

TECHNICAL REPORT 02-11

Canister Options for the Disposal of Spent Fuel

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This report was prepared on behalf of Nagra. The viewpoints presented and conclusions reached are those of the author(s) and do not necessarily represent those of Nagra.

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Abstract

Canister design concepts for the disposal of spent fuel in repositories in both crystalline and Opalinus Clay host rocks are proposed, based on a review of the functional and performance requirements for such canisters, proposed design criteria and an assessment of repository conditions and their impact on long-term performance of possible canister materials. Two proposed canister lifetime targets of 1000 and 100 000 years are considered, based on experience from a variety of performance assessment studies in a number of countries, including Switzerland. The two canister options proposed and evaluated in detail that could meet the lifetime requirements are a thick-walled (~15 cm) carbon steel canister and a composite canister with a copper external shell and a cast iron insert to provide structural integrity (the proposed SKB/Posiva canister).

The cast steel canister is at a conceptual design stage, thus from the manufacturing perspective, only the basic feasibility of fabricating a canister shell has been considered. For an evaluation of the long-term integrity, the structural behaviour of the shell under isotropic loading conditions in the repository has been considered, along with a detailed assessment of the impact of various corrosion mechanisms on canister lifetime. The corrosion evaluation indicates that the short (some decades) aerobic phase of the repository would lead to very limited general and pitting corrosion (approximately 1 cm). Subsequent anaerobic corrosion is expected to occur at a rate of $1 \mu\text{m a}^{-1}$. Evaluation of other mechanisms such as microbial corrosion, stress-corrosion cracking and hydrogen damage indicates that they are not expected to lead to canister breaching, thus a lifetime for a steel canister is expected to be at least 10 000 years, well in excess of the target lifetime of 1000 years. The structural analysis indicates that, for the expected total depth of corrosion of 2 cm, the canister has sufficient strength that structural loads would not lead to breaching within 10 000 years.

The corrosion assessment of the copper canister for crystalline and Opalinus Clay repository conditions suggests a lifetime of at least 100 000 years, in line with Swedish and Finnish assessments.

Zusammenfassung

Im vorliegenden Bericht werden Auslegungskonzepte für die zur Entsorgung von abgebranntem Brennstoff in einem Tiefenlager in den Wirtgesteinen Kristallin und Opalinuston vorgesehenen Behälter vorgestellt. Diese Konzepte basieren auf funktionellen und sicherheitstechnischen Anforderungen an solche Behälter, vorgeschlagenen Auslegungskriterien sowie einer Evaluation der im Tiefenlager herrschenden Bedingungen und deren Auswirkung auf die Langzeitsicherheit potenzieller Behältermaterialien. Dabei werden zwei mögliche Zielsetzungen bezüglich der Behälterlebensdauer, d.h. ein Zeitraum von jeweils 1000 und 100 000 Jahren betrachtet, basierend auf den Erfahrungen aus einer Vielzahl von Sicherheitsstudien, die in zahlreichen Ländern, darunter auch der Schweiz durchgeführt wurden. Bei den beiden vorgeschlagenen und im Detail evaluierten Behälterooptionen, die die Anforderungen an die geforderte Lebensdauer erfüllen könnten, handelt es sich einerseits um einen dickwandigen (~ 15 cm) Kohlenstoffstahlbehälter und andererseits um einen sogenannten Composite-behälter, bestehend aus einer äusseren Kupferhülle und einem Gusseisen-Einsatz zur Gewährleistung der strukturellen Integrität (von SKB/Posiva vorgeschlagener Behälter).

Der Kohlenstoffstahlbehälter befindet sich noch im Stadium eines konzeptuellen Entwurfs, und somit wurde aus Sicht der Herstellung lediglich die grundsätzliche Machbarkeit der Fabrikation einer Behälterschale betrachtet. Für die Abschätzung der Langzeitintegrität ist das Strukturverhalten eines solchen Behälters unter isotropen Belastungszuständen im Tiefenlager betrachtet und der Einfluss verschiedener Korrosionsmechanismen auf die Behälterlebensdauer evaluiert worden. Diese Evaluation des Korrosionsverhaltens zeigt, dass die kurze (einige Dekaden dauernde) aerobe Phase im Tiefenlager zu einer sehr begrenzten generellen Korrosion sowie zu Lochfrass (ca. 1 cm) führen würde. Es wird erwartet, dass nachfolgende anaerobe Korrosionserscheinungen eine Rate von $1 \mu\text{m a}^{-1}$ aufweisen. Die Evaluation weiterer Mechanismen, wie mikrobielle Korrosion, Stresskorrosionsbrüche und Korrosionsschäden durch Wasserstoff zeigt, dass solche erwartungsgemäss nicht zum Behälterversagen führen werden und somit die Lebensdauer eines Stahlbehälter mit mindestens 10 000 Jahren angegeben werden kann. Diese Zeitspanne liegt deutlich über der angestrebten Behälterlebensdauer von 1000 Jahren. Die Strukturanalyse zeigt, dass der Behälter gegenüber der erwarteten Korrosionstiefe von insgesamt 2 cm eine ausreichende Wandstärke aufweist, so dass Strukturbelastungen nicht zu einem Versagen innerhalb von 10 000 Jahren führen würden.

Die Evaluation der Korrosion eines Kupferbehälters unter Tiefenlagerbedingungen im Kristallin sowie im Opalinuston ergibt in Übereinstimmung mit schwedischen und finnischen Analysen eine Behälterlebensdauer von mindestens 100 000 Jahren.

Résumé

Ce rapport présente des propositions pour la conception de conteneurs destinés au stockage des assemblages combustibles usés dans un dépôt situé soit dans des roches cristallines, soit dans les Argiles à Opalinus. Ces propositions sont basées sur les exigences fonctionnelles et les performances requises pour ce type de conteneur, différents critères de conception définis dans l'état actuel des connaissances, ainsi que l'étude des conditions de stockage et de leur influence sur les performances à long terme des matériaux envisagés pour les conteneurs. Sur la base de plusieurs études de performances des matériaux réalisées dans différents pays, notamment en Suisse, des durées de vie de 1000 et 100.000 ans ont été fixées comme objectifs pour l'évaluation. Deux types de conteneurs respectant ces contraintes de durabilité sont analysés en détail: d'une part un conteneur en acier au carbone à parois épaisses (~15 cm) et d'autre part un conteneur composite comprenant un emballage en fonte, assurant l'intégrité structurelle, inséré dans une enveloppe en cuivre (le conteneur envisagé par SKB et Posiva).

Le conteneur en acier se trouve en phase de conception, si bien que seule la fabrication de l'enveloppe externe a été prise en compte pour l'étude de faisabilité industrielle. En ce qui concerne l'évaluation de l'intégrité à long terme, on a étudié le comportement structurel de l'enveloppe soumise à des contraintes isotropiques dans le dépôt, ainsi que l'influence de divers mécanismes de corrosion. L'analyse des processus de corrosion indique que durant la courte phase aérobie du dépôt (quelques dizaines d'années), on observerait une corrosion générale et par piqûres très limitée (sur environ 1 cm). Ensuite, la corrosion au cours de la phase anaérobie devrait être de l'ordre de $1 \mu\text{m a}^{-1}$. L'analyse d'autres mécanismes – tels que la corrosion microbienne, la corrosion sous contrainte et l'endommagement engendré par l'hydrogène – montrent qu'ils ne devraient pas entraîner la rupture des parois des conteneurs, dont la durée de vie atteindrait de ce fait au moins 10.000 ans, ce qui dépasse de loin l'objectif fixé de 1000 ans. L'analyse structurelle montre que, pour une corrosion affectant le conteneur sur 2 cm, la résistance de celui-ci serait suffisante pour que, sur une durée de 10.000 ans, les parois restent étanches malgré les charges structurelles.

L'étude de l'impact des mécanismes de corrosion sur le conteneur en cuivre, dans un environnement de dépôt composé soit de roches cristallines, soit d'Argiles à Opalinus, permet d'estimer sa durée de vie à 100.000 ans au minimum, ce qui correspond aux résultats obtenus lors des évaluations réalisées en Suède et en Finlande.

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1 Introduction

This study presents canister design options for the disposal of spent fuel (SF) from Swiss power reactors. The canister options discussed have been evaluated with respect to their potential application in a repository in Opalinus Clay (NAGRA 2002a) or crystalline rock (NAGRA 1994). The functional requirements of a disposal canister are outlined, followed by a discussion of the specific performance requirements, proposed design criteria and expected long-term performance. Canister design options are presented, focusing principally on a thick-walled canister of carbon steel, and a composite canister with a copper external shell and a cast iron insert, based on the SKB/Posiva canister design concept (WERME 1998, RAIKO & SALO 1999). The prospects for using materials other than steel for canister construction in order to reduce hydrogen gas generation are also briefly discussed.

For the carbon steel canister, a design concept has been developed by Nagra. At this early stage of development of the repository program, the design is conceptual in nature, with the feasibility of manufacturing the canister body and the basic structural performance (isotropic loading of the body) evaluated to establish a feasible design concept. Detailed investigations of welding, inspection, stresses in the weld region and behaviour under anisotropic loading scenarios have not yet been performed, because it is judged that good industrial experience with carbon steel will lead to a satisfactory detailed design that deals with all these considerations during an optimisation stage. As a result, these items are not examined in detail. In contrast, the expected short-term and long-term corrosion performance aspects of a carbon steel canister are thoroughly examined, based largely on data that has been published since the Kristallin-1 study (NAGRA 1994). The corrosion assessment provides the basis for the corrosion allowance, which, when added to the minimum wall thickness for structural integrity and a subjective estimated additional thickness that provides a factor of safety, gives the total wall thickness. The minimum lifetime target is 1000 years for the steel canister, although the lifetime assessment presented here indicates an expected lifetime of at least 10 000 years.

For the concept based on a copper shell with a steel insert, the design, structural performance and long-term performance evaluations are based on Swedish and Finnish studies (ANDERSSON 2002, WERME 1998, KING et al. 2001). The latter study provides a detailed evaluation of the expected corrosion performance of a copper canister, including a summary of published canister lifetime predictions. Because of this, and because the range of repository chemistry conditions considered in these assessments largely encompass those expected in a Swiss crystalline or Opalinus Clay repository, the discussion in the present report of the copper canister performance is brief. This copper canister design is at an advanced stage, such that many of the long-term (> 100 000 year lifetime) corrosion and structural performance issues are well understood and confident lifetime predictions can be made. In addition, the technology of fabrication, welding and inspection of the canister are sufficiently developed that there is confidence in the overall technical feasibility of the design.

Costs of the two canister concepts are not addressed, but they are not expected to be the dominant factor in selecting a reference design.

2 Design Development Process

The development of a design for a SF disposal canister requires that a proper design process be established and followed. The stages of the process are as follows:

1. determination of functional requirements
2. determination of performance requirements
3. development of design criteria
4. preliminary development and review of design concepts that are likely to meet the design criteria and assessment of potential long-term performance,
5. detailed assessment of selected design(s), including fabrication and structural loading studies
6. design review to select a preferred concept for detailed design
7. design optimisation through pilot-scale fabrication trials
8. final design

The present study deals with the first four stages outlined above. In order to provide background for a discussion of canister performance, the repository concepts are first briefly outlined.

3 Repository design concepts

3.1 Kristallin-I

The crystalline repository concept has been described in detail in NAGRA (1994). It involves tunnel emplacement of steel canisters containing vitrified HLW in a repository at a depth of 900-1200 m in crystalline basement rocks in Northern Switzerland. The crystalline basement in Northern Switzerland consists of gneisses with granitic intrusions, and is overlain by thick (several hundred metres) Mesozoic sediments. The low-permeability domain of crystalline rock exists below about 900 m. Groundwater salinities are expected to range from 0.003 - 0.2 mol Γ^{-1} Cl. The canister emplacement concept is shown in Figure 1. The steel HLW canisters, with a design lifetime of 1000 years, are surrounded by bentonite blocks having a dry density of 1.7 Mg m^{-3} and an initial moisture content of $\sim 15 \%$. A total of 2693 canisters of HLW were assumed in the Kristallin-1 study. The emplacement tunnels have a diameter of 3.7 m. Although the disposal of SF was not discussed in Kristallin-1 (NAGRA 1994), it is envisioned that the same emplacement concept could be used for SF disposal canisters.

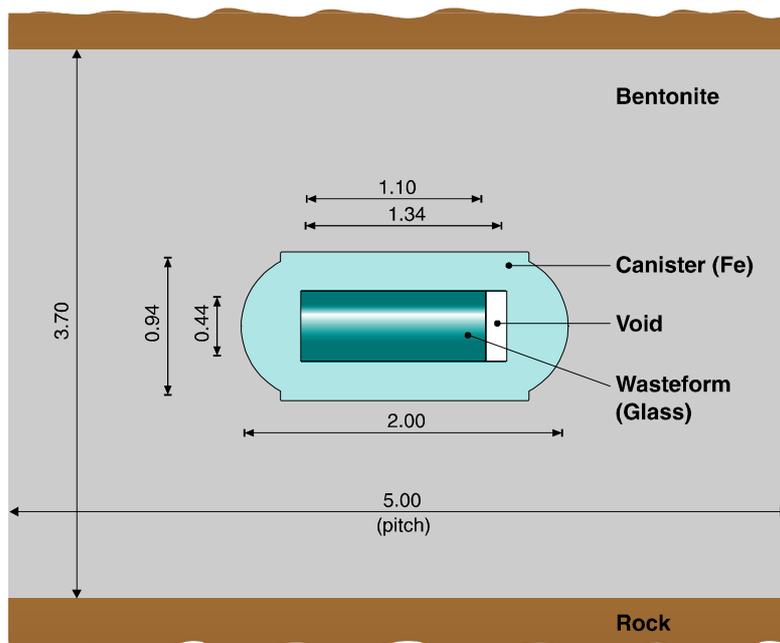


Fig. 1: Cross-section of Kristallin-I emplacement tunnel for HLW canisters

3.2 Opalinus Clay

Opalinus Clay is a Jurassic claystone formation of ~ 100 m thickness found at a depth of 450 to 850 m in the Zürcher Weinland region of northern Switzerland (NAGRA 2002a). The formation is characterised by a low porosity ($\sim 12 \%$) and an extremely low hydraulic conductivity ($2 \times 10^{-14} \text{ m s}^{-1}$). The SF/HLW waste emplacement tunnels, with a diameter of 2.5 m, would be excavated in the centre of the Opalinus Clay formation. These tunnels are expected to be self-

supporting during the operational stage of a few years, based on experience with excavations in Opalinus Clay. A longitudinal section through the SF and HLW emplacement tunnels is shown in Figure 2. Highly compacted bentonite blocks support the canisters and the region around the canisters is backfilled with granular bentonite using a pneumatic or conveyor system. The granular backfill has a moisture content of $\sim 2\%$ (NAUNDORF & WOLLENBERG 1992), and comprises $\sim 80\%$ by volume very dense pellets ($\sim 2.1 - 2.2 \text{ Mg m}^{-3}$) and 20% bentonite powder. Upon emplacement, the material is expected to have an average dry density of 1.5 Mg m^{-3} (NAGRA 2002a). The design and operations of the repository are described in detail in NAGRA (2002b).

