-6}$ m^{-1}	3.95×10^3 m	1.25×10^3 m	3.95×10^2 m
$\Lambda = 10^{-5}$ m^{-1}	1.25×10^3 m	3.95×10^2 m	1.25×10^2 m
$\Lambda = 10^{-4}$ m^{-1}	3.95×10^2 m	1.25×10^2 m	3.96×10^1 m
$\Lambda = 10^{-3}$ m^{-1}	1.25×10^2 m	3.95×10^1 m	1.25×10^1 m
$\Lambda = 10^{-2}$ m^{-1}	3.95×10^1 m	1.25×10^1 m	3.96 m
$\Lambda = 10^{-1}$ m^{-1}	1.25×10^1 m	3.96 m	1.26 m
$\Lambda = 1$ m^{-1}	3.95 m	1.25 m	4.05×10^{-1} m

Table 2 Predicted size of the corroding surface of the steel.

For the range of parameters considered, these numbers are summarised by the formula

$$\frac{0.03955}{\sqrt{\Lambda w}} \tag{67}$$

It follows that if

- the crack width w is small
- $\Lambda = i/nFC_oD$ is small

then the crack in the copper canister will not 'yawn'.

5. IMPLICATIONS FOR CANISTER PERFORMANCE AND SUMMARY

In this report we have considered the effect of an early failure of the copper overpack. It is assumed that there is a circumferential crack in the copper canister. The crack will allow water to flow into the annular gap between the steel and copper vessels, which will cause corrosion of the steel. Because the metal oxide produced by the corrosion reaction occupies a greater volume than the metal, the corrosion of the steel will eventually lead to stresses being exerted on the inside of the copper canister.

The calculations in this report suggest that a typical stress on the copper canister is of the form

$$\frac{d\sigma_\theta}{dt} = 3 \times 10^{11} \frac{d\delta}{dt} \tag{68}$$

where

$$\frac{d\delta}{dt}$$

is the rate of increase in thickness of the corrosion residue in the annulus

$$\frac{d\delta}{dt} = 1 - 100 \ \mu\text{m/year}$$

if the corrosion is aerobic, and

$$\frac{d\delta}{dt} = 0.1 - 1 \ \mu\text{m/year}$$

if the corrosion is anaerobic.

The stresses on the copper canister will increase, as predicted by the above equations, until the thermodynamic limit of the corrosion reaction is reached.

The maximum stresses on the copper canister occur when the steel is corroding aerobically. The amount of oxygen in the repository will decrease as the metals corrode, and the rate at which the stresses increase could fall by as much as two orders of magnitude as the corrosion switches to the anaerobic form.

Stress corrosion cracking (SCC) of copper has been studied using slow strain rate tests. Examples of SCC of pure copper have been observed in $NaNO_2$ solutions and in the US J-13 groundwater [7,8]. SCC occured when the strain rate was in the range

$$10^{-7} - 10^{-5} \ \text{s}^{-1}$$

These strain rates are several orders of magnitude larger than those calculated from the stresses obtained in this report (68) using the strain-stress relationship (7). However, the resistance of copper to SCC when immersed in the Swedish repository groundwater and subjected to the calculated stresses should be determined before any firm conclusions are made.

A diffusion calculation implies that for most combinations of aeration and saturation conditions in the repository, corrosion will continue to take place over a significant fraction of the steel surface, and therefore the crack in the copper canister is unlikely to 'yawn'. The most likely scenario that could lead to a widening of the crack in the copper canister occurs if the repository is saturated with groundwater and the corrosion is aerobic. Such conditions exist for a relatively short time in the lifetime of the repository; the aeration period of the repository should be determined in order to confirm the conclusion that the crack will not yawn.

6. ACKNOWLEDGEMENT

This work was funded by SKB, Sweden. The authors would like to thank C.C. Naish and D. Blackwood of the Applied Electrical Department, AEA Technology for many helpful discussions during the course of this work.

