



Conceptual Design for a Deep Geologic Repository for Used Nuclear Fuel

Annex 1

Metallurgical Aspects of Used Fuel Container Design

December 2002

NOTICE to the Reader

“This document has been prepared by CTECH Radioactive Materials Management, a joint venture of Canatom NPM Inc. and RWE Nukem Ltd. (“Consultant”), to update the conceptual design and cost estimate for a deep geologic repository (DGR) for long term disposal of used nuclear fuel. The scope is more fully described in the body of the document. The Consultant has used its professional judgment and exercised due care, pursuant to a purchase order dated October 2001. (the “Agreement”) with Ontario Power Generation Inc. acting on behalf of the Canadian nuclear fuel owners (“the Client”), and has followed generally accepted methodology and procedures in updating the design and estimate. It is therefore the Consultant’s professional opinion that the design and estimate represent a viable concept consistent with the intended level of accuracy appropriate to a conceptual design, and that, subject to the assumptions and qualifications set out in this document, there is a high probability that actual costs related to the implementation of the proposed design concept will fall within the specified error margin.

This document is meant to be read as a whole, and sections or parts thereof should not be read or relied upon out of context. In addition, the report contains assumptions, data, and information from a number of sources and, unless expressly stated otherwise in the document, the Consultant did not verify those items independently. Notwithstanding this qualification, the Consultant is satisfied that the updated conceptual design and cost estimate was carried out in accordance with generally accepted practices in a professional manner .

This document is written solely for the benefit of the Client, for the purpose stated in the Agreement, and the Consultant’s liabilities are limited to those set out in the Agreement.”

Summary

The overall objective of the work described in this document is to confirm that the materials and design proposed for the Used Fuel Container (UFC) are reasonable to use as the basis for the conceptual design for the Deep Geologic Repository (DGR). Ontario Power Generation (OPG) have selected oxygen-free phosphorus-doped (OFP) copper as the preferred option for constructing the outer shell of the UFC. One factor in making this selection was the fact that there is a large existing body of knowledge about the behaviour of copper in repository environments and about techniques for constructing and inspecting large copper vessels, based mainly on work carried out for the Swedish, Finnish and Canadian authorities. This document presents a summary view of the key metallurgical aspects of using copper for the outer shell of the UFC. It deals with:

- behaviour of the specified UFC materials under the conditions expected in the DGR
- assessing the constructability and inspection of the UFC design.

The document outlines the environments that the UFCs will encounter and discusses the various modes of corrosion that might affect the copper shell during each stage of operation. The role of creep in the deformation of the outer copper shell is reviewed. Studies carried out to investigate methods for constructing the outer copper container are reviewed and recent discussions with potential manufacturers of the copper shell are summarised. Possible methods for inspecting the seal weld are reviewed. It is concluded that it is reasonable to use the proposed UFC design as the basis for the conceptual design of the DGR

Contents

1	Introduction	1
2	UFC Materials and Environment in a Deep Geologic Repository	2
2.1	UFC MATERIALS	2
2.2	ENVIRONMENT DURING MANUFACTURE AND STORAGE	3
2.3	ENVIRONMENT DURING FILLING, SEALING AND REPOSITORY EMPLACEMENT	3
2.4	ENVIRONMENT AFTER SEALING IN REPOSITORY	4
3	Compatibility of Container Materials with DGR Environment	7
3.1	CORROSION DURING MANUFACTURE, STORAGE, FILLING AND SEALING	7
3.2	CORROSION AFTER EMPLACEMENT IN THE REPOSITORY	8
3.3	CREEP DEFORMATION OF THE UFC COPPER SHELL	13
4	Constructability and Inspection of UFCs	15
4.1	CONSTRUCTION OF A UFC	15
4.2	INSPECTION OF UFC	17
5	Conclusions	18
6	References	18

1 Introduction

The overall objective of the work described in this report is to confirm that the materials and design proposed for the Used Fuel Container (UFC) are reasonable to use as the basis for the conceptual design of the Deep Geologic Repository (DGR). The specific aims of this document are to address the requirements stated in the technical specifications for the project [1], namely:

- to assess the compatibility of the specified materials under the conditions expected in the DGR
- to assess the constructability of the UFC design.

In line with the customer's guidelines the task is based on the following assumptions and principles. A factor in OPG's selection of copper for the corrosion barrier of the UFC was the existence of a large knowledge base on copper behaviour and fabrication technology which derives from years of R&D work done by Sweden and Finland, to which OPG has also contributed. Selecting copper gives the benefit of using a well-known material and utilising existing technologies specially developed for nuclear waste applications. Other material choices (such as titanium) would require the commitment to carry out an extensive R&D programme in order to be able to obtain equivalent data to that already available for copper.

Therefore, CTECH was not requested to carry out any additional investigations on copper behaviour but to use the information available from recent research. Accordingly, this assessment draws heavily on the literature provided by OPG and the documents published by SKB and Posiva. For example, SKB have recently published a comprehensive review of the large body of information available about the corrosion of copper [2].

This report presents only a summary view and makes references to the extensive literature base. It is divided as follows. In Section 2 the materials proposed for the UFC are outlined and a brief description of the environmental conditions expected during the waste management process is given. Section 3 deals with the compatibility of the UFC materials with the environmental conditions expected during the used fuel management process. In Section 4 the feasibility of constructing and inspecting the proposed UFC design is discussed.

2 UFC Materials and Environment in a Deep Geologic Repository

The purpose of this section is to outline the materials that will be used for the UFC and to describe the environments that the containers are expected to encounter. In assessing the performance of the UFC materials it is important to characterise the environment at each stage of the operations. Details of the proposed design of the UFC, and the processes by which the UFCs will be handled and placed in the repository, are given in the body of the main report [3].