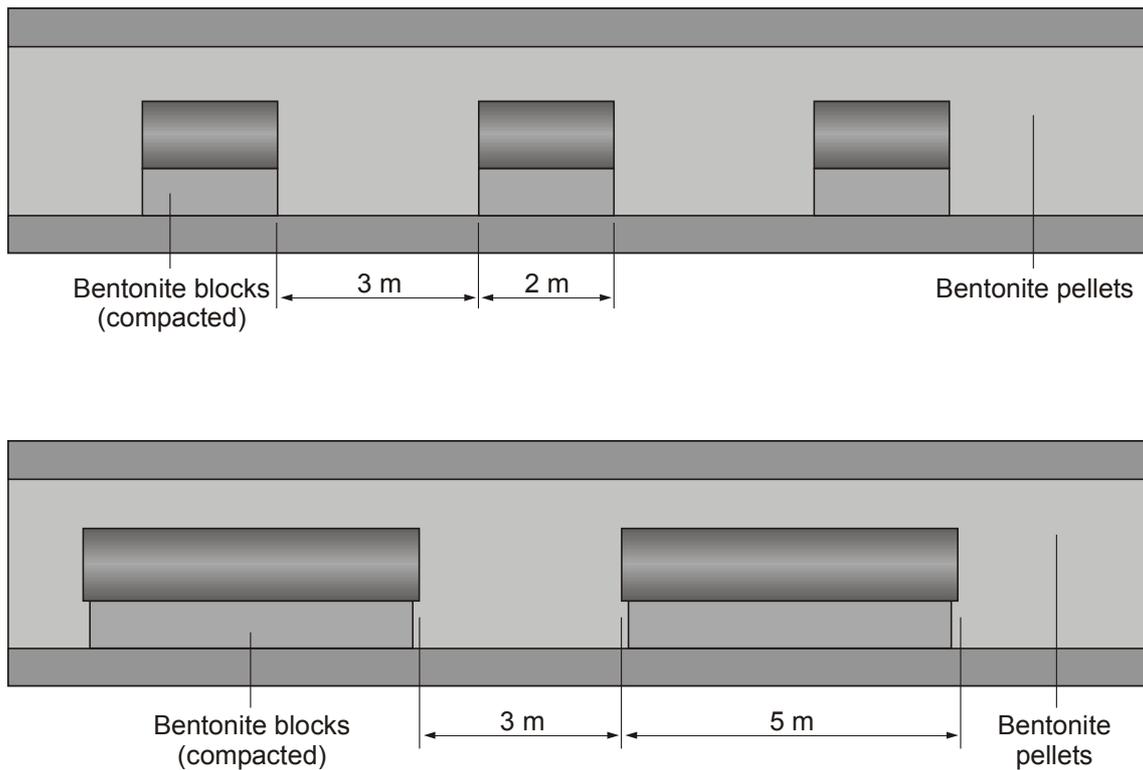


Fig. 2: Longitudinal section through disposal tunnels in Opalinus Clay for canisters of HLW (top) and SF (bottom) at completion of canister and backfill emplacement. Tunnel diameter is 2.5 m

4 Functional Requirements

The most basic functional requirements of the SF disposal canister are to contain and isolate the SF so that safety is maintained at a high level for workers and the public during fuel encapsulation, transport and handling of canisters, emplacement of canisters in the final repository and in terms of meeting the long-term performance requirements of the disposal system. The canister design achieves this by providing

- a method of isolation of the SF that permits safe handling, interim buffer storage and emplacement in the repository,
- a structural barrier against the hydrostatic pressures and other mechanical forces that might compromise containment, by providing an absolute barrier to radionuclide release for the design lifetime of the canister, and
- a transport barrier against contaminant release to the groundwater after eventual breaching, by inhibiting the transport of radionuclides through the breached part of the canister.

The canisters must be designed for all SF types produced by Swiss power reactors, including UO_2 and MOX fuels from BWR and PWR reactors. The quantities of SF to be encapsulated and their typical characteristics are given in Table 1, based on an assumption of operation of the reactors over a period of 60 years. The data in Table 1 are for reference SF, based on detailed review of the types of SF assemblies, their dimensions, burnups and other characteristics, given by McGINNES (2002). For 60 years power plant operation, 2065 canisters are required for encapsulation of the quantities of fuel given in Table 1, comprising:

- 935 canisters of BWR fuel, with 9 assemblies per canister,
- 680 canisters of PWR fuel, with 4 assemblies per canister, and
- 450 canisters with UO_2 and MOX fuel combined, (3 UO_2 assemblies and 1 MOX assembly).

A criterion of a maximum of 1500 W per canister at the time of emplacement has been selected in the Opalinus Clay study (NAGRA 2002a) based on consideration of the temperatures that would be reached by the various components of the disposal system. This is discussed in more detail in Section 6.2. Based on the decay heat data of McGINNES (2002), JOHNSON et al. (2002) show that, for BWR and PWR UO_2 SF assemblies with the average burnups in Table 1, the canister loadings of 9 BWR or 4 PWR assemblies will give a canister heat output of less than 1500 W at the time of emplacement with 40 years of cooling. In the case of MOX fuel with its somewhat higher heat production, loading of 1 MOX assembly with 3 PWR assemblies is the preferred arrangement. The latter canisters would require 55 years of cooling to remain within the 1500 W constraint at the time of emplacement. Alternatively, other measures such as combining the MOX assembly with three somewhat lower burnup UO_2 fuel assemblies is possible.

Tab. 1: Quantities and characteristics of reference power reactor fuels for direct disposal, based on a postulated operating lifetime of 60 years for Swiss nuclear power plants (NAGRA 2002a)

	BWR UO ₂ Fuel	PWR UO ₂ Fuel	PWR MOX Fuel	BWR MOX Fuel
Total mass (tIHM)	1491	1581	128	17
Avg. Burnup (GWd/tIHM) ¹	48	48	48	48
Mass ² per FA ³ (t)	0.27 and 0.294	0.48 and 0.666	0.482 and 0.655	0.289
FA length ¹ (m)	4.48	4.29 and 3.52	3.52 and 4.29	4.48
FA width ¹ (m)	0.139 × 0.139	0.215 × 0.215 0.198 × 0.198	0.215 × 0.215 0.198 × 0.198	139 × 0.139

¹ IHM = initial heavy metal (U + Pu)

² Masses and dimensions are for typical fuel assemblies. Detailed characteristics vary according to the fuel design and are given by McGINNES (2002).

³ FA = fuel assembly

5 Performance Requirements

5.1 Long-term Safety Requirement

It is generally agreed that the required canister lifetime is a derived criterion, established on the basis of total system performance assessment calculations. Nonetheless, there is recognition on the part of regulators and repository proponents that a minimum canister lifetime of ~1000 years is required, because of the high hazard potential arising from fission products during this time (e.g. HSK 1993). Furthermore, such a time frame ensures that the period of resaturation of the near field, which involves high temperatures and high thermal gradients, will be over by the time canister breaching by corrosion might occur. The adoption of a minimum lifetime of ~1000 years thus avoids the requirement of modelling radionuclide transport during this transient phase, when there may be significant uncertainties in transport parameters.

Canister lifetime targets ranging from 1000 to 100 000 years have been proposed in various national programs for HLW and SF disposal. The broad range relates to different judgements made about the relative contributions to safety of the different components of the disposal system. In Sweden (SKB 1999), Finland (VIENO & NORDMANN 1996) and Canada (WIKJORD et al. 1996), copper canisters with a design lifetime of 100 000 years have been proposed for SF disposal. In other assessment studies related to crystalline rock, minimum lifetimes of 1000 years were proposed for spent fuel disposal in titanium canisters (GOODWIN et al. 1994) and for HLW disposal in carbon steel canisters (NAGRA 1994, JNC 2000). In the recent Belgian study (NIROND 2001), both SF and HLW are assumed to be in stainless steel canisters with a minimum lifetime of 1000 years. In all cases, the canisters provide sufficient isolation in the context of the studies such that doses are well below the regulatory requirements.

The effect of the design lifetime of SF canisters on system performance has been examined in the context of the Opalinus Clay safety case (NAGRA 2002a). The maximum dose from ^{129}I , the principal dose contributor, decreases very little as canister lifetime is increased from 10^3 to 10^5 years, because of the very long half-life of this nuclide, although the time of peak dose is obviously affected. A distribution of canister breaching times over time can significantly reduce the peak dose relative to that for the unrealistic assumption that all canisters are breached simultaneously. However, this approach is not generally taken in performance assessment calculations, because the distribution of breaching times is difficult to quantify, thus adopting a poorly justified broad distribution of breaching times may lead to underestimation of dose.

The results of Nagra studies (NAGRA 2002a) suggest that for SF disposal in a low permeability host rock such as Opalinus Clay, containment for 1000 years may provide a significant margin of safety when seen in the context of total system performance assessment. The influence of a much longer canister design lifetime, such as 10^5 years, has also been evaluated in the case of a repository in Opalinus Clay (NAGRA 2002a), thus the prospects of attaining such performance are addressed in some detail in the present study. For the case of a repository in crystalline rock, no formal performance assessment study has been made by Nagra for the case of SF disposal. The materials and design options to achieve the 10^3 and 10^5 year targets are discussed in Sections 7.1 and 7.2.

5.2 Industrial Safety Requirements

Requirements for safety during canister fabrication and fuel encapsulation will be determined by present-day radiation protection and other industrial safety criteria. It is obvious that such criteria will have to be met at all stages. In the present study, it has been assumed that fuel

elements should be emplaced in canisters with no treatment or conditioning (i.e., no rod consolidation or fuel cutting) to avoid complex procedures and to limit the production of secondary wastes.

Developments in the SKB Encapsulation Plant Project (ANDERSSON 2002) will provide essential information on issues such as canister fabrication, handling, reliability, welding and inspection. These developments should be closely followed in relation to evaluating the design concepts discussed in the present study.

5.3 Retrieval

Retrievability of canisters needs to be considered in terms of the operational and post-closure phases of the repository. It is clearly essential that retrievability be possible during the operational phase (15 to 30 years). The concept for emplacement of SF and HLW canisters in Opalinus Clay envisions that bentonite backfill is emplaced around the canisters as each canister is placed and that emplacement tunnels are sealed at the ends with highly compacted bentonite. In the case of crystalline rock, tunnels may have a larger diameter (up to 3.7 m vs. 2.5 m for the reference case for Opalinus Clay). In both cases, excavation into the bentonite to retrieve canisters is not intrinsically difficult, thus retrievability is considered possible as long as canisters remain structurally sound (Nagra 2002b). This implies a period of easy retrievability of the order of at least some hundreds of years. Furthermore, the pilot repository concept incorporated in the Opalinus Clay study (NAGRA 2002a) permits development and testing of retrieval procedures, if required.

5.4 Projected Repository Conditions and Service Limits

The repository conditions in crystalline rock and Opalinus Clay in terms of the time frame of resaturation, temperature, pressure, mechanical loads on the canister and groundwater composition are summarized in Table 2 and further discussed below. Given the uncertainty regarding the final disposal medium and the depth, the canister design concepts are evaluated here with respect to each of these potential repository environments.

Resaturation Time

There is considerable uncertainty in the permeability of crystalline rock in which a repository might be sited. The calculations of ANDREWS et al. (1986), which did not consider thermo-hydraulic coupling, suggest that for tunnel emplacement, saturation of the bentonite is likely to occur within 100 to 1000 years for rock with an average hydraulic conductivity of $10^{-12} \text{ m s}^{-1}$. For the situation in which a high conductivity (10^{-9} m s^{-1}) fracture zone intersects the tunnel, the resaturation time could decrease to ~ 10 years. The coupled thermo-hydro-mechanical modelling of BÖRGESSON & HERNELIND (1999) illustrates that it takes only a few years to saturate blocks of highly compacted bentonite (HCB) emplaced at an initial degree of saturation of 0.6 in the case of relatively permeable granite. Although the latter results are for borehole emplacement, they may also be representative for the case of tunnel emplacement of canisters and bentonite in relatively permeable crystalline rock. The possibility that buffer saturation times may be quite short in crystalline rock and the initially rather high moisture content of precompact blocks (relative to the granular bentonite backfill proposed in the Opalinus Clay repository design) suggests that no credit can be given to a delay in the initiation of corrosion processes.

In the case of Opalinus Clay, the low hydraulic conductivity ($\sim 2 \times 10^{-14} \text{ m s}^{-1}$) and the absence of conductive fractures leads to slow transport of water into the near field (NAGRA 2002a). Furthermore, the use of a low moisture content (2 %) granular bentonite as a backfill around the canisters, combined with a high temperatures at the canister surface, may lead to some delay in the time at which corrosion might initiate, although, as discussed in Section 6, the likelihood of a protracted partially saturated period may make aspects of the corrosion assessment more complex.

Tab. 2: Expected Conditions in Crystalline Rock and Opalinus Clay Repositories*

Host rock	Repository depth ¹ [m]	T at repository depth [°C] Ambient ² Max ³	Hydrostatic Pressure ⁴ [Mpa]	Lithostatic Pressure ⁵ [Mpa]	Bentonite saturation time	Bentonite Swelling Pressure ⁶ [Mpa]	Salinity ⁷ Cl [mol /l] Typ. / Max
Crystalline	900 -1200	45 - 55 ~150	9 -12 + glacial: 5	-	> 10 years	4 - 15	3×10^{-3} 0.2
Opalinus Clay	450 - 850	30 - 45 ~160	4.5 - 8.5	15 - 22 + glacial: 5	> 100 years	2 - 4	0.1 0.3

* sedimentary rock would creep and the canister would eventually experience the combined lithostatic and hydrostatic loads; at some later time the load could increase by a further 5 MPa from glaciation; whereas the canister would not experience a lithostatic load in crystalline rock

¹ crystalline: Project Gewähr: 1200 m (NAGRA 1985); Kristallin-I: "about 1000 m" (NAGRA 1994); Opalinus Clay: 450 - 850 m (NAGRA 2002a)

² ambient temperature estimated from NAGRA (2002a)

³ max. canister surface temperatures from JOHNSON et al. (2002) and SATO et al. (1997), based on an initial thermal loading of 1500 W/canister at emplacement

⁴ assumption: max. thickness of ice sheet: 500 m

⁵ assumption: average density of rock: 2600 kg/m³

⁶ bentonite swelling pressure range for crystalline rock for a dry density of 1650-1750 kg/m³ (NAGRA 1994) and for a dry density of 1500 kg/m³ Opalinus Clay (Nagra 2002a) based on DIXON (2000)

⁷ values based on NAGRA (1985) for crystalline rock and on NAGRA (2002a) for Opalinus Clay

Temperature

The maximum temperature and the temperature-time behaviour of the canisters are determined principally by the canister and areal thermal loading, which are controllable parameters, and the ambient temperature and thermal properties of the bentonite backfill and rock. For the crystalline repository option, the ambient rock temperature at the nominal repository depth of 900 to 1200 m is 45 to 55 °C, whereas the ambient temperature for Opalinus Clay at a nominal repository depth of 450 to 850 m is 30 to 45 °C. The temperature at the reference depth for a repository in Opalinus Clay of 650 m is 38 °C (NAGRA 2002a). Thermal analysis calculations performed by SATO et al. (1997) for crystalline rock and JOHNSON et al. (2002) for Opalinus Clay indicate that, for the same thermal loading (1500 W/canister at emplacement, 3 m spacing along tunnels and 40 m spacing between tunnels) SF canister surface temperatures may reach a maximum of ~150 to 160 °C. The impacts of elevated temperatures are discussed in Section 6.2.

