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Comments on "SKB RD&D-Programme 98" Focused on Canister Integrity and Corrosion

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In order to avoid confusion of chapter numbering between the present document and the reviewed, there is no numbering in the present document.

Abstract

According to the Act on Nuclear Activities the nuclear utilities are requested to submit a comprehensive programme for research and development every third year, aiming at the safe storage of radioactive waste produced by the nuclear power plants. The latest was published by SKB in September 1998 and is called the ”RD&D Programme 98”. The work presented in the present report was commissioned by SKI and is a result of reading the ”RD&D Programme 98” and related reports with focus on canister production, integrity and corrosion.

We find that those parts of the programme often are difficult to follow owing to the lack of detail in the Programme and in one of the supporting reports. In our opinion this will make the work difficult to monitor by SKI and SKB. We also feel that the interpretation of information already available often is overoptimistic. As a consequence the difficulties ahead are understated and the programme is allowed to converge too quickly.

We agree that the materials choices for both the inner and outer canisters are appropriate providing they both can be produced commercially and in a satisfactory metallurgical condition, that they can be quality assured and that no further unforeseen difficulties arise. We also agree that alternative technologies merit consideration for production of the outer canister and that alternative joining processes should be studied. We are actually concerned that greater prominence is not given to the alternatives in the programme.

We believe that it should be possible to develop a satisfactory canister for disposal of high level nuclear waste according to the general method proposed by SKB and with the proposed capacity within the time-scale of the overall programme. We do not believe, however, that all the difficulties have been recognised. As a consequence of this the results to date are interpreted optimistically. We believe that progress should be subjected to more professional review within SKB and that a higher level of metallurgical support is required.

We disagree that suitable full size canisters have already been created and that production technology is available for both canisters at full size. We also disagree that the long time durability is ascertained. I. a. it is easy to find corrosion mechanisms and handling procedures for the canister system that have to be demonstrated not to be harmful.

We feel that there are many areas, which need further evaluation but are granted too little space in the programme. This is valid for i.a. effects of non-uniform loading and creep, welding, quality control, effects of radiolysis and corrosion properties. We also consider that more information should be provided on the detail and timing of the development plan for the trial fabrication programme of the canister, the canister test programme, determination of quality standards and development of non destructive testing procedures.

We also feel that insufficient emphasis has been placed on the further development on alternatives to high power electron beam welding, non-destructive testing and over all handling.

Copper will be exposed for both general and different kinds of localised corrosion in the repository. The complex mechanical, chemical and microbial environment with high pressures varying in time and location and with oxygen, chloride, sulphur and carbon bearing compounds present will cause different types of attacks that are going to prevail during different time periods.

The procedures of production, handling and treatment of the canister throughout the processes of filling, transportation and deposition are crucial for its later, corrosion-related integrity throughout the storage period in the repository. There is a risk that due to systematically induced faults, many canisters may have later corrosion related problems. The QA system should be developed to cover all steps of canister handling.

We feel a large uncertainty when expressions like "known corrosion processes" and "sufficient" safety factors are used in the reasoning, especially as the corrosion part throughout the series of RD&D Programmes to our opinion has been allowed to diminish too rapidly.

The conclusive part of the present report contains about forty points of detailed comments on the RD&D Programme 98.

Sammanfattning

Enligt kärntekniklagen krävs att de svenska kärnkraftproducenterna lägger fram en plan för forskning och utveckling vart tredje år. Planen skall inriktas på säkert förvar av det radioaktiva avfall som produceras av kärnkraftverken. Den senaste planen publicerades av SKB i september 1998 och kallas "FUD-program 98". Det arbete som presenteras i föreliggande rapport har utförts på uppdrag av SKI och är ett resultat av granskning av "FUD-program 98" och underliggande rapporter med fokus på tillverkning, integritet och korrosion hos avfallsbehållaren.

Vi tycker att de delar av programmet som vi granskat ofta är svårpenetrerade beroende på bristen på detaljer bl a i kapitel 7 i programrapporten och i en av underlagsrapporterna. Vi menar att detta faktum kommer att försvåra både SKIs och SKBs egen uppföljning. Vi tycker också att tolkningarna av föreliggande information ofta är överoptimistiska, att framtida svårigheter underskattas och att programmet därför tillåts konvergera alltför snabbt.

Vi samtycker till åsikten att materialvalen till både den inre och den yttre behållaren är godtagbara under förutsättning att båda kan produceras kommersiellt och i ett metallurgiskt godtagbart tillstånd, att de kan kvalitetssäkras och att inga ytterligare svårigheter uppstår. Vi samtycker också till att alternativa sätt att producera den yttre behållaren förtjänar att övervägas och att alternativa förslutningsprocesser bör studeras. Vi ställer oss frågande till varför inte större utrymme ges åt alternativen i programmet.