2.1 UFC MATERIALS

The outer container will be manufactured from oxygen-free phosphorus-doped (OFP) copper [4]. The phosphorus is added to improve creep ductility (see Section 3.3) and it is oxygen-free to confer superior resistance to uniform and localised corrosion processes. SKB has compiled a technical specification for the OFP copper [5], which gives the chemical composition for the material (see Table 1 below). The material may obey either of the specifications given in the table, but in addition trace impurities should be in the following ranges: O<5 ppm, P 40-60 ppm, H<0.6 ppm, S<8 ppm. It is specified that the grain size for the lid and copper tubes shall be less than 360 µm.

	Cu	Ag	As	Fe	S	Sb	Se	Te	Pb
UNS C10100	99.99%	25	5	10	15	4	3	2	5
EN 133/63 Cu-OF1	rem	25	5	10	15	4	4	4	5
	Bi	Cd	Mn	Hg	Ni	O	Sn	Zn	
UNS C10100	1	1	0.5	1	10	5	2	1	
EN 133/63 Cu-OF1									

Table 1. Composition of material selected by SKB (max ppm) for outer copper shell

The inner container will be made from carbon steel (typically SA516 grade 70 for shell plate (C 0.28 wt%; Mn 0.85% wt% [6,7]) and SA105 for the ends).

2.2 ENVIRONMENT DURING MANUFACTURE AND STORAGE

During manufacture and storage the vessels will be exposed to an atmospheric environment. The humidity and atmospheric composition are unknown at present. It is possible that the units will be manufactured remotely, before being transferred to a storage area at the packaging plant. It is understood that the UFCs will be used and transferred to the DGR within a short period of being received at the packaging plant (up to 6 weeks based on first in, first out). The heat emitted from the surface of the container will raise the surface temperature, which will reduce the local relative humidity and prevent any condensation forming on the container surface.

2.3 ENVIRONMENT DURING FILLING, SEALING AND REPOSITORY EMPLACEMENT

After loading the UFC, the inner (steel) lid will be bolted in place, sealing the inner vessel. Following this operation, the UFC will be moved to the electron beam (EB) welding station, where the copper lid will be placed on the container and seal welded. Since the EB welding operation is carried out under vacuum, the interstitial space between the UFC inner and outer shells will remain at vacuum. The inert gas used to fill the inner vessel may diffuse into the annulus after a long period of time, once the mechanical seal of the inner vessel has failed. Steps will be taken to dry the gas used to fill the inner vessel, to avoid hydrogen generation by anaerobic corrosion of steel. During these operations the surfaces of the UFC components will be exposed to indoor atmospheric conditions.

After sealing, the UFC will be placed on to the lower part of the bentonite sleeve and the upper bentonite sleeve will be added, so that in effect all the outer copper surfaces will be placed in contact with oxygenated bentonite. The moisture content of the precompacted bentonite blocks will initially be 85% saturated [2,8] corresponding to a moisture content of 17%, but a moisture profile is expected to develop near the surface of the UFC due to the elevated surface temperature of the UFC (i.e. the moisture level will be lower near the UFC surface than the bulk concentration in the bentonite). Initially the surface temperature will be of the order of 30°C, but this will increase after the bentonite shell has been placed around the container. The design limit for the temperature of the outer surface of the UFC has been set at 100°C. Thermal analysis predicts a peak temperature of 97°C, which will occur sixteen years after emplacement in the repository [9]. The elevated surface temperature may lead to local evaporation and precipitation of dissolved species such as sodium chloride and gypsum (calcium sulphate).

The bentonite sleeve will be 250 mm thick. The complete assembly will then be placed in a repository emplacement room, within a pre-assembled structure constructed with buffer and dense backfill blocks. The buffer material surrounding the bentonite shell will be composed of 50% silica sand and 50% bentonite. Outside the buffer material will be a layer of dense backfill (a compacted mixture of crushed granite aggregate, glacial lake clay and sodium bentonite clay), followed by light backfill (a compacted mixture of crushed granite aggregate and sodium

bentonite clay) and finally the host rock. It is expected that the two halves of the bentonite shell will seal together after being saturated with groundwater [14].

2.4 ENVIRONMENT AFTER SEALING IN REPOSITORY

2.4.1 Groundwater composition

The site of the DGR has not been agreed and so it is not possible to be precise about the groundwater composition. However, data are available for the crystalline rock formations in the Canadian Shield. Gascoyne has published compositional data for a range of locations at depths up to 1000m [10]. Shallow fracture groundwaters are generally dilute calcium-sodium-bicarbonate waters (TDS <0.3 g/L). At greater depths (200-400m below surface) there is a chloride and sulphate component in the formation water and at still greater depths (>500m), waters of increasing salinity are found (>50 g/l salinity). Below 200m, the pH is typically in the range 7.5 to 8.8.

2.4.2 Buffer materials

The major components of a typical bentonite, Wyoming bentonite MX-80, and one which is a candidate buffer material for the DGR and has been used in research programmes by SKB, is as follows [11]:

Element	Percentage
SiO ₂	66.9
Al ₂ O ₃	20.8
Fe ₂ O ₃	4.7
TiO ₂	0.2
MgO	3.1
CaO	1.9
K ₂ O	0.6
Na ₂ O	0.6
P ₂ O ₅	0.1
unknown	1.1

A typical sodium bentonite composition is as follows: Si 29.5; Al 8.4; Fe 3.0; Na 1.6; Ca 1.3; Mg 1.3; K 0.5; C 0.4; Ba 0.4; S 0.3; H 0.8; O 52.5%. Details of the other sealing materials are given in reference [12].