Pressure

Service loads on canisters after disposal will arise from groundwater pressures, mechanical loading from the swelling pressure of the buffer, lithostatic loading (in the case of sedimentary rock) and glacial loading. As noted in Table 2, for the crystalline rock case, the maximum load is 25 -30 MPa plus a further 5 MPa during glaciation. For a repository in Opalinus Clay at a maximum depth of 850 m the maximum isotropic load would be ~25 MPa (the lithostatic pressure during glaciation.) Differential loads may arise because of non-uniform rewetting of the buffer in the crystalline rock case (WERME 1998), although the total pressure in such a situation is likely to be relatively low, because, as groundwater pressure increases, more uniform wetting will occur. Anisotropic loading of the canister is unlikely to occur in Opalinus Clay, even though there are anisotropic stresses in the rock (the maximum principal stress at 650 m is 22 MPa), while the lithostatic stress and minimum horizontal stress are 15 MPa). The reason for this is that waste emplacement tunnels would be aligned with the maximum stress, thus the stress state within the sealed tunnel would be nearly isotropic.

It should also be noted that an internal pressure would be generated in the long term within an intact canister by a combination of fission gas release (release of Xe and Kr, should cladding fail as a result of, e.g., creep rupture) and He release from actinide decay. A maximum release of ~120 moles of gas would result after 100,000 years in the case of 4 PWR elements, assuming that all the fission gas and He is released from the fuel assemblies into the inner cavity of the canister. Assuming a design concept similar to that of SKB (a cast metal insert), this would generate a pressure of about 1 MPa, which is very small relative to the external load.

Groundwater Chemistry

The salinities of crystalline and Opalinus groundwaters are given in Table 2. Crystalline groundwaters are expected to have a very low salinity ($\sim 3 \times 10^{-3} \text{ mol L}^{-1}$), whereas the Opalinus Clay porewater has a salinity of 0.1 to 0.3 mol L⁻¹. The uncertainty in the former case is much greater, because salinities up to 0.3 mol L⁻¹ may be encountered. In contrast, there appears to be no realistic prospect of a salinity outside the range of 0.1 to 0.3 mol L⁻¹ for the case of Opalinus Clay (NAGRA 2002a). Both groundwaters would be reducing. The Opalinus Clay has a large redox buffering capacity because of its high pyrite content (~1 %) (NAGRA 2002a). The concentration of sulphide, which may cause corrosion of both steel and copper, is expected to be very low in both cases, as it will be controlled by pyrite solubility. Reliable measurements of sulphide concentrations are not available, as concentrations are normally below the detection limit of ~0.005 ppm (e.g. PEARSON & SCHOLTIS 1993). The potential for microbial reduction of sulphate by sulphate-reducing bacteria and its impact on corrosion is discussed in Section 7.1.1.

6 Design Criteria

6.1 Impermeability

The canister must be impermeable to water for its required lifetime.

Initial Impermeability

Initial impermeability would be assured by inspection of the closure weld using ultrasonic and/or X-ray techniques followed by helium leak testing (if possible). For copper canisters with a 5 cm thick shell (WERME 1998), a criterion has been set of < 1 in 1000 canisters having defects that would be larger than acceptance specifications. Presumably, the acceptance specification itself would be the absence of a defect or defects that would represent a significant probability of a through-wall breach being present or developing later under service conditions. Canisters with unacceptable defects would be repaired or replaced. At the present stage of development of canister fabrication technology and non-destructive inspection systems for thick copper, and considering that the start of repository operation would be about 2050 in Switzerland, it is not considered justified to define the acceptance specifications themselves. There is no elaboration provided on the origin of the SKB criterion and the quantitative acceptance criterion in terms of a defect size is not yet established. The criterion can thus be considered to be a preliminary manufacturing quality target.

For steel canisters, no acceptance specifications have been developed. However, there is considerable experience in welding and inspection of thick carbon steel (see the discussion in JNC 2000), thus there is good reason to believe that high quality fabrication and welding can be achieved.

The question of estimating the proportion of canisters that might be prematurely breached is a performance assessment issue discussed in Section 7.1.

Chemical Durability

There is considerable flexibility regarding the required lifetime for a SF disposal canister as noted in Section 5.1. Given the possibility that overall system performance is expected to provide a large margin of safety in the case of a 1000 year minimum lifetime in the context of a low permeability geosphere, it appears reasonable to retain a minimum containment target of 1000 years as one option (note the *expected lifetime* of a steel canister is about 10^4 years, see Section 7). Pending the outcome of further studies, the option to adopt a canister with a much longer lifetime (up to 100 000 years) should be maintained. The prospects for achieving both these targets are thus evaluated in this report.

Mechanical Integrity

As noted in Section 5.4, for both the crystalline and sedimentary repositories, the design load on the canister may be as much as ~30 MPa. The extent of anisotropic loading has not yet been determined. The canister should have sufficient strength to withstand these loads with a suitable margin of safety. A discussion of the selection of a factor of safety is given by Werme (1998), who suggests a factor of 2 to 2.5 for the Cu canister with a cast iron insert in the context of a repository in granite and this factor is also adopted in the present study. The present study has not evaluated the factor of safety in more detail as this requires both further development of design concepts and a more comprehensive analysis of maximum stresses in the repository environment.

The loaded canister should be capable of being handled in the encapsulation facility, during post-loading transfers and shipment and within the final disposal facility without detrimental effects on the long-term performance of the canister. This implies that damage to the canister shell of the type that would significantly enhance long-term corrosion or induce additional stresses that significantly increase the risk of stress-assisted failure mechanisms should be avoided.

Residual Barrier Function After Breaching

Loss of the containment function when breaching by corrosion or other mechanisms occurs (e.g. localised penetration or collapse of a heavily corroded canister when the wall thickness is significantly reduced) will result in pathways through the residual canister for radionuclide transport. These pathways may be tortuous, if breaching occurs through corrosion product-filled penetrations. Alternatively, there may be significant displacement if the failure occurs because of ductile overload after significant wall thinning. In any case, the combination of diffusion-controlled transport conditions and the presence of large quantities of redox-active corrosion products with high surface areas is expected to facilitate reduction reactions (e.g. Tc(VII) to Tc(IV)), providing an important barrier function long after canister breaching has occurred. The occurrence of such redox processes is often considered in performance assessment studies, although sorption on canister corrosion products is normally conservatively neglected. The value of a canister material such as steel (with redox-active magnetite as the corrosion product), as compared to a passivated metal such as titanium or an inert material such as a ceramic, is apparent.

6.2 Chemical and Physical Interactions with Buffer and Rock

The canister should be designed such that its physical and chemical interactions with the buffer and surrounding rock are not detrimental to their containment function. For example, chemical and physical interactions should not lead to degradation of surrounding barriers in a way that could increase the rate of mass transport of radionuclides. Some processes that need to be considered include chemical interaction of corrosion products with bentonite, excessive temperatures, volume changes arising from corrosion product formation and the production of H₂ in excess of the solubility in bentonite pore water.

Chemical Interaction of Corrosion Products with Bentonite

Chemical interaction between the canister corrosion products and bentonite may affect the mass transport properties of the buffer. Precipitation of Fe₃O₄ or other corrosion products in the pores of the buffer away from the immediate vicinity of the canister surface will reduce the permeability of the bentonite and inhibit subsequent mass transport of radionuclides from a breached canister, in addition to acting as a redox barrier. Occupation of adsorption sites by Fe(II) and Fe(III) could reduce the sorption capacity of the buffer close to the canister surface, depending upon the relative strengths of the Fe-bentonite and radionuclide interactions. However, such effects are likely to be limited to the buffer closest to the canister and there is expected to be sufficient buffer thickness that the outer parts buffer will be unaffected.

Temperature

The maximum temperature at the surface of a canister is determined by the following factors:

1. the thermal loading of the canisters, specifically the quantity of fuel and its decay heat at the time of final emplacement of the canister,
2. the thermal properties of the surrounding bentonite backfill and rock (thermal conductivities and heat capacities and ambient rock temperature).
3. the spacing between canisters and the emplacement tunnels,

The canister material selected should be able to withstand the materials degradation processes that occur for the range of temperatures that can reasonably be expected to occur. Thermal analysis studies (JOHNSON et al. 2002) for the Opalinus Clay repository for carbon steel SF canisters with a thermal output of 1500 W at the time of emplacement illustrate that canister surface temperatures are likely to reach 150 to 160 °C several years after emplacement. This maximum is determined principally by the canister thermal loading and the slow resaturation process, which maintains relatively dry conditions in the bentonite (and thus low thermal conductivity of the bentonite, increasing the canister surface temperature). In the case of crystalline rock, the ambient rock temperature is higher because of the greater repository depth, thus initial canister surface temperatures may be somewhat higher (SATO et al. 1997).

There are no strict criteria for establishing a maximum near-field temperature, but it is generally considered that the concerns regarding assurance of good long-term performance of the bentonite buffer, rather than those associated with corrosion performance of the canister, are the most stringent (JOHNSON et al. 2002). Specifically, the canister surface should be maintained at a sufficiently low temperature that the desirable plasticity and hydraulic and diffusion properties of the buffer material are preserved. The most important potential degradation processes involve cementation as a result of silica dissolution and precipitation, and precipitation of sparingly soluble salts (e.g. calcite and anhydrite) at or near the canister surface. Several studies have proposed a design maximum canister surface temperature of 100 °C (e.g. JOHNSON et al. 1994, WERME 1998, BEL & BERNIER 2001). These criteria appear to have been set not because there are fundamental limitations in the performance of the canister materials above this temperature, but principally because of concerns about bentonite integrity at temperatures above 100 °C and because of lack of confidence in the coupled THM models describing the evolution of near-field conditions over the first few hundred years after emplacement. It is therefore not possible to view the maximum canister service temperature in isolation as a canister materials performance criterion.

Maximum service temperatures for the canister are well above 100 °C for repository environments in Switzerland. Because of the low thermal conductivity of granular bentonite prior to resaturation and the relatively high ambient temperature (38 °C) in Opalinus Clay at repository depth, it is difficult to keep temperatures below 125 °C throughout the entire bentonite barrier (JOHNSON et al. 2002). For tunnel emplacement of canisters with either 4 PWR or 9 BWR fuel assemblies with a heat output of 1500 W at emplacement (burnup of 48 GWd/tHM with 40 years cooling), calculations of temperature evolution show that the maximum canister surface temperature will reach ~150 °C and at the point in the bentonite midway between the canister and the tunnel wall may reach ~110 °C. To achieve lower temperatures would require much longer cooling times for the fuel. At a temperature of 150 °C, for granular bentonite with an initial dry density of 1.5 Mg m⁻³, a ~50 % drop in swelling pressure occurs due to cementation, whereas at 125 °C, only minor reduction in swelling is observed (PUSCH et al. 2002). As a result, the thermal constraint for the bentonite presently specified for the design of the SF / HLW emplacement tunnels in Opalinus Clay is that the outer

half of the bentonite should remain below ~ 125 °C, so as to retain maximum swelling capacity in at least this region to ensure a good quality hydraulic seal around each canister. The thickness of buffer retaining good sealing properties would still be sufficient (~ 0.35 m) to provide a substantial diffusion barrier. Results of assessment calculations for Opalinus Clay (NAGRA 2002a) and for crystalline rock (NAGRA 1994) confirm that the assumed loss of the transport barrier function of the inner half of the bentonite (an extreme assumption, given the limited alteration that might occur at temperatures of 125 to 150 °C) has no significant impact on dose, confirming the statements above that criteria should not be set in isolation from system performance assessment. There is thus no reason to limit maximum canister surface temperatures to 100 °C.

Volume Changes

Corrosion of carbon steel or cast iron under anaerobic conditions is driven by water reduction, thus it will continue until there is complete conversion of the steel to magnetite. This is likely to take hundreds of thousands of years at a typical long-term uniform corrosion rate of ~ 1 $\mu\text{m/a}$ (see Section 7.1 for a discussion of corrosion rates). The volume increase for this reaction is a factor of 2.1, thus the density of the buffer would gradually increase as a result of its compaction due to magnetite buildup. For a 2.5 m diameter tunnel such as that proposed for Opalinus Clay, significant excess loads on the residual canister wall are likely to result only after some tens of thousands of years (JNC 2000), as the total corrosion depth in 10,000 years for a corrosion rate of ~ 1 $\mu\text{m/a}$ is only ~ 1 cm. This is unlikely to affect the mass transport properties of the buffer significantly, except in the region of the buffer closest to the canister.

Gas Generation

The use of iron-based materials either for an insert, as in the SKB canister design, or as primary containment also raises the question of the impact of hydrogen production on the mass transport properties of the buffer and on disposal system performance as a whole (NAGRA 2002a). The corrosion rate of steel is sufficiently high that, coupled with low hydrogen solubility and the low rate of diffusion of dissolved hydrogen through the buffer, gas bubbles will form at the canister surface. Laboratory experiments by PUSCH et al. (1985, 1987) have demonstrated that gas escapes through the largest pores in the bentonite before extreme pressures build up. TANAI et al. (1997) tested the Japanese bentonite Kunigel VI (to be used in a Japanese HLW repository with a carbon steel canister) and French Fo-Ca clay and they compared the results with the experiments of PUSCH et al. (1985) on MX-80 bentonite. They found that in all three materials, the critical breakthrough pressure was about the same as the respective swelling pressure (in the Fo-Ca clay it was actually about 40 % higher than the swelling pressure). In the experiments of HORSEMAN et al. (1997), the gas breakthrough pressure was found to be equal to the sum of the swelling pressure and the equilibrium external water pressure. Because the volume of the gas pathways through the buffer would be extremely small relative to the total volume of the buffer (SWIFT et al. 2001), the hydraulic and diffusion properties of the buffer, and thus the transport rates of dissolved radionuclides, are unlikely to be affected by this process. The impact of hydrogen gas buildup on overall disposal system performance is discussed in some detail in NAGRA (2002a).