Vi anser att det bör vara möjligt att utveckla en tillfredsställande bra behållare för högaktivt avfall i enlighet med de allmänna metoder som SKB föreslår. Vi tror också att detta kan ske med föreslagen produktionskapacitet inom det allmänna programmets tidsram. Däremot tror vi inte att alla svårigheter ännu har förutsetts. Konsekvensen av detta är att de FUD-resultat som redan uppnåtts tolkas alltför optimistiskt. Vi anser att framstegen fortlöpande bör underställas en mer professionell och självkritisk granskning inom SKB och att en högre nivå av metallurgisk support behövs.

Vi ansluter oss inte till åsikten att en fullstor behållare redan har tagits fram och att produktionsteknologier redan finns för fullstora behållare. Vi samtycker inte heller till åsikten att den långsiktiga hållbarheten är säkerställd. Bland annat är det lätt att finna exempel på korrosionsmekanismer och hanteringsmetoder för behållaresystemet där bevis måste tas fram att de inte inverkar negativt på den långsiktiga säkerheten.

Vi anser att det föreligger många områden som är i behov av ytterligare utvärdering men ges ett alltför litet utrymme i programmet. Detta gäller bl a effekterna av anisotrop last och kryp, svetsning, kvalitetskontroll, radiolyseffekter, korrosionsegenskaper, mm. Vi tycker också att mer information bör tillhandahållas vad avser detaljer och tidplaner i programmet för provtillverkning. Detta gäller också utveckling av metoder för icke förstörande provning.

Vi tycker dessutom att otillräckliga insatser gjorts för att ytterligare utveckla alternativ till högeffektselektronstrålesvetsning, gjutning av insats, oförstörande provning och allmän hantering.

Koppar kommer att exponeras för både allmän och olika typer av lokal korrosion i förvaret. Detta är inte märkligt med tanke på den komplexa mekaniska, kemiska och mikrobiella miljö som kommer att råda och som dessutom kommer att variera i tid och rum. Olika tidsperioder och olika positioner i förvaret kommer därför sannolikt också att uppvisa skilda typer av angrepp. Dessa komplexa förhållanden borde utredas bättre.

Vi anser att den allmänna hanteringen av kapslarna vid produktion, transport och deponering i förvaret är kritisk för deras senare korrosionsbeteende. Det finns en risk för att systematiska fel någonstans i den totala hanteringskedjan kan orsaka att många kapslar senare går sönder för tidigt genom en kombination av hanteringspåverkan och korrosion. QA systemet bör vidareutvecklas för att säkert täcka hela hanteringskedjan.

Vi känner en stor osäkerhet när uttryck som "kända korrosionsprocesser" och "tillräckliga säkerhetsfaktorer" används i resonemangen, speciellt som korrosionsdelen enligt vår åsikt tillåtits att minska alltför snabbt i programserien.

Slutsatsdelen av föreliggande rapport omfattar ca fyrtio detaljerade kommentarer till FUD-program 98.

Background

General

According to the Act on Nuclear Activities (SFS 1984:3) the Swedish nuclear utilities are requested to submit a comprehensive programme for research and development aiming at the safe storage of radioactive waste produced by the nuclear power plants. The programme is supposed to be submitted every third year and the latest was published in September 1998 [1]. SKB is the utility owned organisation responsible for the programme.

The Nuclear Power Inspectorate (SKI) is the government agency responsible for reviewing the programme. This process is founded on internal SKI reviewing, considerations by a set of organisations with technical or societal etc. competence and also by consultants.

In the present report comments on the programme are accounted by the authors from the point of view of canister production, integrity and corrosion. The reports reviewed are the "RD&D Programme 98" [1] and related background reports, namely [2, 3].

The present SKI report is also registered as STUDSVIK/M-99/1 in the Studsvik Material AB report series.

Previous RD&D Programmes

The previous RD&D Programmes (92 and 95) [4, 5] and our comments on those [6, 7] also form an important foundation for the reviewing of RD&D Programme 98 [1]. The previous comments form a background to the evaluation of the present programme, as it is possible to point out differences as well as interactions between comments on the old programmes and the formulation of the new.

In our previous comments on canister integrity and corrosion, see Hermansson et al [6] and Bowyer and Hermansson [7], some important conclusions were made. We are concerned that many main conclusions made on previous programmes are still valid for the RD&D Programme 98. Some important of those are listed here.

- First of all we want to underline that we think the RD&D Programmes, including RD&D Programme 98 are very impressive.