2.4.3 Composition of conditioned groundwater

For the purposes of assessing the corrosion behaviour of the UFC materials it is necessary to estimate the composition of the aqueous phase in contact with the metal surfaces. The composition will be derived from equilibration of the groundwater with the chemical components of the sealing materials (i.e. the groundwater will be 'conditioned' by the sealing materials before it reaches the surface of the container).

After each emplacement room is closed the groundwater will gradually resaturate the sealing materials. SKB [2] have estimated that this process is most likely to occur over a period of six to thirty five years, but periods up to several hundred years cannot be ruled out. As groundwater saturates the bentonite, it is likely that the clay will expand inhomogeneously, so there will be uneven contact between the bentonite and the surface of the copper. SKB assume that contact between the bentonite porewater and the UFC will not occur until the bentonite is fully saturated [2]. In the SKB situation, it is estimated that total porewater replacement will occur within ~14,000 years.

The pH of the saturated bentonite porewater is expected to be mildly alkaline. While calcite is present the pH will be ~8.4 decreasing to about ~6.8 after removal of the calcite [2].

The ammonia concentration is expected to be low (a few mg/litre [2]); it is possible that it may be removed by an irreversible cation exchange process but this has not been verified experimentally [2].

The chloride concentration will depend on the specific chloride concentration in the groundwater. For the purposes of assessing materials compatibility it is necessary to assess the corrosion behaviour in groundwaters with a chloride concentration up to 50 g/litre.

The sulphate concentration in the porewater will increase when it comes into contact with the groundwater due to dissolution of gypsum (calcium sulphate) [2].

The predominant cations will be sodium, calcium, magnesium and silicon [10].

Some concentration of groundwater constituents is expected at the surface of the UFC as a result of the elevated temperature [2].

2.4.4 Temperature and degree of oxygenation

During the exposure period in the repository the temperature and oxidising ability of the environment will change with time, as shown graphically in reference [13]. The major oxidant will be residual oxygen, which will be consumed by reaction with the minerals in the sealing materials, by microbial activity [2], as well as by reactions with the UFC copper shell. Some oxidising species may be formed by radiolysis of the trapped water and oxygen, but these will decrease with time as the radioactivity decays. SKB predict negligible concentrations of radiolysis products on the external surface of the UFC.

The length of the aerated period has been modelled by taking account of the various mechanisms for consumption of oxygen, and it has been estimated that the maximum duration of the aerated period will be 670 years. SKB have estimated that the aeration period in their repository will be between 7 and 290 years. The temperature of the UFC surface will reach a maximum value of approximately 97°C after about 16 years, following which it will fall to the ambient temperature, 20-30°C, after 10^4 - 10^5 years.

The waters tested in the Canadian shield are anoxic, with Eh values typically in the range 0 to 200 mV in near surface waters, falling to a range of –50 to -250 mV at depths greater than 600m [10]. The oxygen content of the water decreases rapidly with increasing depth. At 60m the oxygen concentration has decreased by 99.98% compared to the oxygen concentration of water at the surface. At depths of several hundred metres, dissolved oxygen is virtually absent due to reaction with iron-bearing minerals, and the Eh is believed to be controlled by the Fe(II)/Fe(III) couple [10]. UFCs in the deep repository will first be exposed to oxidising conditions, owing to the residual air trapped in the repository. During this period, aerobic corrosion of the UFCs would be possible. As time passes, the residual oxygen will be consumed by reaction with the minerals and the copper outer shell of the UFCs, and when full groundwater resaturation has occurred, the conditions will be fully anoxic with negative Eh values applying.

2.4.5 Microbial activity

Microbial activity is likely in the repository, as deep granitic groundwaters have been found to contain diverse, metabolically active bacteria [2]. Swedish and Canadian studies [2,14] have shown that the level of microbial activity close to the UFC will be determined by the availability of water in the bentonite. Below a certain thermodynamic water activity, a_w , (i.e. a thermodynamic term used to describe the effective concentration or availability of water in the bentonite clay [15]; the lower the value of a_w the lower the concentration of available water) the microbial populations cannot survive. The threshold a_w for microbial activity has been measured as 0.96 and studies have shown that this activity cannot be sustained in partially saturated or fully saturated bentonite. Furthermore the drying effect of the heat emitted from the UFCs will reduce the local water activity still further around the UFC and lead to a microbiologically inactive zone.

In the fully compacted state the bentonite blocks will have a water activity below that required to allow microbial growth [14]. SKB and OPG results indicate that the number of micro-organisms will decrease rapidly during swelling of the bentonite [2] and that only spores will remain after the full expected swelling pressure has been achieved. Eventually even viable spores will be eradicated from fully compacted buffer and repopulation by microbes is unlikely due to the small pore space in the bentonite clay. The fully compacted bentonite blocks will not allow permeation of new fresh microbes from outside the vicinity.

The radiation levels at the surface of the copper containers will be too low to reduce any microbial activity [14] because of the attenuation of the radiation by the thick inserts and the copper walls (see Section 2.4.6). The temperature of the UFCs will not be high enough to kill any naturally occurring microbial populations in the bentonite [14], although the populations will be reduced at the highest expected temperatures.

Microbial activity will be possible in the 50%/50% bentonite/sand mixture because the water activity will be higher than the threshold value but is unlikely that it will be sustainable within the 100% bentonite jacket of the UFC. Activity will likely be viable in the outer regions of the emplacement room and at the interface between the sealing materials and rock [14].