If continuing research associated with the significance of hydrogen buildup and transport in repositories cannot resolve these issues, there are material selections that could obviate these difficulties altogether. The uncertainties regarding the use of iron-based materials can be avoided by the use of a copper canister with a bronze insert. The extent of corrosion of copper and bronze would be negligible even over very long time frames, and there would be no

hydrogen generation because copper is thermodynamically stable in anoxic groundwater in the absence of sulphide. The principal issues related to the use of a bronze insert are increased cost relative to mild steel or cast iron and the lack of industrial capability for casting bronze at this scale. The limited contribution that Cu and its corrosion products would make to redox buffering (as compared to Fe corrosion products) should also be noted. A further alternative to cast iron as insert is stainless steel, which has a markedly lower anoxic corrosion rate ($< 0.01 \mu\text{m/a}$ (WADA et al. 1999)), although its cost is substantially higher than that of carbon steel.

Transport Limitations

A further consideration in performance assessment calculations that adopt the SKB reference design is the approach to be taken in modelling release from a small defect in the external Cu shell. In several performance assessment studies (SKB 1992, VIENO & NORDMAN 1996, WIKJORD et al. 1996, SKB 1999), credit was taken for the reduced mass flux from the canister arising from the small defect size in the Cu shell. This leads to significant decreases in dose (a factor of 15 in VIENO & NORDMAN 1996 and 6 in WIKJORD et al. 1996) relative to the case in which the canister provides no mass transport resistance. Implicit in adopting such an approach is the assumption that the defect will not increase in size with time. This can reasonably be argued for corrosion of the Cu itself on the basis of the lack of oxidants in the repository. The argument that the defect size in the Cu shell would not be influenced by the volume changes of the enclosed steel insert (BOND et al. 1997) is more problematic (see discussion above and in Section 7.1), although it must be acknowledged that significant increases in defect size are likely to take a very long time, given the low corrosion rate of steel under anoxic conditions.

Criticality

Canisters for the disposal of SF must be designed to preclude criticality even upon water ingress, either for the undisturbed configuration of the fuel or for configurations that might arise from cladding breaching and dissolution of fuel and separation of Pu from the fuel matrix or fuel slumping. Criticality assessments for spent LWR fuel disposal must deal with the issue of burnup credit or with design approaches that would preclude criticality, because unirradiated fuel is sufficiently reactive that several close-packed fuel assemblies in the presence of sufficient water may result in a critical configuration. Calculations for MOX and UO_2 fuel of average burnup indicate that the reactivity¹ decreases for the first few hundred years after discharge of the fuel from the reactor due to ingrowth of ^{241}Am , a strong neutron absorber (KÜHL et al. 2003). It gradually increases back to its initial value over hundreds of thousands of years as ^{241}Am decays and ^{235}U grows back in from decay of ^{239}Pu . This means that the potential for criticality requires evaluation for the entire period for which assessment calculations are performed.

Several criticality assessments have been performed in the context of SF canister design studies. For example, ANTILLA (1996) performed a criticality assessment for PWR and BWR assemblies placed in a Cu/steel canister for undisturbed geometries (i.e., no fuel slumping). He calculated the effective neutron multiplication factor (k_{eff}), assuming that water had filled the canister and that neutron reflection around the canister was maximally effective. Values of k_{eff} of < 0.95 were considered to be sub-critical. The canister design loaded with eleven BWR elements was sub-critical for burnup values of $>10 \text{ GwD/tIHM}$. AGRENIUS (2002) performed

¹ Reactivity is a measure of the number of excess neutrons produced by fission

criticality calculations for canisters of BWR and PWR fuel for the full range of burnups and enrichments of the fuel presently stored in CLAB (the Swedish central interim wet storage facility). His calculations show that k_{eff} would be less than 0.95 for canisters containing any of the fuel presently in storage under the assumption of breached canisters filled with water surrounded by saturated bentonite.

KÜHL et al. 2003 discuss preliminary criticality calculations for the steel canister design concept assumed in Section 7.2 and show that, for UO_2 SF, a moderate burnup (> 22 and > 11 GWd/tHM for PWR and BWR fuel, respectively) could prevent criticality. There are good prospects that this could be administratively assured in the canister loading procedure, because methods of confirming the burnup of each fuel assembly are available (LEBRUN & BIGNAN 2001). A knowledge of the burnup and decay time of the fuel assemblies is also required in order to assure the combined thermal output of all assemblies in a given canister does not exceed the thermal criterion (assumed to be 1500 W per canister).

Alternative or supplementary approaches to the use of burnup credit would be to use neutron absorbers in the canister or to fill the canister void space with a particulate material that would effectively reduce moderation by reducing the quantity of water that could enter a breached canister, thus preventing criticality. The latter approach may be preferred because reliance on neutron absorbers requires that it be shown that they remain in the canister and are not leached out for extremely long periods of time. KÜHL et al (2003) have shown that for the steel canister design discussed in Section 7.2, criticality would not occur for unirradiated fuel enriched to 4.5 % if the water density is less than 0.4 g cm^{-3} averaged over the canister internal void volume. This would be the water density in a canister filled with typical sand, suggesting that filling the void space in the canister with a particulate such as silica sand would be a feasible method of precluding criticality.

Radiation Field

The dose rate at the surface of the canister should be low enough that radiolysis of pore water in the bentonite does not produce significant quantities of radiolysis products that could accelerate corrosion of the canister. The criterion proposed by SKB is that the surface dose rate of the canister be $< 1 \text{ Gy h}^{-1}$ (SKB 1997). Corrosion rates of copper are not increased at such low dose rates (KING et al. 2001). MARSH & TAYLOR (1988) observed negligible increases in carbon steel corrosion rates at dose rates of $< 3 \text{ Gy h}^{-1}$, a value which is recommended as a surface dose rate criterion for a steel canister from the perspective of corrosion processes. The surface radiation dose rate for the steel canister design discussed in Section 7.2 is $< 35 \text{ mGy hr}^{-1}$, well below the threshold for radiation-enhanced corrosion. Thus for wall thicknesses proposed in the present study, corrosion due to radiolysis would be insignificant.

7 Canister Design

7.1 Materials options

Several materials exist that could meet a required lifetime target of 1000 years for the temperature and chemistry conditions of interest. The most promising are mild steel and cast iron, copper, nickel-based alloys and titanium alloys. Comprehensive summaries of their corrosion performance are given by JOHNSON et al. (1994) and JNC (2000). KING et al. (2001) provide a thorough treatment of copper in relation to its performance in crystalline rock repositories. Of these materials, copper and mild steel are arguably the most predictable in corrosion behaviour in the range of conditions of interest in a Swiss repository. The concept of a thin shell of corrosion-resistant alloy (e.g. Grade-16 titanium or Hastelloy) overtop of an internal support structure of carbon steel is also feasible and has been examined (e.g. JOHNSON et al. 1994, JNC 2000). The following evaluation focuses principally on the corrosion performance of steel and copper and presents an assessment of the corrosion allowance required to give a lifetime of a minimum of 1000 years for steel and 100 000 years for copper. In addition to reviewing results of relevant corrosion studies, the following discussion provides a canister lifetime assessment for steel and copper canisters under the conditions expected in a repository in crystalline rock and Opalinus Clay. There are a few cases in which there are significant differences between the canister service conditions in repositories in crystalline rock and Opalinus Clay, e.g. the possibility of an extended unsaturated phase for the latter, and these are particularly noted. A brief discussion of using alternative canister materials to reduce gas production is included.

7.1.1 Carbon Steel and Cast Iron

There is now a large body of data available to assess the performance of carbon steel or cast iron in a repository in either crystalline rock or Opalinus Clay. These data are reviewed here in order to establish the thickness of steel required to achieve containment for a minimum of 1000 years and to evaluate the expected canister lifetime.

Summarizing briefly the assessment in Kristallin-1 (NAGRA 1994) and Project Gewähr (NAGRA 1985), the following extents of corrosion of carbon steel canisters for a repository in crystalline rock were conservatively estimated for a 1000 year period:

- i) initial consumption of trapped oxygen < 1 mm
- ii) corrosion by sulphide arising from sulphate reduction - 9 mm
- iii) corrosion under anaerobic conditions as a result of reduction of water - 20 mm

As discussed in the following sections, there are new data that suggest that the above estimates significantly overpredict the extent of anaerobic corrosion and corrosion due to sulphate reduction.

Corrosion During the Initial Aerobic Phase

As discussed in Section 5.4, in crystalline rock, there is a possibility that resaturation times could be relatively short, perhaps years or decades. This suggests that moisture levels will rapidly reach levels sufficient to initiate corrosion processes. In contrast, for a repository in Opalinus Clay, resaturation will likely be much slower, thus there may be a delay in the initiation of canister corrosion processes. Nonetheless, as discussed below, the possibility of a protracted partially saturated period may also increase uncertainties with respect to localised corrosion.

It is known that, in air, corrosion of mild steel is extremely slow provided the relative humidity is less than a critical value of ~60 % (BROWN & MASTERS 1982). For granular bentonite backfill, the 2 % initial moisture content (PUSCH et al. 2002) corresponds to a relative humidity of ~5 % (MARSHALL & HOLMES 1979). In contact with bentonite, the critical relative humidity for initiation of aqueous corrosion may be reduced to about 30 to 40 % due to absorption of water by hygroscopic salts, in particular, trace quantities of CaCl₂ or NaCl present in bentonite (MANSFELD & KENKEL 1976). Such a high moisture level at the canister surface is unlikely to be reached for many years in the case of Opalinus Clay, because the high temperature gradient in the bentonite maintains low moisture levels in the hottest part of the bentonite, even when saturation of the outer bentonite is approached (BÖRGESSON & HERNELIND 1999). The corrosion of the steel canisters is therefore expected to be limited (< 100 µm) for the first decades in Opalinus Clay, because the surfaces of canisters will remain dry until the humidity increases sufficiently that a thin film of water can condense and initiate both local and general corrosion. During this time period, much of the oxygen initially present in the bentonite will be consumed by reaction with pyrite and siderite in the outer part of the bentonite and in the Opalinus Clay immediately surrounding the tunnel, which will have a much higher moisture content (WERSIN et al. 2002). At some point, moisture levels at the canister surface will increase sufficiently to initiate corrosion. Under the high temperature conditions, corrosion of steel is expected to occur at an initially rapid rate if there is oxygen remaining in the bentonite. There is a possibility that preferential wetting of the canister surface could occur in the coolest areas, with wetter areas being anodic and drier areas supporting oxygen reduction, leading to localised corrosion, thus both uniform and pitting corrosion are considered possible.

One approach to evaluating the total extent of corrosion during the initial aerobic phase is to estimate the expected depth of corrosion on a simple mass-balance basis, assuming that no other processes consume oxygen. As noted in Kristallin-I (NAGRA 1994), this gives a uniform corrosion depth of ~1 mm for a steel HLW canister.

The oxygen entrapped in the bentonite would gradually be consumed by corrosion of steel and oxidation of small quantities of pyrite and siderite that are present in the bentonite (MULLER-VONMOOS & KAHR 1983). WERSIN et al. (1994a) estimated that for the KBS 3 repository design, the time to 99 % oxygen depletion would be 7 to 280 years, assuming oxidation of pyrite in the buffer was the dominant oxygen consumption process. Their calculations were performed for a temperature of 25 °C and they noted that the rate would be ~10 times faster at 60 °C. This suggests that the duration of oxic corrosion of steel may be of the order of decades. However, studies summarised in JNC (2000) indicate that, in saturated bentonite, reduction of oxygen quickly becomes negligible and water reduction becomes the dominant cathodic reaction, because of slow migration of oxygen in saturated bentonite.

Calculations and discussion of oxygen consumption rates in the case of the Opalinus Clay repository near field are presented by WERSIN et al. (2003), who conclude that the time to 99 % oxygen depletion may range from 2 years (realistic case) to 100 years (pessimistic case).

It is not certain, however, that uniform corrosion would be the most important mechanism under these conditions. The possibility that pitting corrosion could also occur clearly must be examined. It has been argued that pitting would only be possible during the aerobic phase of the repository when passivation may lead to susceptibility to this process (JNC 2000). Although the severity of pitting may be greater under aerated conditions, it is questionable if this provides evidence that pitting (or non-uniform general corrosion) will not occur under anaerobic conditions. It may be more appropriate to focus on the degree of non-uniformity of corrosion, as is discussed below. There are several approaches that can be considered in evaluating the possible extent of pitting corrosion.

The Probability of Pitting

It is useful first of all to consider whether passivation of carbon steel occurs in bentonite pore water. This has been studied at a temperature of 80 °C using electrochemical techniques by TANIGUCHI et al. (1998), who suggested based on these results that passivation does not occur in compacted saturated bentonite and that only active corrosion occurs, thus pitting would not occur. In spite of this observation, it is prudent to consider several approaches to estimating the extent of pitting corrosion.

Pitting and Crevice Corrosion

Studies of pitting and crevice corrosion of carbon steel discussed in JNC (2000) provide a good assessment of the possible extent of pitting and the data and arguments are briefly summarised here. Under bentonite porewater conditions, bicarbonate will promote passivation, whereas chloride will promote passive film breakdown. The fact that the repassivation potential is close to the passivation potential suggests that crevice and pitting corrosion may be possible. Even though the oxygen reduction current is unlikely to be maintained because of slow oxygen transport, thus propagation should be stifled, this may only apply under saturated conditions. During the partially saturated phase, crevice and pitting corrosion might occur.