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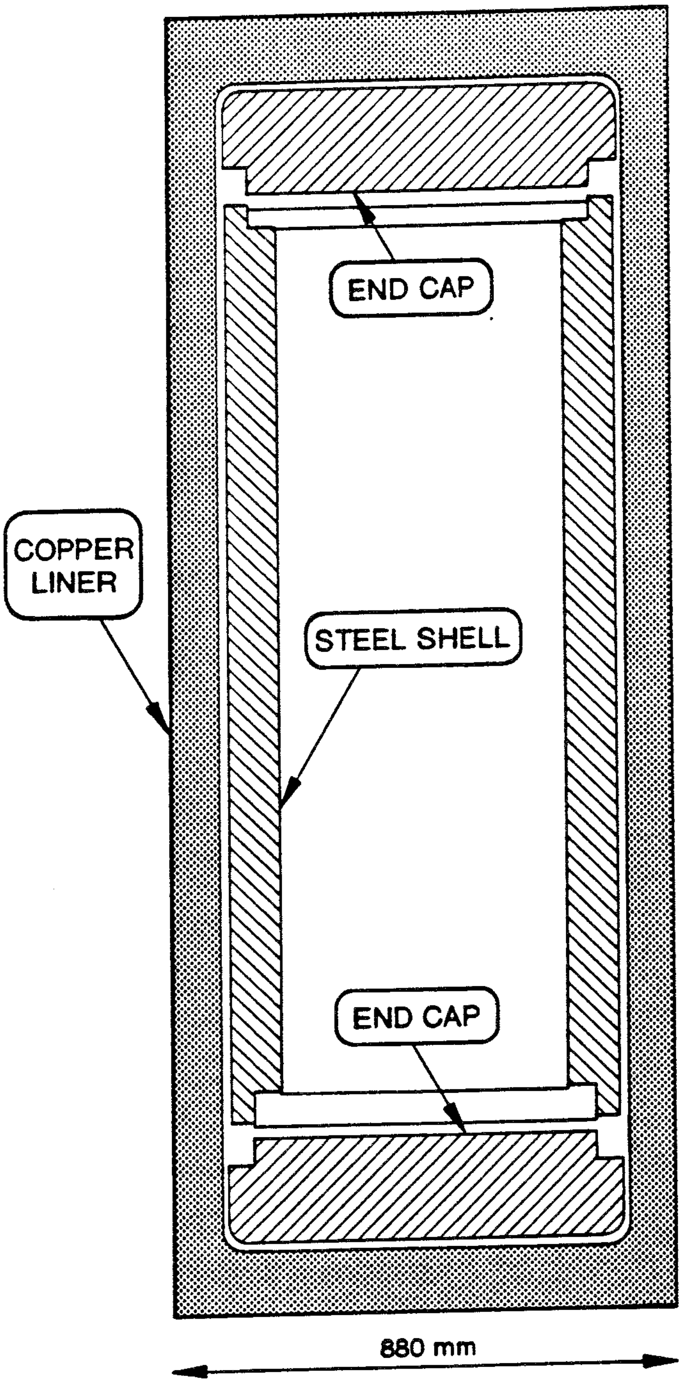

Figure 1
The Advanced Cold Process Canister.