Microbial activity remote from the surface of the UFCs, particularly that of sulphate reducing bacteria during the early post-closure phase, may result in the formation of sulphide, which is a potential corrodent for copper (see Section 3.2.2.4). SRB activity correlates well with the presence of pyrite (iron sulphide) in geological formations at the Finnish sites [2], indicating that there had been long-term SRB activity leading to the reduction of sulphate to sulphide. SRB activity was observed at many of the depths investigated at Finnish sites. The concentration of sulphate in the bentonite can be up to tens of mmol/litre [2] and this could be a nutrient source for SRB activity.

The production of nitrite, ammonia and organic acids (e.g. acetate) by bacterial action is possible. These species are all known agents for the stress corrosion cracking of copper. Their probable concentration is taken into account when assessing the risk of stress corrosion cracking occurring [2].

2.4.6 Radiation levels

The gamma absorbed dose rates at the surface of a range of container designs were evaluated by Hanna [16]. If it was assumed that fuel bundles were cooled for a period of thirty years before packaging, and that the UFC had a lid thickness of 3.2 cm and a shell thickness of 2.5cm, the maximum predicted contact dose rate at the surface of the UFC was 0.908 Gy/hour.

2.4.7 External pressure on UFC

An external pressure will develop on the exterior of the UFC due to the hydrostatic head and bentonite swelling pressure. The normal isotropic pressure loading prior to glaciation is estimated to be ~15 MPa and the glaciation isotropic pressure loading is estimated to be up to ~45 MPa [17]. The pressure will cause the outer copper vessel to deform collapsing onto the inner carbon steel vessel.

3 Compatibility of Container Materials with DGR Environment

In this section the effect of the environments that will be experienced by the UFC on the various possible types of corrosion affecting the UFC are discussed.

3.1 CORROSION DURING MANUFACTURE, STORAGE, FILLING AND SEALING

During this period the container materials will undergo atmospheric corrosion. The critical relative humidity for corrosion is generally believed to be in the range 50-70% for metals [2];

below this value there is insufficient water to form a continuous electrolyte film on the surface. The elevated surface temperature of the UFCs will tend to reduce the local humidity below the critical relative humidity for corrosion. Very little corrosion would be expected during these operations with a corrosion rate of $<1 \mu\text{m yr}^{-1}$ expected [2], provided cleanliness is preserved. It is assumed that steps will be taken to prevent surface contamination.

3.2 CORROSION AFTER EMPLACEMENT IN THE REPOSITORY

3.2.1 Atmospheric corrosion prior to saturation

It is predicted that it could take several thousand years to resaturate the repository with groundwater [2]. While the bentonite is unsaturated the surfaces of the UFC will be exposed to a gaseous environment, whose water content would be expected to be low as moisture will be adsorbed by the bentonite clay. There will be a moisture gradient away from the surface, due to the heat produced by the UFC [2,18] and it is difficult to predict the concentration of water at the surface of the UFC. If the activity of water is low, the rate of gas phase oxidation will be slow, with the most probable corrosion product being Cu_2O . The predicted depth of attack due to this form of corrosion is negligible ($<1 \mu\text{m}$ over 10,000 years).

If sufficient moisture were available to form a thin continuous surface electrolyte film, which is unlikely due to the elevated temperatures of the UFC, the rate of corrosion could be more rapid given the rapid supply of oxygen through a porous sealing material layer and the possibility of forming concentrated solutions on the surface. There are no data for the atmospheric corrosion rate of copper at elevated temperatures but it is likely to be considerably less than $1 \mu\text{m yr}^{-1}$ [2,18], even with an unlimited supply of oxygen to the surface. The rate is likely to decrease with time due to the build up of a protective corrosion product film [18], which will mainly be formed from cuprous oxide.

Regardless of the exact corrosion mechanism, the total amount of corrosion would be restricted by the limited inventory of oxygen in the repository. On mass balance considerations, the maximum depth of corrosion would be expected to be approximately $300 \mu\text{m}$ over the entire surface of the container [2]. The actual value is likely to be considerably less than $300 \mu\text{m}$ because some of the oxygen would be consumed by reaction with the sealing materials. More detailed modelling shows that the total depth of attack due to corrosion in a thin liquid film is likely to be $<90 \mu\text{m}$ [18]. In summary, atmospheric corrosion is unlikely to have a significant impact on the integrity of the UFC.

3.2.2 Aqueous corrosion after saturation

After repository closure, groundwater will eventually permeate through the sealing materials and come into contact with the surface of the UFC. There are two main types of attack to consider, namely general corrosion and pitting corrosion.

3.2.2.1 General corrosion

On the basis of the Pourbaix diagrams for copper in a range of chloride concentrations, it can be seen that the potential range for thermodynamic stability extends from above the hydrogen evolution potential to more negative electrochemical potentials. This shows that in the absence of oxidising species copper will be stable. This has been demonstrated by the study of natural analogues, such as native deposits of copper. In the absence of oxygen the corrosion of copper will be extremely slow and the corrosion process will produce no hydrogen. The period of greatest risk to the copper is when it is exposed to oxygenated conditions, before oxygen is consumed by corrosion reactions or by reaction with surrounding minerals. The question that needs to be addressed therefore is whether the corrosion rate of copper during this period will be sufficiently low to provide the necessary protection of the interior of the UFC. This question has been addressed in the work carried out by OPG and partners. The detailed mechanism of general corrosion of copper and the supporting experimental evidence is thoroughly reviewed in reference [2] and there is no need to cover it in detail here. The key points are summarised below.