ISHIKAWA et al. (1994) studied corrosion of carbon steel in seawater, synthetic groundwater and bentonite/aerated water mixtures. An extreme value analysis of the corrosion data gave the equation:

$$P = x + 7.5x^{0.5} \quad (1)$$

where P is the upper limit of the corrosion depth, and x is the average corrosion depth (in mm). This suggests that the pitting factor PF (the ratio of the maximum depth of corrosion to the average depth of corrosion) is relatively small and that it decreases with average corrosion depth. This is apparent if Equation 1 is reformulated as

$$PF = 1 + 7.5x^{-0.5} \quad (2)$$

Another approach discussed in JNC (2000), involves comparison of the pitting factors measured in the laboratory experiments of TANIGUCHI et al. (1999) and those reported by ROMANOFF (1989) for burial of steel and iron samples in soil for periods of up to 15 years. These data are compared in Figure 3, from which it is clear that the pitting factor decreases with the average corrosion depth, declining to a maximum value of about 5 for an average corrosion depth of 1 mm. The slope of the line shown in Figure 3 defining the upper bound of the observed PF value has a slope of -0.66, slightly greater than the value of -0.5 inferred from Equation (2). Comparison of Equation (2) with the data in Figure 3 shows good agreement at corrosion depths of less than ~0.2 mm, with higher PF values predicted at greater average corrosion depths. A further analysis in JNC (2000) suggests that, even using extreme value statistical analysis, the pitting factors are likely to be small, such that maximum pitting depths would be in the range of 1 cm. Furthermore, the decrease in PF with increasing corrosion depth (and also, therefore, increasing time) indicates that the corrosion will become less localised and more uniform as time progresses.

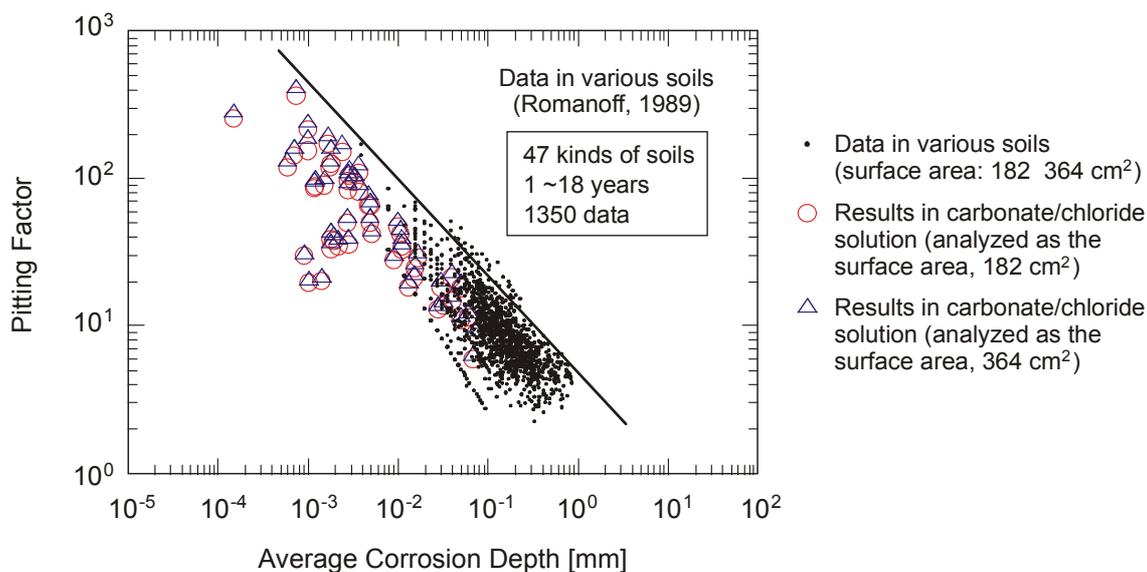


Fig. 3: Relationship between average corrosion depth and pitting factor of carbon steel in carbonate/chloride solutions and in various soils (JNC 2000)

It is concluded from these studies that the corrosion allowance for pitting is no more than 1 cm. Furthermore, should the oxic period last longer than several decades, this conclusion is unaffected, given the relationship shown in Figure 3.

Microbial Corrosion and Corrosion Due to Sulphide

Sulphide may be present in bentonite porewater, where its concentration and availability may be controlled by pyrite solubility and by reduction of sulphate by sulphate-reducing bacteria (SRB). Previous assessments of the impact of microbial corrosion have considered that SRB might reduce all the available sulphate to sulphide, thus providing an oxidising agent for steel canister corrosion (NAGRA 1994). Since the time of these assessments, considerable progress has been made in evaluating the viability of SRB in bentonite. These studies have confirmed that SRB are unable to thrive in compacted bentonite. For example, studies summarised in JNC (2000) showed that SRB introduced into bentonite at the beginning of steel corrosion experiments were absent at the completion of the tests and that the introduction of the bacteria had no effect on the corrosion rate. PEDERSEN et al. (2000a) have shown in experiments lasting 15 months at temperatures of up to 70 °C that only spore-forming bacteria survive in highly compacted bentonite and that their numbers are reduced markedly over time. In other studies, the activity of SRB was observed to cease at saturated densities higher than 1.5 Mg m⁻³ (PEDERSEN et al. 2000b). Furthermore, PUSCH (1999) has shown that SRB are immobile in bentonite with a saturated density exceeding 1.9 Mg m⁻³, thus they will not be able to migrate towards the canister from the surrounding rock. As a result, it is expected that bacteria will have a negligible impact on canister corrosion in compacted bentonite.

Diffusive transport of sulphide towards the canister may also occur if SRB are viable in the rock, thus producing sulphide that may migrate across the bentonite, and by dissolution of sulphide from the pyrite naturally present in the bentonite. This has been considered by WERSIN et al. (1994b), who estimated a corrosion rate of Cu of 4×10^{-6} mm a⁻¹, assuming that

amorphous pyrite naturally present in bentonite combined with diffusive transport through bentonite controls the flux of sulphide. Combined with a pitting factor of 5, they derived a maximum rate of corrosion due to sulphide of $2 \times 10^{-5} \text{ mm a}^{-1}$. The conclusions for copper are also broadly applicable to the case of the steel canister considered in the present study.

Anaerobic Corrosion

There is a large body of data available on corrosion of carbon steel under anaerobic conditions. Although the rates are low enough that this process is of relatively low importance for assessing the corrosion allowance (e.g. even at a relatively high rate of $10 \mu\text{m a}^{-1}$, the reduction in wall thickness is only 1 cm in 1000 years), the determination of the rate is of great importance for assessing the effects of hydrogen gas production in a repository. As a result, a comprehensive discussion of anaerobic corrosion of carbon steel is presented here, with the objective of determining the expected long-term rate.

A large number of measurements of carbon steel and cast iron corrosion have been made over the past twenty years in order to evaluate the corrosion performance of steel canisters. KING & STROES-GASCOYNE (2000), for example, include over 250 measurements in their review. The selection of a best estimate corrosion rate for steel for anaerobic conditions from this data requires some care, because, for a variety of reasons, much of the data is not relevant to long-term anaerobic corrosion in bentonite porewater. Table 3 provides a data set for corrosion under anoxic conditions that has been selected because the measurements clearly represent conditions of interest, in particular, elevated temperatures in dilute groundwater or bentonite/groundwater systems and attempts to eliminate oxygen.

The rates in Table 3 are grouped into two categories, those derived from weight loss in immersion tests and those inferred from direct measurements of H_2 evolution. The former give higher rates, because they represent an integrated rate, which may reflect a short aerobic phase with a high rate, a rapidly decreasing anaerobic rate as the protective film grows and a low steady-state rate, as illustrated in Figure 4 from SMART et al. (2001). This is particularly problematic in immersion tests in wet bentonite, in which O_2 is trapped in the pores. SIMPSON & VALLOTON (1986), for example, measured a rate at 80°C of 20 to $68 \mu\text{m a}^{-1}$ over 816 h, while the rate over 6648 h was 4 to $10 \mu\text{m a}^{-1}$ (see Table 3). The results from long-term weight-loss immersion tests given in Table 3 suggest that the corrosion rate eventually drops below $10 \mu\text{m a}^{-1}$ at 80°C . Weight loss measurements are considered to give unrealistically high corrosion rate estimates for either assessing long-term steady-state anaerobic corrosion rates or estimating H_2 evolution rates. They are nonetheless of some interest, given that in many cases they involve long-term contact of bentonite with steel. Further illustration of the trends in the corrosion rate data of Table 3 are shown in Figures 5 and 6.

It is also worthwhile to examine analogue studies that have derived corrosion rates from the study of archaeological artefacts that have been buried in various soil and sediment environments for up to 2000 years, which have been summarised by MILLER et al. (2000). Corrosion rates ranged from 0.1 to $10 \mu\text{m a}^{-1}$ (also shown in Figure 5). Some caution must be used in applying the rates derived from such studies, as some of the specimens may have been retrieved from oxidising environments, thus results may be skewed towards higher rates. Nonetheless, they provide average rates over a time frame even greater than the minimum canister design lifetime, and permit the establishment of an upper bound rate.

Tab. 3: Summary of carbon steel corrosion rates measured by weight loss or H₂ generation for anaerobic conditions relevant to crystalline and Opalinus Clay repositories

Time	Temperature [°C]	Solution Composition	Corrosion Rate [$\mu\text{m a}^{-1}$]	Reference & Comments
Up to 2000 years (analogue studies)	ambient	soil/sediment pore water (compositions unknown)	0.1-10	MILLER et al. (2000)
816 h	80	groundwater - saturated bentonite	24-68	SIMPSON & VALLOTON (1986)
816 h	140	groundwater - saturated bentonite	20-40	SIMPSON & VALLOTON (1986)
6648 h	80	groundwater - saturated bentonite	4-10	SIMPSON & VALLOTON (1986)
6648 h	140	groundwater - saturated bentonite	5-13	SIMPSON & VALLOTON (1986)
6 months	170	humid clay	0.02-0.34	DEBRUYN et al. (1991)
6 months	170	claywater	0.5 to 1.1	DEBRUYN et al. (1991)
700 days	80	compacted bentonite	2-6	JNC (2000)
5000 h	90	syn. granite water	<0.1	MARSH & TAYLOR (1988)
9 months	90	clay water	25	LANZA & RONSECCO (1986)
100-380 h	80	Böttstein groundwater	2.7	SIMPSON (1989), H ₂ evolution
100-380 h	80	Säckingen groundwater	1.7	SIMPSON (1989), H ₂ evolution
195 h	80	Bentonite/ groundwater slurry	27	SIMPSON (1989), H ₂ evolution
195 h	80	Bentonite/ groundwater slurry	37	SIMPSON (1989), H ₂ evolution
~5000 h	21	Böttstein groundwater	0.05-0.1	KREIS (1991), H ₂ evolution
7500 h	30	Bentonite water	0.15	PEAT et al. (2001), H ₂ evolution
4500 h	50	KBS groundwater sat'd with FeCO ₃	< 1	SMART et al. (2001), H ₂ evolution
5000	85	Äspö water (0.3 mol/l Cl ⁻)	0.1	SMART et al. (2001), H ₂ evolution
5000	50	Bentonite-equilibrated water	0.05	SMART et al. (2001), H ₂ evolution

The corrosion rates derived from H₂ evolution rates also illustrate the extrapolation problems associated with slow growth of the protective film. The slow approach to a steady-state corrosion rate is illustrated in Figure 4 from SMART et al. (2001). It is notable that all the corrosion rate values derived from H₂ evolution rates over short times (hundreds of hours) are significantly higher than the long-term steady-state values (Note – only the rates at the end of the testing periods are shown in Figure 6). This is confirmed by inspection of the corrosion rates vs. time curves of SIMPSON (1989) and the rate curves of KREIS (1991), which drop by a factor of 5 to 10 between 200 h and 5000 h. The anomalously high rates measured by SIMPSON (1989) for bentonite slurries are likely to arise from a similar effect. Based on all these observations, the data from the long-term H₂ evolution experiments appear to be the most reliable. These data include conditions relevant to both crystalline and Opalinus Clay repositories, including moderate salinity, contact with bentonite-equilibrated water and temperatures of up to 85 °C (canister surface temperatures decline to this value after about 500 years). These data show that the use of a rate of 1 µm a⁻¹ in safety analysis calculations is likely pessimistic and that the actual rate may be somewhat lower.

Estimate of the Corrosion Allowance

Consideration of the corrosion depths in 1000 years for all the processes above gives the following:

- Uniform corrosion by oxygen during the aerobic phase – < 1 mm
- Pitting corrosion during the aerobic phase – 10 mm
- Corrosion due to sulphide – 0.02 mm
- Anaerobic corrosion – 1 mm

These findings suggest the total corrosion allowance required is less than 1.5 cm and that for a 10,000 year lifetime the corrosion allowance is only slightly greater than 2 cm. The reduction in the required corrosion allowance relative to the estimate of ~3 cm in NAGRA (1994) arises from improved understanding of anaerobic corrosion and more realistic estimates of potential non-uniform corrosion and corrosion due to sulphide, including SRB.

Stress-Corrosion Cracking

Stresses in the region of a final closure weld are expected to be relatively high, perhaps approaching the yield stress (ATTINGER & DUIJVESTIJN 1994). Components with such stresses are typically heat treated if they are to be exposed to environments that might initiate SCC. Such heat treatments of welds are common practice and are typically done at temperatures of 500 to 600 °C. This may not be viable for SF canisters, as it is desirable to ensure that the cladding temperature remains below 400 °C to prevent creep rupture. While some fuel cladding failures would be acceptable, because the canister would already be sealed, large numbers of failures should be avoided because of complications that would arise if a canister was shown to be defective and fuel had to be unloaded. As an alternative to heat treating, the weld region could be shot-peened. Shot-peening introduces surface compressive stresses and reduces the probability of crack initiation (PARKINS 1985, 2000). For this application, shot peening has the advantage that it could easily be performed remotely at ambient temperature and may also peen undetected crack-like defects on the surface that may otherwise act as stress raisers. Unfortunately, unlike heat treatment, shot-peening only affects the stress levels in the near-surface region (a few tenths of a mm) and any beneficial effect is lost once this layer is

corroded. However, shot-peening could be a suitable strategy for mitigating SCC processes during the early oxic period in the evolution of the repository environment.

In spite of the high stresses, there is good reason to believe that SCC would not occur. The three requirements for SCC are (i) a susceptible material, (ii) an aggressive environment, and (iii) a suitable tensile stress. Only certain environments cause the SCC of mild steels: caustic, nitrate, phosphate, CO-CO₂, high-temperature water (>200°C), and HCO₃⁻/CO₃²⁻ solutions (CIARALDI 1992). Of these environments, the only one likely to be present in the repository is a HCO₃⁻/CO₃²⁻ solution.

There are two forms of SCC of mild steel in HCO₃⁻/CO₃²⁻ solutions, the so-called high- and low- (or near-neutral) pH SCC. Table 4 summarizes the environmental and mechanical conditions necessary for high- and low-pH SCC, derived from operating experience with C-Mn pipeline steels. Although there are differences in the susceptibility of different microstructures, all mild steels are susceptible to this form of attack to some degree.