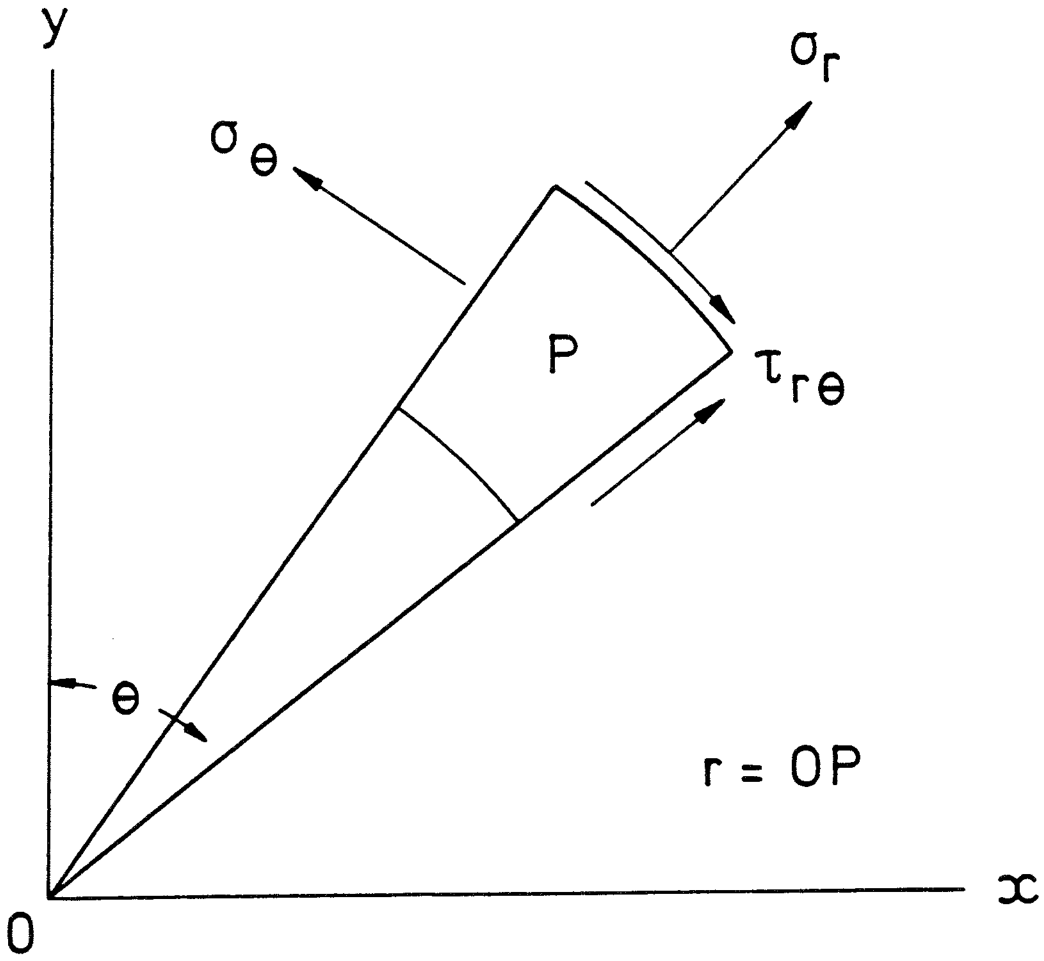

Figure 2
Components of the stress tensor in two-dimensional cylindrical polar coordinates.