A mechanism has been proposed for the general corrosion of copper [13,19,20]. In essence, in highly oxygenated conditions and/or low chloride concentrations the final corrosion product contains cupric ions (Cu(II)) in the form of mixed chloride/hydroxide species ($\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2$), whereas in low oxygen concentrations and/or low chloride concentrations the final corrosion product will be predominantly cuprous oxide, $\text{Cu}(\text{I})_2\text{O}$. The predominant cathodic reaction is reduction of oxygen.

Corrosion experiments in bentonite buffer have shown that the corrosion rate is controlled by diffusion of oxygen and chloride through the bentonite [2]. In well aerated buffer material the corrosion rate is determined by the rate of transport of Cu(II) away from the surface and in low oxygen conditions it is controlled by diffusion of oxygen to the surface of the copper. Good agreement is claimed between measured values of E_{corr} and the values predicted on the basis of electrochemical models.

In the presence of Cu(II), the copper concentration near the surface is high, because it is readily absorbed by the bentonite, whereas in the presence of Cu(I) the concentration profiles are shallow, because its complexes are less readily absorbed by the clay.

Experiments on corrosion coupons and heated copper tubes embedded in bentonite have yielded average corrosion rates of $3 \mu\text{m yr}^{-1}$ [21]. Other experiments in compacted bentonite / sand mixtures have given corrosion rates in the range $30\text{-}50 \mu\text{m yr}^{-1}$ [22,23].

As oxygen within the repository is consumed the rate of general corrosion will decrease and eventually cease unless it is supported by reduction of water on a sulphide film.

A detailed mechanism has been proposed for the corrosion of copper and this has formed the basis of a predictive 1-dimensional model. The modelling shows that the oxygen available in the repository will be consumed after 670 years at which point the depth of attack due to general corrosion will be $11 \mu\text{m}$ [13,20,24].

3.2.2.2 Pitting corrosion

A number of experimental studies have been carried out to investigate the possible pitting of copper in a bentonite environment. In experiments carried out with bentonite-filled, groundwater-saturated, small-scale copper containers [25], Aaltonen did not find any pitting after 12 months' exposure at 80°C. The Eh value inside the container decreased to below -300 mV. In other experiments [21,26] on heated copper tubes in bentonite the attack was uneven, but there were no indications of pitting. Brennenstuhl et al [27] carried out experiments to investigate the corrosion of copper under deposits of bentonite. Some roughening of the copper surfaces was observed, but no pitting. It was concluded that although under-deposit corrosion was a possible degradation mechanism for UFCs its rate and extent would be very small and would cease when the oxygen in the repository has been consumed.

During resaturation, following a period in which a layer of bentonite next to the container was desiccated, the contact between the copper surface and the bentonite may not be uniform. Points of contact may be possible pit initiation sites [2].

Type I and Type II pitting of copper is well known to occur in potable water, as discussed in reference [13], but such pit morphologies have not been observed on copper embedded in bentonite.

The extreme value statistics approach has been applied to the prediction of the long-term behaviour of copper [2,13]. The probability of a pit exceeding 9 mm depth during a million years was predicted to be $<10^{-21}$; the probability of exceeding 5 mm in 1,000 years was $<10^{-11}$ [13].

3.2.2.3 Stress corrosion cracking

It is necessary to demonstrate that stress corrosion cracking (SCC) of copper is not possible in the predicted repository conditions. The two primary requirements for stress corrosion cracking to occur are tensile stresses exceeding a certain threshold value and the presence of an SCC agent. The possible causes of stress cracking of copper in the repository have been thoroughly reviewed in reference [2]. It is concluded [2] that the risk of SCC will diminish with time as the repository environment evolves. The most likely time for SCC to occur is during the early period when oxygen and other oxidants such as Cu(II) from corrosion of the UFC, will be present. A number of reasons are postulated for the chemistry of the environment not being conducive to SCC. These include:

- the short period over which oxygen will be available to support SCC;
- the high concentration of chloride is expected to lead to the formation of a $\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2$ film, which will cause general corrosion and prevent SCC;
- the concentrations of SCC agents will be too low.

Laboratory experiments are in progress to establish the risk of SCC over short time periods. It is suggested that the stresses and strain rates required for SCC will not occur on the copper

outer shell [17], even during the early stage of repository life, when the external hydrostatic pressures will increase and the copper outer shell will deform to fit around the inner steel component.

Finite element analysis (FEA) [17] has shown that the external isotropic pressures in a DGR would cause the copper shell to collapse on to the carbon steel inner vessel by plastic deformation. This would result in closure of the gap between the inner and outer containers. The FEA shows that the external surfaces in the regions around the welded lid closure would always be subject to compressive stress, thus minimising the risk of the initiation and propagation of SCC near the electron-beam lid closure welds. The FEA also predicts that there would be some regions of tensile stress in the surface of the UFC, but that there would always be sub-surface regions of compressive stress, which would tend to prevent stress corrosion cracks penetrating the full thickness of the walls. Furthermore, it is postulated that crack tip creep processes will relieve the stresses in the container surface and relieve crack tip stresses before crack advance can occur [2].

There are a number of unresolved questions in the field of SCC of copper, including the effect of electrochemical potential on the SCC of copper in ammonia solutions and how this relates to the corrosion potential in the repository, the effect of acetate on the SCC of copper and the effect of welds on SCC susceptibility. Further work may be needed in this area [2,28].

3.2.2.4 Microbially influenced corrosion

Microbially influenced corrosion (MIC) of copper can occur in conditions that are conducive to growth, of a number of different species, leading to increased general corrosion rates or SCC [14,29]. Possible causes of MIC are SRBs, which form sulphides, organic acids produced by anaerobes, and nitrogen-metabolising microbes, which form species such as nitrate, nitrites and ammonia. The nitrogen-containing species are potential agents for SCC of copper.