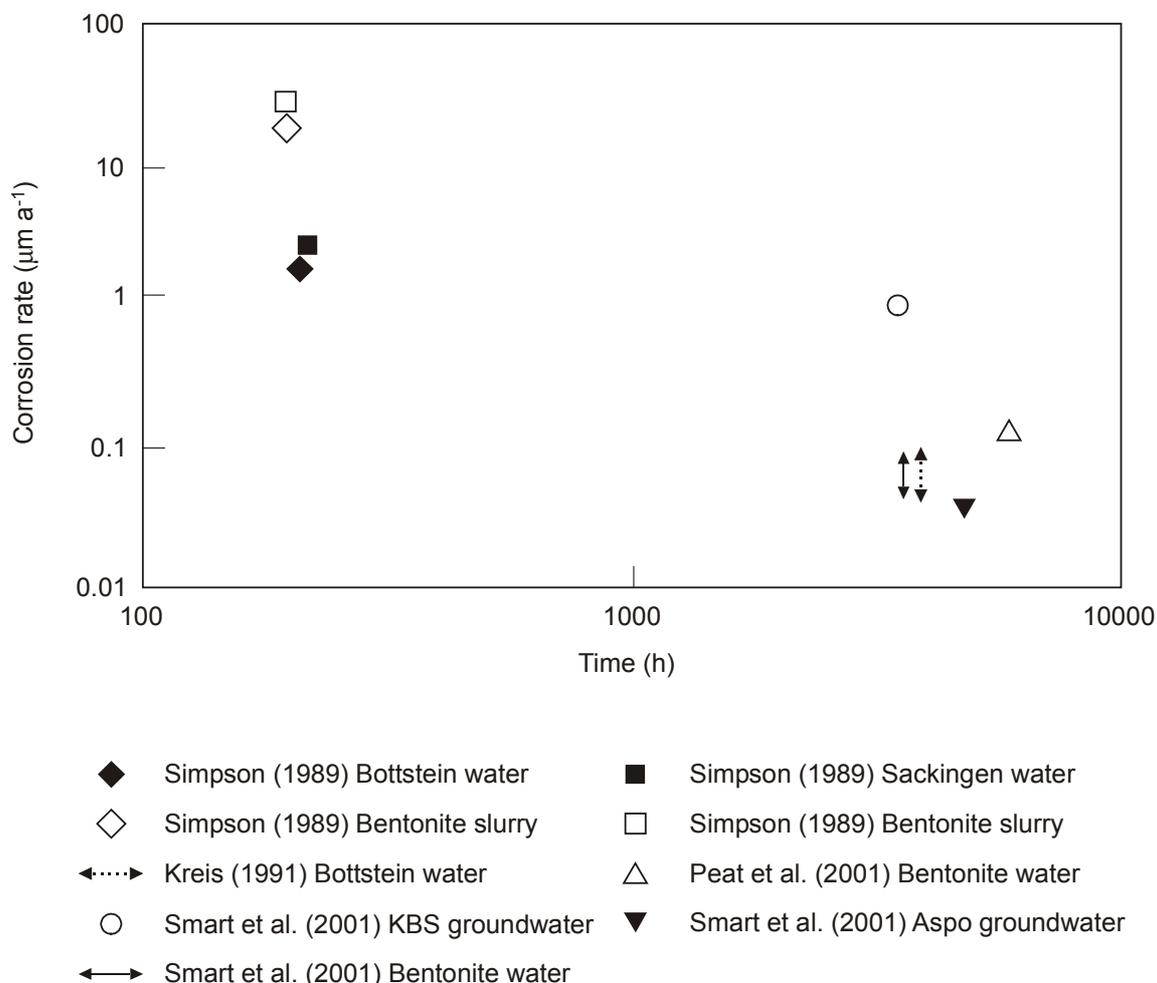


Fig. 6: Corrosion rates of carbon steel under anoxic conditions based on H₂ generation rates (data from Table 3)

Tab. 4: Environmental and mechanical conditions for high- and low-pH SCC of mild steel

Parameter	High-pH SCC	Low-pH SCC
Solution	Concentrated $\text{HCO}_3^-/\text{CO}_3^{2-}$, typically $\sim 1 \text{ mol}\cdot\text{L}^{-1}$	Dilute $\text{HCO}_3^-/\text{CO}_3^{2-}$ solution, typically $\sim 0.001\text{-}0.010 \text{ mol}\cdot\text{L}^{-1}$
pH	pH 9.3-10.5	pH 5.5-7.0
Potential/redox conditions	Well-defined potential range that changes with temperature. -0.52 to $-0.67 \text{ V}_{\text{SCE}}$ at ambient T Anodic to E_{CORR} under anaerobic conditions.	Occurs at E_{CORR} under anaerobic conditions
Temperature	Rate increases with temperature, with an apparent activation energy of $42 \text{ kJ}\cdot\text{mol}^{-1}$	No apparent effect of temperature
Tensile stress	Threshold stress decreases with amplitude of cyclic loading. Cyclic loading apparently required for crack propagation.	Observed over wide range of tensile stresses (60-100 % of yield stress), but cyclic loading apparently required for crack growth
Mechanism	Film-rupture dissolution	Uncertain

Although canisters in a Swiss repository will be exposed to some of the conditions listed in Table 4, not all of the necessary conditions for either high- or low-pH SCC will be present. Of these necessary conditions, the two most unlikely to be met in the repository are the required pH range for cracking and the need for cyclic loading. From a mechanistic viewpoint, it is significant that the pH ranges for low- and high-pH SCC are centred on the pK_a values for the equilibria between H_2CO_3 and HCO_3^- ($\text{pK} = 6.4$) and between HCO_3^- and CO_3^{2-} ($\text{pK} = 10.3$). Buffering by the bentonite and surrounding Opalinus Clay will produce a pore-water pH of ~ 7.3 (CURTI & WERSIN 2002), in between the necessary pH ranges for the two forms of SCC. Cyclic loading is believed to be necessary for crack propagation as it affects the plasticity at the crack tip. Although cyclic loading is a common feature of pipeline operations, the same condition will not affect canisters in the repository.

Hydrogen Damage

Hydrogen can have a number of effects on the mechanical and chemical (corrosion) stability of structural materials. The important processes that affect the mechanical properties of mild steel are (CRAIG 1987): (i) embrittlement during plastic deformation due to exposure to gaseous H_2 , (ii) hydrogen stress cracking (also referred to as hydrogen-induced cracking (HIC) and, in sulphide-containing environments, sulphide stress cracking), (iii) the loss of ductility during plastic deformation, (iv) high-temperature hydrogen attack, (v) blister formation, (vi) damage due to hydrogen picked up during casting and forging, (vii) hydrogen cracking of welds, (viii) micro-perforation, and (ix) degradation of creep properties. Hydrogen can also affect the rate of anodic dissolution (GU et al. 1999) and the stability of surface films (YU et al. 2002).

A number of these processes are not important for cast-steel canisters in a repository (high-temperature hydrogen attack, micro-perforation) and the detrimental impacts of others can be avoided or minimized by prudent engineering design and manufacturing practices (loss of ductility and embrittlement during plastic deformation, hydrogen pick-up during casting and

welding, degradation of creep properties). Furthermore, the effects of hydrogen on the rate of anodic dissolution and the stability of surface films are implicitly included in the experimental corrosion rates used above to predict the canister lifetime.

The hydrogen damage processes of most concern for a canister are hydrogen stress cracking (HSC) and blister formation. Hydrogen stress cracking (HSC) involves the diffusion of hydrogen to highly stressed regions in the material and results in the formation of internal cracks. As such, the severity of HSC increases with increasing strength of the material, increasing applied and residual stress, and increasing hydrogen content. It is most severe at a temperature of ~ 20 °C. Blister formation is more important for low-strength steels and involves the diffusion of atomic hydrogen to internal defects (such as voids, laminations, and inclusions), where it forms H_2 . The internal pressure of molecular H_2 is sufficient to cause localized plastic deformation and the formation of blisters. Blister formation is particularly important in sulphidic environments.

Since both HSC and blister formation depend on the atomic hydrogen concentration in the steel, it is of interest to estimate the hydrogen content of a canister in the repository. Atomic hydrogen may enter the steel by the dissociation of gaseous H_2 or as a result of the reduction of H_2O on the canister surface. For an H_2 partial pressure of 10 MPa at a temperature of 100 °C, JNC (2000) estimated a hydrogen concentration of $0.065 \mu\text{g}\cdot\text{g}^{-1}$. The concentration of hydrogen absorbed during corrosion can be estimated from hydrogen permeation measurements. For a range of dilute ($\sim 0.01 \text{ mol}\cdot\text{L}^{-1}$) HCO_3^- - based groundwaters, CHENG et al. (2000) report a maximum diffusible hydrogen concentration of $\sim 0.04 \mu\text{g}\cdot\text{g}^{-1}$. In addition, some hydrogen will be immobilized in trap sites within the steel matrix, but the total H content is likely to be $< 0.1 \mu\text{g}\cdot\text{g}^{-1}$. The presence of sulphide will increase the hydrogen concentration (sulphide poisons the H atom recombination reaction), with the addition of 10 ppm HS^- to the solution increasing the hydrogen content to $0.07 \mu\text{g}\cdot\text{g}^{-1}$.

As noted above, the severity of HSC increases with the strength of the material and the magnitude of the total stress. The grade of cast steel proposed for the canister is a relatively low-strength material, with a yield strength of 256 MPa (ATTINGER AND DUIJVESTIEN 1994). According to the data presented in JNC (2000), such a material would only be subject to HSC at a hydrogen concentration $> 10 \mu\text{g}\cdot\text{g}^{-1}$, one to two orders of magnitude higher than is possible in the repository. The possibility of HSC crack initiation can be reduced even further by heat treatment of the welds on the bottom and in the body of the canister and by shot peening of the final closure weld, the latter to introduce compressive surface residual stress and to peen potential stress-intensifying crack-like defects.

Blister formation appears to be equally unlikely for cast-steel canisters in a Swiss repository. The threshold hydrogen concentration for blister formation decreases with decreasing strength, with a lower threshold of $\sim 4 \mu\text{g}\cdot\text{g}^{-1}$ (JNC 2000). Although the threshold [H] for blistering is lower than that for HSC for low-strength steel, the value is still at least 10 times higher than is likely in the repository. Furthermore, blistering of the surface (where the hydrogen concentration will be highest) is unlikely to affect the structural integrity of the canister.

In summary, hydrogen damage is unlikely to affect the integrity of the canister, primarily because of the use of a low-strength material and because of the low hydrogen concentration in the steel.

Lifetime Assessment for a Carbon Steel Canister

The lifetime assessment for a carbon steel canister considers that uniform and pitting corrosion by oxygen during the aerobic phase would lead to a maximum wall penetration of about 11 mm. Corrosion due to sulphide, based on the assumption of transport being limited by low sulphide solubility and diffusive transport in the bentonite, may lead to about 0.02 mm of corrosion. Studies of the viability of sulphate-reducing bacteria (SRB) indicate that they will not contribute to corrosion in compacted bentonite. Anaerobic corrosion will result in about 10 mm corrosion in 10 000 years. This suggests that the total corrosion in 10 000 years is likely to be about 2 cm, i.e., only a small fraction of the ~15 cm wall thickness of the reference design discussed in Section 7.2. The question of residual stresses in the weld region and their interaction with possible stress-assisted failure mechanisms should be explored as the detailed design and welding techniques for the canister are further developed. Nonetheless, the possibility of failure by SCC or hydrogen-assisted damage at present appears remote, thus a lifetime assessment of at least 10 000 years appears reasonable.

7.1.2 Copper

The performance of copper canisters has been extensively treated in the context of Swedish (SKB 1992, 1999), Finnish (VIENO & NORDMANN 1996) and Canadian (JOHNSON et al. 1996) safety assessment studies. The following summary of the expected performance of a copper canister in crystalline and Opalinus Clay repositories is based on the comprehensive evaluation by KING et al. (2001). Before summarising the results from KING et al. (2001), the main differences between the repository conditions assumed in their assessment and those for potential Swiss repositories are noted.

Previous assessments of copper canister performance in the context of Swedish, Finnish and Canadian repository concepts have considered repositories in crystalline rock which have relatively low ambient temperatures at repository depth (10 to 15 °C). As noted in Table 2, this is considerably lower than for crystalline rock and Opalinus Clay in Switzerland, in which ambient rock temperatures at repository depth are ~35 to 45 °C. The consequences of this and the high average thermal output of the fuel in the Swiss case because of its high average burnup are that peak canister surface temperatures after emplacement would be significantly higher (JOHNSON et al. 2002). As noted in several studies, the most stringent thermal constraint for the engineered barrier system relates to the maximum temperature for the bentonite buffer (see discussion in JOHNSON et al. 2002), thus no thermal criteria for steel or copper canister materials have to date been specified. However, because the copper canister lifetime assessments summarised in KING et al. (2001) are for maximum canister temperatures of 100 °C, there remains a question of extrapolating the results to the higher temperatures of interest here. This question remains to be examined in detail, but no significant differences in the corrosion mechanisms or properties of copper are considered likely over the temperature range from 100 to 150 °C, thus the assessments presented by KING et al. (2001) are considered to be valid for the higher maximum temperatures of interest in the present study.

The suitability of copper as a canister material for deep geologic disposal of nuclear waste stems from (i) its thermodynamic stability under anoxic conditions and (ii) the tendency for copper to undergo uniform, rather than localized, corrosion in repository environments. Examination of the potential-pH (Pourbaix) diagramme for the Cu-H₂O system reveals an appreciable zone of immunity over a wide range of pH within the stability field of H₂O. Thus, although copper canisters will corrode due to the trapped O₂ in the buffer, corrosion should cease once all of this O₂ has been consumed. Under anoxic conditions, corrosion of copper is possible only if sulphide ions are present, in which case H₂O can act as an oxidant. Although copper does

undergo localized corrosion, the presence of high chloride concentrations ($>0.01 \text{ mol}\cdot\text{L}^{-1}$) promotes active (i.e., uniform) dissolution. Although bicarbonate ions can lead to passivation of copper (KING et al. 2001), active dissolution still predominates in chloride-based groundwaters. The groundwater $[\text{Cl}^-]$ for crystalline rock and Opalinus Clay (Table 2) are sufficiently high that the extent of localized corrosion of copper canisters in a Swiss repository should be minimal.

Uniform Corrosion

It is convenient to discuss the extent of uniform corrosion in terms of the evolution of redox conditions within the repository. Under oxic conditions, corrosion is supported by the reduction of O_2 on the canister surface. As discussed in Section 7.1.1, some of the trapped O_2 will be consumed by reaction with oxidizable minerals in the buffer, but a fraction will reach the canister surface. Because of the restrictive mass-transport conditions, the rate of corrosion will be limited by the rate of diffusion of O_2 through the buffer (KING et al. 2001). The corresponding anodic reaction will be the dissolution of Cu as soluble cuprous-chloride species (such as CuCl_2^-). Oxidation of Cu(I) to Cu(II) will consume additional O_2 .