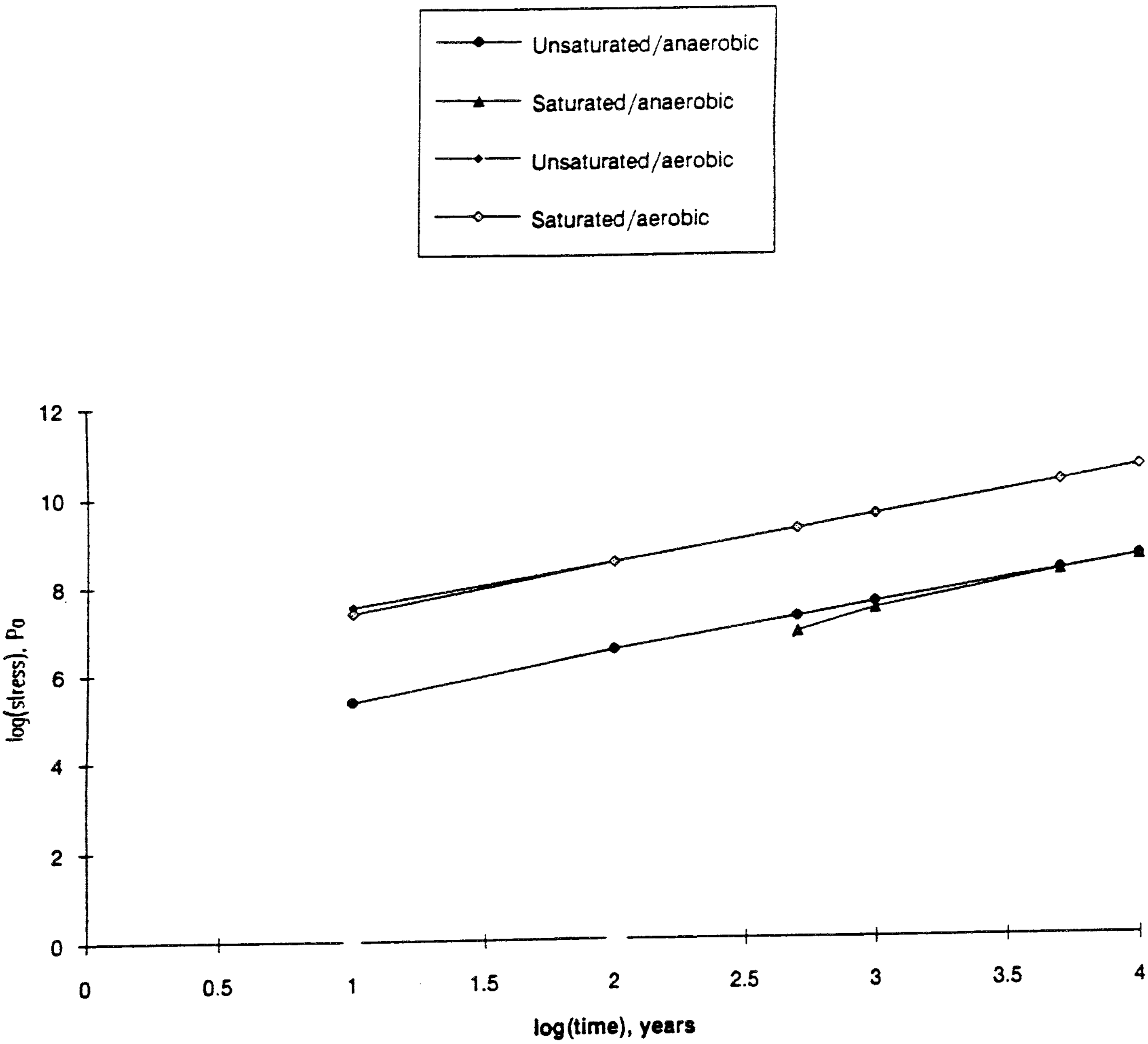

Figure 3
The hoop stress σ_θ exerted on the inner surface of the copper canister.

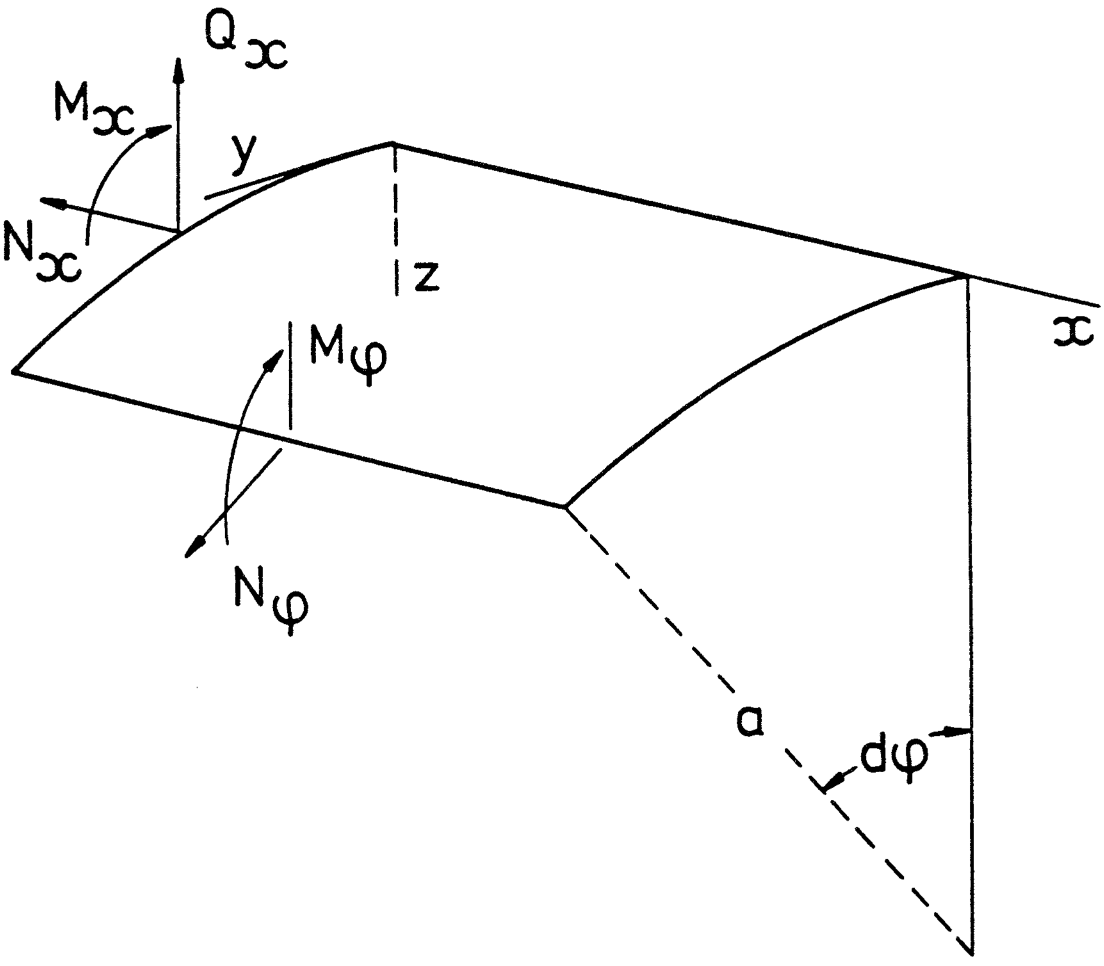

Figure 4
The forces acting on a small element of a thin cylindrical shell.

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3 CTH
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