The effect of a range of possible microbial metabolites on the corrosion of copper in simulated groundwater was investigated by Jain and Ogundele [14]. Sulphide, and to a slight extent nitrite, were found to increase the corrosion rate of copper in simulated groundwater. The results with other metabolites were inconclusive, but did not appear to cause a significant increase in corrosion rate. The situation with polysaccharides may warrant further investigation.

A strong argument against the possibility of MIC of the copper containers is that microbes will not be viable in the region of the buffer surrounding the container, due to the low water activity (see Section 2.4.5), provided that the density of the bentonite is sufficiently high. Furthermore it is believed that cycled aerated-deaerated conditions are required for SRB-induced MIC [14] to occur.

In the longer-term it is postulated that the most important effect of microbial activity in the repository would be the production of sulphide by SRB activity in regions which are remote to the container [2,14]. Microbial activity will be possible in the 50%/50% bentonite/sand mixture because the water activity will be higher than the threshold value, but activity will be severely limited. The HS^- ions produced by the SRBs could diffuse through the sealing material and

react with the surface of the copper however, the maximum predicted corrosion rate due to this mechanism is only 0.001 $\mu\text{m}/\text{yr}$. On this basis the additional corrosion damage caused by microbial activity has been modelled and shown to be negligible [29].

3.2.2.5 Galvanic corrosion

Where two dissimilar metals come into contact in the presence of an electrolyte there is the possibility of enhanced corrosion occurring due to galvanic corrosion. If there were a leak of groundwater into the annulus between the inner and outer containers there would be a possibility of galvanic interactions between the copper and the carbon steel insert. This is an issue that is not addressed in the OPG corrosion studies because it is assumed that there will be no perforations in the outer copper container.

3.2.2.6 Crevice corrosion

Crevice corrosion in copper is believed to be self-limiting because it is driven by a concentration cell which disappears when the concentrations of Cu(I) ions at the anodic and cathodic sites become equivalent [2]. It is assumed that as the lid and base will be welded on to the body of the UFC, there will be no crevices exposed externally to the repository environment and therefore crevice corrosion is not an issue.

3.2.2.7 Weld corrosion

No preferential corrosion of welds has been found in tests to date [30].

3.2.3 Radiolysis effects

There is literature evidence that SCC of brasses can occur at high dose rates (e.g. $6 \times 10^{-4} \text{ Gy hr}^{-1}$) in moist air at ambient temperature, due to the formation of radiolysis species, particularly NH_3 and NO_2^- . However, no positive shifts in corrosion potential were observed when copper was exposed to a dose rate higher than that expected in the repository and it was concluded that there was no significant formation of oxidising radiolysis products at such dose rates [18].

Because of the low dose rate, due to the internal shielding by the carbon steel inner container, it is predicted that the concentrations of radiolysis products such as NO_2^- would not be sufficient to cause SCC of copper [18]. Combined crevice corrosion / U-bend tests [18] in an irradiated (5 Gy hr^{-1}) groundwater vapour phase did not result in any crevice corrosion or SCC, but general corrosion resulted in the formation of a green patina.

3.2.4 Natural analogues

Measurements on natural analogue artefacts support the corrosion rate estimates for copper and provide supporting evidence for the proposed corrosion mechanisms. Extreme value statistics have been applied to predict the maximum pit depth based on archaeological analogues (in particular the *Kronan* cannon and naturally occurring deposits of native copper [8]). They are consistent with laboratory data and modelling predictions.

3.3 CREEP DEFORMATION OF THE UFC COPPER SHELL

It is anticipated that the copper outer shell of the UFC will be subjected to creep deformation. At the expected maximum temperature of ~100°C and an external pressure of about 15 MPa, the external copper shell will undergo plastic deformation and creep while collapsing onto the inner load-bearing steel vessel. Through this process, the total elastic and plastic strain developed in the copper vessel must not exceed the minimum value for creep strain-to-failure of the material [31].

Extensive R&D efforts in the Swedish, Finnish and Canadian nuclear used fuel management programmes have been made to improve the creep properties of the copper material, to ensure that creep rupture can be ruled out as a failure mode for their used fuel containers. Creep data have been collected and analytical methods have been developed for predicting the long-term creep behaviour of the copper vessel in a DGR.

3.3.1 Development of a Copper Material with Improved Creep Ductility

Early SKB creep test results indicated that standard oxygen-free copper (ASTM UNS C10100) exhibited unacceptably low creep strain to failure. The poor creep ductility of this material was believed to be caused by segregation of sulphur (S) to the grain boundaries where it forms films that lead to embrittlement of the matrix [32]. A sulphur content in OFP copper higher than 7 ppm, was found to result in reduced creep strain to failure. SKB tests indicated that the addition of a small quantity of phosphorus (P) (40 to 60 ppm) resulted in significant improvement of creep ductility and an increase in the creep strain to failure [33-36]. It was also demonstrated that electron beam welding does not affect the creep ductility of OFP-Cu [37].