The higher temperature in a Swiss repository will not affect the basic mechanism of the uniform corrosion of copper. The dissolution of Cu as CuCl_2^- has been shown to be the predominant anodic reaction in Cl^- solutions at temperatures up to 175°C (BACARELLA AND GRIESS 1973). The higher temperatures will lead to an increase in corrosion rate due to an increase in the rate of diffusion of O_2 to the container surface, but the overall extent of corrosion will still be limited by the amount of trapped O_2 . If anything, the higher temperatures in a Swiss repository will result in less canister corrosion because a greater fraction of the trapped O_2 will be consumed by reaction with oxidizable minerals at the higher temperature (KOLAR AND KING 1996).

Once all the O_2 has been consumed, uniform corrosion of the canister is possible only in the presence of sulphide ions. The concentration of HS^- is expected to be low ($<0.005 \mu\text{g}\cdot\text{g}^{-1}$) and is controlled by the solubility of pyrite (Section 5.4). Microbially produced sulphide can only be produced in the rock or at the buffer/rock interface (because of the lack of microbial activity in compacted bentonite) and will similarly have to diffuse to the canister surface through the buffer to cause corrosion. For these reasons, the corrosion rate will be determined by the rate of supply of sulphide to the canister surface (KING et al. 2001). Increasing temperature will increase the rate of diffusion, but an increase in temperature from 100°C to 150°C will only result in an approximate doubling of the rate (for a typical activation energy of $15 \text{ kJ}\cdot\text{mol}^{-1}$ for diffusive mass transport).

As discussed in Section 7, another consequence of the higher temperature may be an extended unsaturated phase. The critical moisture content for corrosion of copper will be the same as for steel canisters, since the condensation of moisture is dependent on the relative humidity and the presence of hygroscopic salts rather than the exact nature of the surface. KING et al. (2001) considered various vapour-phase corrosion processes, and concluded that corrosion during the saturation phase would be relatively benign. Once a thin moisture film has formed, but before complete saturation of the buffer, the rate of corrosion could be higher than in saturated buffer because of the faster rate of O_2 diffusion through vapour-filled pores. However, as discussed above in the context of the higher repository temperature, although corrosion will be faster under these conditions the maximum extent of corrosion will remain limited by the amount of trapped O_2 .

Pitting Corrosion

Although the pitting of copper is a well-known phenomenon, the repository environment is not expected to support extensive localized corrosion of a copper canister. Sustained pitting attack requires a continuous supply of oxidant and a passive, or at least a non-active, surface. In addition, the pitting of copper is favoured at room temperature, with general dissolution predominating at higher temperatures (KING et al. 2001).

Instead of discrete pitting, the surface of a copper canister is likely to undergo "uneven uniform corrosion." Copper coupons exposed to groundwater-saturated compacted buffer at temperatures up to 100 °C exhibit roughened surfaces with all areas attacked to some degree (KING & KOLAR 2000). This form of attack is consistent with a non-permanent separation of anodic and cathodic sites. Discrete pitting attack requires the permanent separation of the electrochemical reactions.

Other Potential Degradation Processes

Copper is known to be susceptible to SCC in certain, very specific, environments. Studies in several countries have shown that copper does not undergo SCC in either groundwater or simulated pore-water solutions (KING et al. 2001). Therefore, SCC is possible only if aggressive species such as ammonia, acetate or nitrite ions are introduced into the repository. In the absence of radiolysis effects, these species can be formed in a repository only through microbial activity. Since microbial activity is precluded by the use of highly compacted bentonite, there is no credible mechanism for the formation of an aggressive SCC environment in the repository.

Oxygen-free grades of copper are not susceptible to degradation by hydrogen.

Lifetime Assessment for a Copper Canister

Following the example of other programmes that have considered the use of copper canisters, the lifetime of copper canisters in a Swiss repository is estimated here based on the assumption of both uniform corrosion and pitting.

Uniform corrosion is assumed to occur during both the oxic and anaerobic phases of the evolution of the repository environment. Conservatively assuming that all of the trapped O₂ will support the corrosion of copper as Cu(I) results in a maximum depth of corrosion under oxic conditions of 0.070 mm. Under anaerobic conditions, the rate of corrosion is assumed to be $8 \times 10^{-6} \text{ mm}\cdot\text{a}^{-1}$, based on the estimate of WERSIN et al. (1994b) (Section 7.1.1) increased by a factor of 2 to account for the higher rate of diffusion of HS⁻ at the higher temperature in a Swiss repository. Therefore, for a period of 10⁵ a, the maximum depth of uniform corrosion is estimated to be 0.87 mm.

Pitting is treated using a pitting factor, with a conservative estimate of 5 (KING et al. 2001). Thus, the maximum depth of uniform corrosion and pitting in 10⁵ years is equal to $5 \times 0.87 \text{ mm} = \sim 4.4 \text{ mm}$.

Assuming the same minimum wall thickness of 50 mm as defined for the SKB canister, the lifetime of a copper canister in a Swiss repository is $>10^5$ years. The estimated lifetime is consistent with lifetimes for copper canisters predicted in other programmes (KING et al. 2001).

Because the copper shell is not a structural member, the results above also imply that 50 mm is substantially more than is required for protection against all forms of corrosion and that a 20 to 30 mm wall thickness would be sufficient for a lifetime of $>10^5$ years. It is expected that the selection of a thickness within this range would be determined by mechanical integrity requirements related to stresses induced in the shell during handling.

7.1.3 Alternative Canister Materials to Limit Hydrogen Gas Production

As noted in Section 5, the use of canisters constructed from cast iron or carbon steel in the case of a SF/HLW repository in low permeability host rock raises questions about the significance of hydrogen gas production and buildup and the potential impact on system performance and safety. Although there are good reasons to believe that gas production is unlikely to compromise safety (NAGRA 2002a), it is nonetheless clear that the need to analyse all the impacts of gas production complicates the development of clear safety arguments, simply by virtue of the complexity of the models required and the increased data requirements. As noted earlier, the use of copper as a canister material surrounding a steel insert would ensure that production of H_2 in the repository would be greatly delayed². The use of ceramic canister materials (e.g. alumina) would similarly be effective, although residual questions about delayed cracking remain unresolved (JOHNSON et al. 1994). Another alternative would involve the use of a thin shell (~1 cm) of highly corrosion-resistant material such as Grade 16 titanium or Hastelloy surrounding a load-bearing insert. These alloys have extremely low general corrosion rates ($< 0.01 \mu\text{m a}^{-1}$). The cost of such materials is high (e.g. the cost of a 1 cm shell of a Ni or Ti alloy would be 3 to 5 times the cost of a 3 cm shell of Cu). It is also unclear if the adoption of such materials would provide the same canister lifetime potential as does a copper shell.

The use of cast bronze instead of cast steel for the insert in the case of a copper canister would essentially completely eliminate H_2 gas buildup in the near-field. The cost increase is significant (a factor of at least 5 in raw material costs for cast bronze vs. cast steel), but another significant concern is the limited capability for bronze casting for structures of this scale (BOWYER 1998). It would be possible, of course, to cast the insert in two or three short sections that are then welded together.

It should also be noted that the advantage of a potentially large capacity for scavenging radiolytic oxidants could be lost in switching from a steel to a bronze matrix, because of the inability of Cu to effectively reduce U(VI), Np (V) and various redox-sensitive fission products.

7.1.4 Probability of Premature Breaching and Breaching Mode

As discussed in Section 6.1, the SKB criterion (WERME 1998) of < 1 in 1000 canisters having defects that would be larger than acceptance specifications sheds little light on the significance of a defect in terms of long-term performance of the canister. For example, weld defects may exist that are too small to detect with the inspection technique, but there may be, nonetheless, some probability of a through-wall breach developing. In addition, it is reasonable to assume that some defective canisters might pass inspection because weld defects that are larger than the acceptance specifications go undetected because of process failure or operator error. There is presently no basis for estimating the proportion of such initially defective canisters that might be produced. This requires the gaining of considerable experience with the material in question, in particular the canister manufacturing and welding techniques and the development and

² The production of H_2 as a result of oxidation of Cu by water in the presence of sulphide can occur. At the corrosion rate supported by sulphide diffusion to the canister ($8 \times 10^{-6} \text{ mm a}^{-1}$), the gas production rate would be 300 to 400 times lower than for carbon steel.

refinement of a suitable inspection technique. As a result, the estimation of an initial canister defect rate for performance assessment calculations must be performed in another fashion. The most appropriate basis for such an estimate would appear to be from experience with pressure-retaining components that are carefully inspected. The probability of undetected manufacturing defects leading to an early through-wall breaching has been estimated to be 1 in 5000 in Canadian studies (DOUBT 1984). This estimate is based on the actual number of failures observed for highly inspected pressure-retaining components (e.g. pressure vessels and CANDU reactor pressure tubes). This merely represents the starting point for deriving a suitable value for performance assessment calculations, as various aspects related to manufacturing techniques and inspection limitations may argue that lower or higher values could be adopted. In the absence of other data, but considering the high end of the data discussed by DOUBT (1984), it is recommended that for Nagra performance assessment calculations, a case involving breaching of a single Cu canister upon emplacement (~1 defect per 2000 canister) be considered. Such breaching could be considered to be of the pinhole type, with a maximum diameter of 1mm.

A limitation of the steel canister in terms of long-term performance is the difficulty of defining the conditions for modelling the role of the residual canister (and corrosion products) after canister breaching, in particular its impact on mass transport of nuclides into the buffer. Although it is apparent that transport limitations would arise, it is difficult to quantify the size and number of penetrations that would occur. It is possible that the breach mode might involve ductile rupture after significant wall thinning, or the remote possibility of a stress-corrosion crack of the weld region. This contrasts to the case of a copper canister, where for the case of a prematurely defected canister, radionuclide transport would occur from a single defect, the size of which is likely to remain constant for many thousands of years, allowing modeling of such transport limitations to be used in assessments of copper canisters (see Section 5.1).

For Cu canisters, a hole defect in the weld region appears to be the only reasonable assumption regarding breaching. Adoption of a value of 1 in ~2000 for prematurely breached canisters for performance assessment calculations does not suggest that this rate would actually be expected to occur. Given improvements in manufacturing and inspection technology, it is reasonable to assume that this value is rather pessimistic.

7.2 Canister Design Options

The structural strength requirements for the canister argue for a high strength material either as the internal structural member (e.g. the cast iron of the Cu/cast iron canister of SKB (1999)), or as the shell itself. The following concepts are based on these two approaches.

Design Concept 1 – Carbon Steel

Concept Development

The use of a cast iron or steel insert in a copper canister, as proposed in WERME (1998) gave rise to the concept of simply adopting the insert itself as a canister. Increasing the effective wall thickness of the insert from 5 cm in the SKB design so as to provide the required corrosion allowance (an additional generous allowance of ~10 cm wall thickness was assumed) would then give the basic canister design parameters. Subsequent examination of the difficulties encountered in casting the massive insert from carbon steel as a single unit (ANDERSSON 1998) suggested that other fabrication technology should be considered. The use of cast iron instead of carbon steel was not considered because of concerns regarding the long-term integrity of the final closure weld.

Discussions regarding manufacturing techniques indicated that it would be feasible to cast the canister in three sections as solid bodies, followed by machining a central square cavity and welding the sections together. In order to permit insertion of fuel assemblies, cross-plates could be inserted into machined grooves to provide the required number of fuel assembly positions, as illustrated in Figures 7 to 9. A canister base would be welded on, followed by annealing the canister and milling the surface to improve inspectability. Both the canister lid and base are assumed to have a thickness of 13.5 cm. The lid design is shown in Figure 9, which also shows a proposed lifting ring for handling. The actual canister lengths would vary from 4 to 4.9 m, depending on the fuel assembly types in question, thus a 5 m length was considered in the feasibility study. The mass (empty) would be ~26 t for the case of 4.9 m length canister for disposal of 9 BWR assemblies. Preliminary Structural Analysis

Preliminary Structural Analysis

The structural performance of the canister was examined by creating a finite-element model of the canister and evaluating the steady-state stresses as a function of wall thickness using the ANSYS code. Decreased wall thicknesses are assumed to mimic the effect of slow uniform corrosion of the outer canister surface. Canister failure is postulated if the tensile stresses near the corner of the quadratic cavity exceed 100 MPa. Under such circumstances, propagation of small cracks (with an assumed initial crack depth of 0.5 mm) would occur from the outer canister surface into the canister wall, leading eventually to canister failure. The stresses in the wall were calculated for two isotropic loading cases at pressures of 30 and 40 MPa. The details of the analysis are given by SPRYSL & GRIBI (2002). The results indicate that a wall thickness of > 4.2 cm is sufficient to withstand a load of 40 MPa, thus there is a considerable corrosion allowance available for the reference wall thickness of ~15 cm. (Note that a maximum corrosion depth of <2 cm in 10 000 years was estimated in Section 7.1.1, thus there is considerable scope for defining a reduced wall thickness, which would simplify manufacturing). Further structural analysis calculations for anisotropic loading scenarios including stresses in the head-to-shell region are required, although based on the analysis done in Sweden for the copper/steel canister, the conclusions are not likely to be dramatically affected, i.e. a 5 cm effective wall thickness is still more than sufficient to deal with all anisotropic loading scenarios considered (WERME 1998).

The final weld would result in a region of high stresses surrounding the weld region. In many applications, this would be dealt with by annealing to reduce stresses. This does not appear to be a feasible prospect in the case of a SF canister³, as this would require heating the entire canister to a temperature of 600 to 650 °C. This exceeds the maximum permissible cladding temperature for dry storage of ~400 °C, established to prevent creep rupture from occurring. As noted in Section 7.1.1, despite the stresses being high, the likelihood of SCC occurring is very small. Stresses can be further reduced by shot peening the weld region as discussed in Section 7.1.1. Nonetheless, some further examination of sealing concepts appears warranted, including the use of a threaded lid seal, followed by a weld, as in the case of the HLW canister (NAGRA 1994).