Based on the above reported results, an oxygen-free phosphorus-doped (OFP) copper material was developed by the Swedish and Finnish nuclear fuel waste management programmes [38,39]. This material conforms to the specifications of the oxygen-free copper ASTM UNS C10100 and meets additional requirements which include: S content < 8 ppm, P content from 40 to 60 ppm and grain size from 180 to 360 µm. The requirement for creep ductility has been specified to be a minimum strain-to-failure of 10% at a temperature of 100°C. This material is currently the reference corrosion-barrier material for used-fuel containers adopted by the Swedish, Finnish and Canadian used nuclear fuel programmes [39-41]. *#Note: Raiko and Salo 1999 was referenced here in the addendum but not given in the reference list – it has been assumed that it should have been 1996.*

3.3.2 Calculated Maximum Creep Strain

Structural analyses were carried out by OPG for the Canadian UFC case to estimate the extent of radial, axial and hoop creep strains of the copper vessel over the UFC design life in a DGR. The maximum accumulated creep strain in the copper corrosion barrier is determined by the size of the gap between the outer copper shell and the inner steel vessel and by the thickness of the copper shell [42]. Maximum strain values were calculated for the Canadian UFC, with a 25-mm-thick copper vessel, gap values of 1 mm and 1.75 mm. These values corresponded to

maximum accumulated creep strains of 4% and 7%, respectively, which are less than the minimum 10% elongation of the strain-to-failure requirement for OFP copper.

3.3.3 Development of Creep Data for OFP-Cu Material

Extensive creep tests were carried out by the Swedish/Finnish and Canadian programmes to improve understanding and the capability for predicting the long-term creep behaviour of the copper vessel of a UFC during its design lifetime in a DGR.

Extensive creep tests were carried out by SKB on OF copper material and OFP-Cu materials in air at relatively high temperatures (175-300°C) and stresses (60-160 MPa) [43-46]. Full creep curves were obtained in these tests, providing strain rates, creep strain and creep rupture data for the primary, secondary and tertiary stages of creep. These data are used to assess the effects of the material chemical composition, grain size, manufacturing and closure welding processes on the creep behaviour of OFP copper.

Copper creep studies in the Canadian programme included tests in air and in simulated Canadian groundwater, at temperatures in the range of 95 to 150°C and stresses from 20 to 100 MPa [47]. Creep rates in the range of 2.8×10^{-13} to $9.9 \times 10^{-11} \text{ s}^{-1}$ were obtained, which could be considered as approximate upper bound limits for creep processes of the UFC copper shell in a DGR. For the range of temperatures and stresses investigated, the dominant creep deformation mechanism is grain boundary controlled diffusion.

Detailed analyses of the OPG data and of SKB data obtained at higher temperatures (175-300°C) and stress conditions (60-160 MPa) indicate that the two data sets are complementary, and constitute a good creep data base for OFP copper, applicable over a relatively wide range of stresses and temperatures [47]. At the higher temperatures and stresses investigated by SKB, the steady-state creep behaviour follows a power-law relationship.

The test results obtained in the Canadian studies indicate that the simulated Canadian groundwater environment has insignificant effect on creep rates for the OFP copper, with respect to those observed in an air environment [47].

3.3.4 Methods for Predicting the Creep Behaviour of the UFC Copper Shell

Creep tests carried out at high temperatures (175-300°C) and stress (60-160 MPa) have indicated that OFP copper would meet the creep ductility requirement of having a minimum value of 10% strain-to-failure. However, it is known that the ductility of copper decreases with decreasing creep strain rate. In comparison with the experimental creep test conditions, the copper vessel would be subjected to lower temperatures and stresses in a DGR and the creep rates that would be experienced by the copper vessel are expected to be lower than those observed in the experimental creep tests. Analytical methods are required for extrapolating the experimental creep data to repository conditions.

An analytical approach that could potentially be used for quantifying the long-term creep properties of the container material was described by Dutton [48]. His study presented an

analytical scheme that adopted the Theta Projection Concept for the extrapolation of experimental creep data for long-term prediction of creep behaviour. Another analytical method has been described by Sandstrom [49] for extrapolating experimental creep strain data to longer times. These data could potentially be used for predicting the long-term creep behaviour of the copper vessel of a UFC.

4 Constructability and Inspection of UFCs

4.1 CONSTRUCTION OF A UFC

The proposed UFC is an assemblage of two major components:

- the inner carbon steel vessel with a bolted lid
- the outer copper shell with a welded lid.

The ability to manufacture and assemble these two items using currently available techniques is discussed in this section.

4.1.1 UFC Copper Shell

Trials to manufacture copper containers with wall thicknesses of 50 mm have been carried out by SKB [5]. This work has extended to the manufacture of copper containers with 30 mm thick walls. Further developments in this area on behalf of SKB are presently examining the benefits of producing seamless tubes by extrusion and pierce and draw techniques, rather than the hot rolling, bending and longitudinal electron beam welding investigated in earlier work [50,51]. Initial problems associated with the integrity of the longitudinal welds of these tubes have been reduced as a result of further development work. Carrying out the welding process at reduced pressure rather than high vacuum, together with using a reduced copper wall thickness, may well result in the hot roll and weld process providing an acceptable solution. Lids and bottoms for containers manufactured using this method have been made by forging continuous-cast bars and this results in a larger grain size than that produced in the container walls.

In both the Swedish and Finnish container development programmes, electron beam welding is a well-established technique for attaching the copper lid to the container shell [51,52,53]. However, friction stir welding has been investigated as an alternative sealing method, with further development work currently being undertaken [5,8].

During the preparation of this report discussions have taken place between CTECH and two potential manufacturers of copper tubes, in order to determine the viability of manufacturing seamless copper containers to the dimensions required for the proposed UFC design. The two techniques considered were the extrusion process, which produces an open ended tube, and the pierce and draw method, which provides a tube with an integral base and open top.

Wyman Gordon Limited of Steventon, Scotland, manufacture extruded tubes and have provided a number of copper extrusions for SKB using the process described in [5]. Mr Bob Collins of Wyman Gordon confirmed that the company has the capability to extrude tubes with an internal diameter of 1092 mm, external diameter of 1194 mm and a length of 3780 mm [54], to allow machining to the required rough machining dimensions. The base of the container, made by forging continuous-cast bars, would be rough machined and then welded to the tube, followed by final machining. The container lid would be made by forging continuous-cast bar and then machining to suit the individual UFC shell, to be ready for welding to the container following loading with used fuel baskets within the DGR facility.

Vallourec and Mannesmann Tubes (V & M Tubes) have manufactured copper tubes with integral bases, using the pierce and draw method, to similar dimensions as those of the proposed UFC design, for Posiva and SKB [5]. During discussions between CTECH and Messrs Wolfgang Grummer and Douglas Crooks of V & M Tubes, confirmation was given that V & M Tubes have the capability at their Reisholz Works in Germany to pierce and draw copper tubes to the required dimensions. In addition, similar tubes had been made for Posiva and SKB; the entire surface of the tubes has been examined using ultrasonic techniques. Further development work is required to ensure that the grain size of the integral base is kept within acceptable limits and V & M Tubes have made proposals to SKB on how this may be achieved. V & M Tubes also have the capability to machine the drawn copper tubes and have undertaken trials for SKB using different machine speeds and feeds to determine their effect on tube surface finish. Tolerances on the finished container required by SKB were achieved within the machining capabilities available to V & M Tubes.

As a result of the above discussions and the work undertaken by SKB and Posiva, CTECH believe that it is viable to produce the proposed UFC copper shell design utilising one (or all) of the manufacturing methods described. The selection of the method to be employed may be influenced by the limited number of suppliers that are able to provide extrusions or pierce and draw tubes. However, this may be balanced by the ability to achieve repeatable high quality longitudinal welds using the roll and weld method.

4.1.2 UFC Inner Carbon Steel Vessel

Costs have been obtained from Babcock & Wilcox (B & W) of Cambridge Ontario [55] to manufacture steel inner vessels to a range of different dimensions. It is assumed that B & W have satisfied themselves that these configurations can be manufactured successfully, giving confidence that the same techniques could be used for the current UFC inner vessel design.

During discussions with V & M Tubes it became apparent that the company had the capability to manufacture a carbon steel vessel sized to fit within the proposed UFC. As with the outer

copper container, V & M Tubes believed that the pierce and draw method was a viable method to produce a steel tube with an integral base to the dimensions required. V&M Tubes had previously produced tubes for the German 'Pollux' container that had finished dimensions of 1012 mm outside diameter, 690 mm inside diameter and 5086 mm long and which weighed approximately 17.2 tonnes.

4.1.3 Summary

The assembly of the two major components of the proposed UFC will require accurate handling of the individual components due to the small dimensional clearance between the two items. V & M Tubes have confirmed that diametral dimensions of copper tubes with a finished wall thickness of 50 mm will remain stable for a considerable period, therefore not presenting additional problems during the UFC assembly process. However, adopting a thinner wall for the copper tube may result in the open-end diameter of the tube distorting with time, due to its self-weight when laid horizontally, or during transport. Such distortion may be overcome by either, assembling the two components soon after the copper tube is finish machined, or introducing temporary support within the open end(s) of the copper tube.

From information reported by OPGI, together with discussions held with Wyman Gordon Limited and V & M Tubes, it is concluded that a number of techniques are available that may be used to manufacture UFCs to the current design. Development work will be required to ensure that repeatable, good quality welds can be achieved for the copper container, either using electron beam or friction stir welding techniques.

4.2 INSPECTION OF UFC

Validation of the UFC closure weld will be carried out using both radiography and ultrasonic non-destructive testing (NDT) techniques. It is proposed to deploy both systems from within the same DGR used fuel packaging plant shielded cell into which the lidded end of the UFC will be raised. Because the ultrasonic testing method requires the introduction of a liquid coupling medium, the weldment will be radiographed prior to deploying the ultrasonic equipment. To simplify the equipment necessary within the cell to undertake both NDT operations the UFC will be rotated past the inspection heads. Furthermore, the equipment within the cell will be kept to a minimum so as to not to expose it to damaging radiation fields. For this reason, as much NDT ancillary equipment as possible will be located outside the cell to provide ease of access for hands on maintenance.

The UFC copper lid weld will be inspected for porosity, a measure of weld quality, and possible sites for localised corrosion [2] and other defects such as cracks. Radiography [8] will be used to detect weld pores, however, the size of pore that can be detected with this type of inspection technique is limited. To extend the scope of weld defects that can be identified, an ultrasonic phase array system will be used. Although this system can detect small pores and cracks, reflection of the ultrasonic beam from large grains within the weld material presents difficulties. The feasibility of using an ultrasonic array technique to inspect container welds for discontinuities that lack volume (e.g. lack of penetration) has been demonstrated [56].

Radiography and ultrasonic inspection are complementary techniques [57]. SKB are also working on the use of the eddy current technique to detect near-surface discontinuities [8].

The grain structure in the lid and base area of the copper container is affected by the fabrication and sealing processes employed. The impact of the grain structure on the ultrasonic inspection of the copper container will need to be investigated further during future container development.

5 Conclusions

The main conclusions from the review of the metallurgical and constructability aspects of the proposed UFC design covered by this report are as follows.

1. Based on the existing body of knowledge, there is a high level of confidence in the use of copper as the outer shell material for the UFC, in terms of its likely corrosion resistance as well as the methods available for its construction and subsequent inspection.
2. Areas of further work to confirm the suitability of copper as the UFC outer shell material have been identified. These include the effect of chemical conditions on copper SCC susceptibility, the effect of grain size on UFC inspection and the significance of microbially-induced corrosion [2].
3. From a review of the metallurgical information currently available the proposed UFC design is viable.

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