³ A similar conclusion is reached for HLW glass, where there is concern regarding devitrification at temperatures exceeding 500 °C

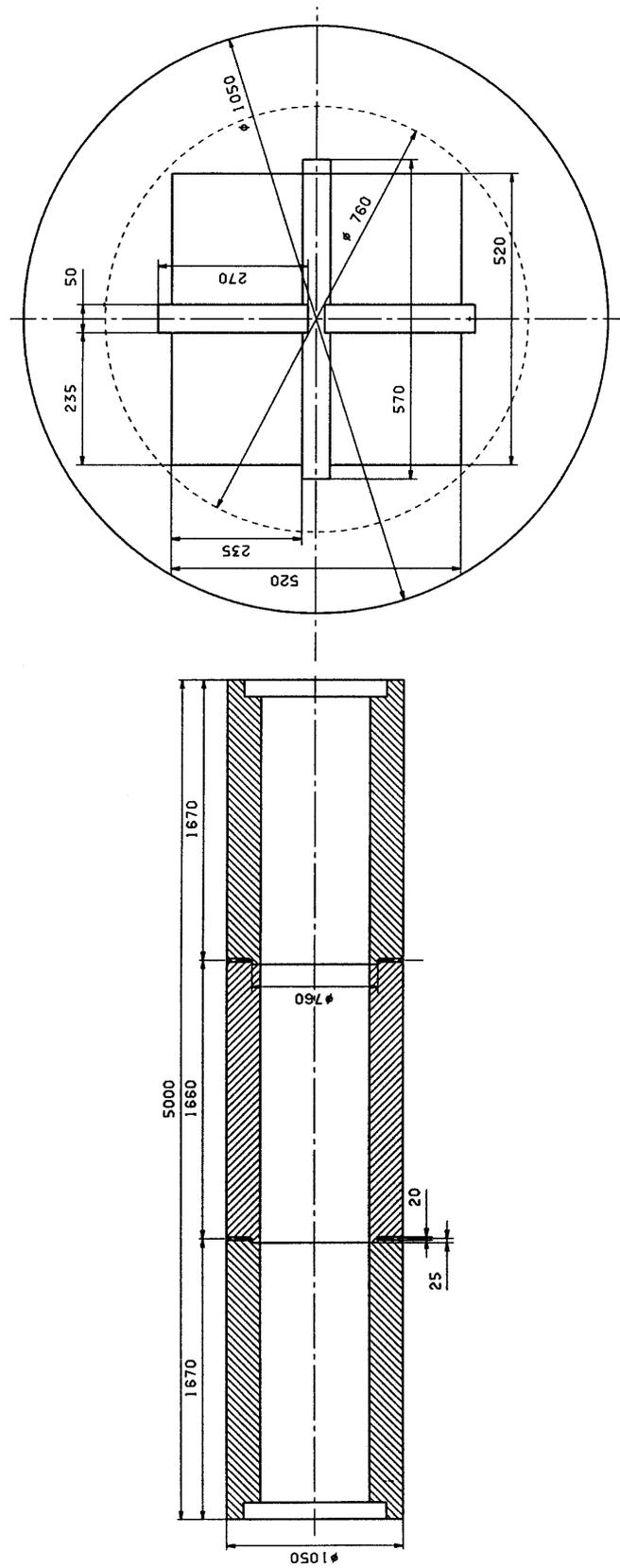


Fig. 7: Cross-section of steel canister for 4 PWR fuel assemblies

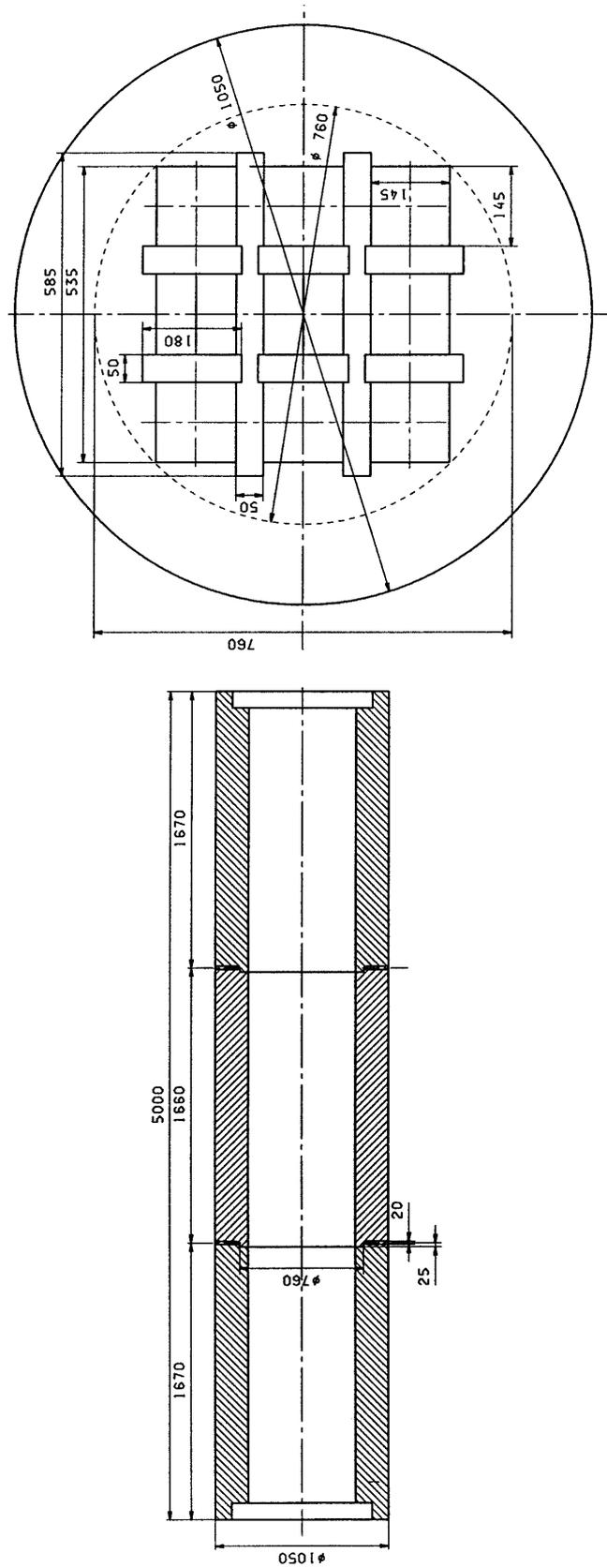


Fig. 8: Cross-section of steel canister for 9 BWR fuel assemblies

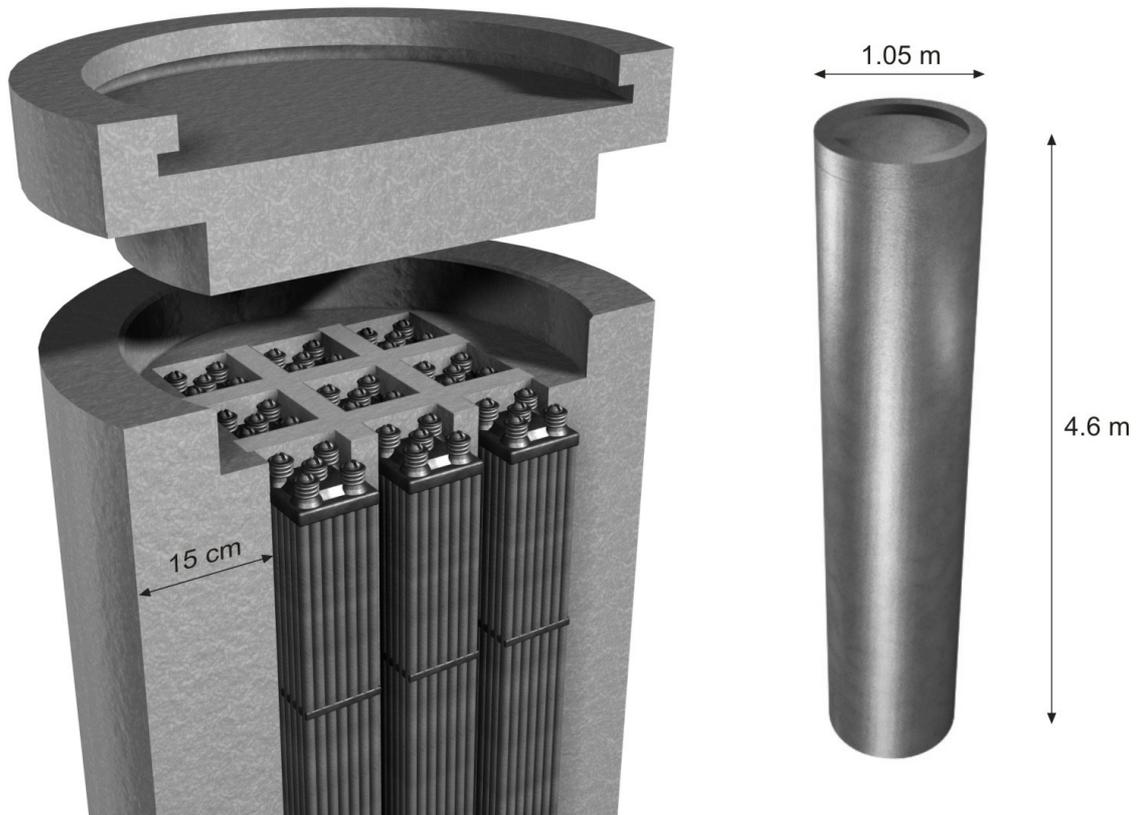


Fig. 9: Carbon steel canister for the disposal of spent BWR fuel

Design Concept 2 – The SKB Cu Canister with Cast Iron Insert

The SKB reference design based on this concept is shown in Figure 10. The feasibility of casting such inserts at full scale has been demonstrated and the procedure is presently being refined (ANDERSSON 1998, BOWYER 1998). Nodular cast iron is both easier to cast and less expensive than mild steel and is now the SKB reference material for the cast insert. The lid would likely be bolted on in the case of a cast iron insert. This approach is preferred because the insert is not a containment barrier, thus the simplest sealing technology should be adopted. Although welding is feasible, the post-welding heat treatment required could expose the fuel to temperature conditions that could affect fuel cladding integrity during the encapsulation operation. The inserts would be modified for four PWR fuel elements and nine BWR elements for disposal of Swiss SF. The load-bearing insert of the SKB design has a collapse pressure of 80 MPa (WERME 1998) (note that this is for a design in which the minimum thickness of steel between the fuel channel and the periphery is 50 mm), far in excess of the maximum load of ~30 MPa required for a repository in either crystalline rock or Opalinus Clay. The outer shell would be copper, with a thickness of 30 to 50 mm, the value probably depending on strength requirements related to lifting and handling rather than to the corrosion allowance requirement, as noted in Section 7.1.2. The canister would not be expected to fail by corrosion, as total corrosion depths of < 5 mm have been conservatively estimated (see Section 7.1.2). A canister of this design would weigh ~25 t and in principle could be adopted for in-room or in-floor emplacement in the repository.

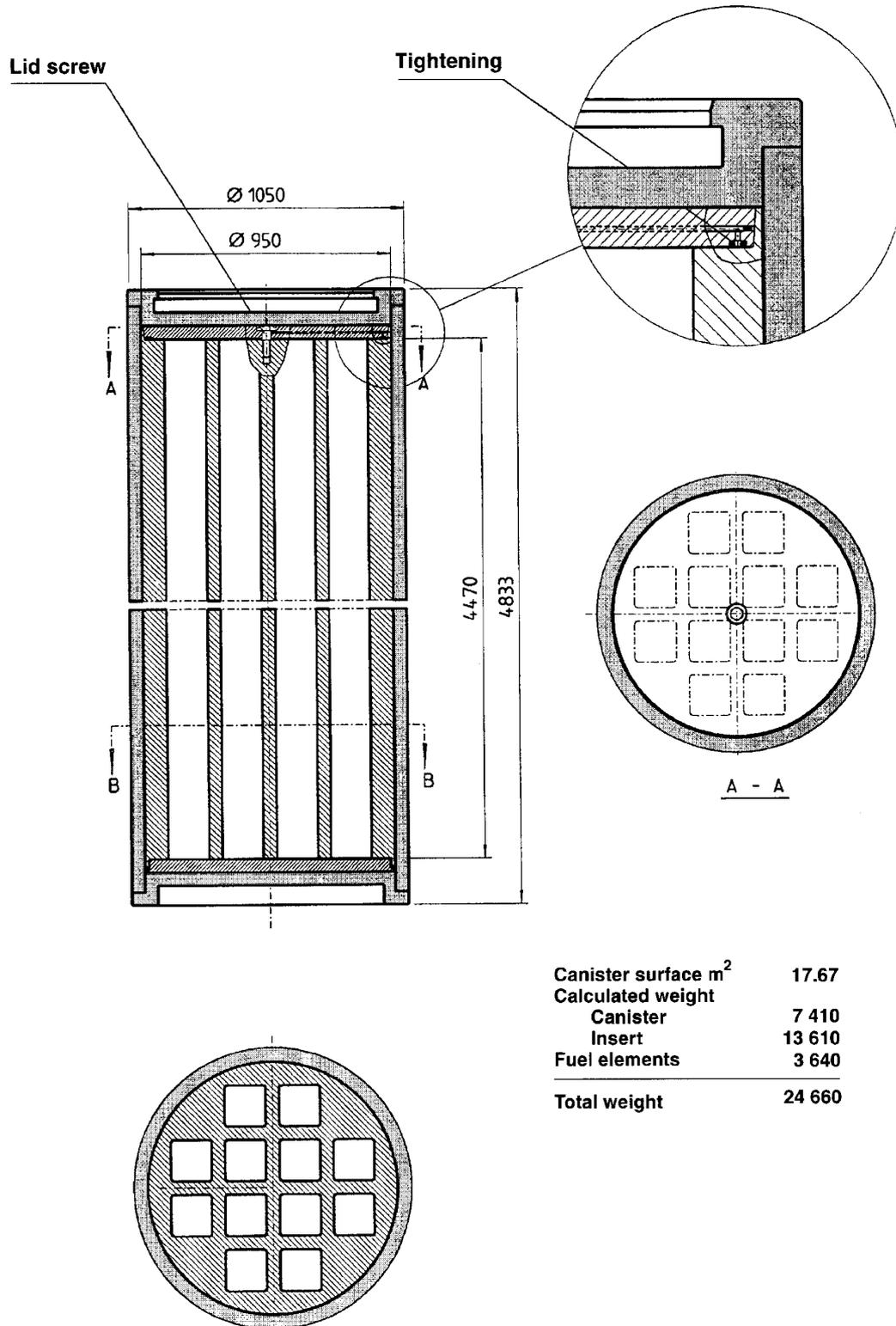


Fig. 10: Reference Cu/steel canister design of SKB (SKB 1997)

8 Conclusions

The design and performance requirements for canisters for the disposal of Swiss power reactor SF have been outlined. Concepts for canisters with 1000 a and 100,000 a lifetimes have been described and the performance issues for these canisters have been reviewed. It is concluded that, should system assessment studies show that a 1000 a lifetime is sufficient, there are reasonable prospects for adopting a cast steel canister as one design option. For long-term containment, the reference copper/cast iron canister of SKB is recommended. A variant of this design, using a cast bronze instead of a steel insert, could also be considered as a potential option.

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