- On the other hand the amount of information is generally too large to comprehend by reviewers in a short time. One obvious consequence of this statement is that SKB cannot be morally free from responsibility for any features, events and processes that could happen whether reviewers commented on it or not.

- It was difficult in RD&D Programmes 92 and 95 [4, 5] to follow how decisions were made relating to programme formulation, etc.

- Irrespective of the movement in importance from R to D, doors should be kept open for qualified R to be able to face future surprises. It was our feeling that R was declining too rapidly in RD&D Programme 95.

- The multi-barrier principle is important. This is still valid but SKB persists in putting most emphasis on the canister. This focuses a very large responsibility on the canister integrity and the requirements that it has to fulfil.

- In RD&D Programme 92 the problem of transfer of scientific knowledge to the practical constructor was emphasised. This problem is still valid and has not yet been addressed by SKB.

- The question of canister corrosion was not considered to be fully understood in RD&D Programme 92 and the writers could trace a tendency of neglect in RD&D Programme 95. This statement is still valid for RD&D Programme 98.

Summary of RD&D Programme 98

The programme [1] briefly describes finished and planned R&D work on encapsulation, storage in a deep geological repository as well as supporting research, development and demonstration.

There are descriptions of guidelines for waste treatment, applicable law, existing nuclear installations for waste handling as well as brief comments on previous programmes.

The programme focuses on the concept of stepwise development of the waste handling system with a deep geological repository as well as on principles and requirements on the knowledge base, radiation protection and safety and on the barrier functions. There are descriptions of the status of the knowledge base for long term safety and for the canister and encapsulation technology. These parts are followed by descriptions of the RD&D programme for the canister and encapsulation, deep repository, safety analyses, supporting RD&D, the programme for the Äspö-lab, for alternative methods and for decommissioning of nuclear installations.

Details of the programme parts reviewed here (mainly canister production, integrity and corrosion) are found mainly in documents [1-3].

Scope and time schedule of the present review

The present task of commenting on the RD&D Programme 98 was commissioned by SKI on Dec. 2, 1998. The intention was to focus mainly on section 7 in the programme and those parts of sections 8 and 9 that are concerned with the canister for disposal of high level nuclear waste. Consequently other chapters are only dealt with very briefly.

The work has been carried out by reading relevant parts of the RD&D Programme 98 [1] and a selection of supporting basic reports, mainly [2, 3].

On Dec. 7-11, 1998 the authors worked together at Studsvik in order to prepare a manuscript of the present report.

Review of the RD&D Programme 98

General

The contract from SKI was focused on chapter 7 and parts of 8 and 9 of SKB RD&D Programme 98 [1] and consequently the main effort is made there. However, minor notes will also be found on other chapters. Beside the main document [1], a couple of related background reports, mainly [2, 3] were also reviewed in an integrated manner.

The introduction of [1], on which the writers have no specific comments, is focused on the guidelines for the waste handling in Sweden. These say that the waste generated by Swedish nuclear power shall be handled inside Sweden. There will be no reprocessing and there are high demands on security, radiation protection and safeguards. The problems with the waste should be solved by generations using the nuclear electricity. Decisions about repository design should be founded on a broad knowledge base. Technical solutions and design should be found within Sweden but supported by foreign knowledge. Review by the authority will be for guidance and the information flow will be open to public.

The goal of the RD&D Programme 98 is, by fulfilling all technical, environmental and security demands, to begin the deposition of a smaller part of the used fuel in a deep rock repository within about 15 years. The fulfilment of this goal requires that an encapsulation plant, a deep repository as well as a transportation system are in operation at the time. The storage is supposed to be carried out according to the KBS 3 concept [8] or a closely similar, optimised concept.

Typical components of the concept are the copper canister and the deep repository at a depth of 500 m in crystalline bedrock.

The authors feel that the goal of the RD&D Programme 98 is clear. However, even though we think that the RD&D Programme 98 is an impressive document, we have many reservations i.a. regarding technical details and the time-scale of the programme. Those details will be discussed in following sections.

Chapters 1 through 4 in part 1 of [1] are of a general nature and are not commented on by us. In Part 2, chapter 5 is only commented on in a general way, mainly concerning time plans. Chapter 6, localisation, was not part of the commission from SKI. Neither is chapter 10, decommissioning, commented on. This leaves the emphasis of this review on chapter 7 and to some extent on chapters 8 and 9.

The foremost requirement of the repository is to isolate the waste and the canister is responsible for the direct isolation. The buffer and rock have retarding tasks. We therefore feel that the canister integrity is very important and the highest demands must be placed on its proper function.

As long as the canister is tight all leakage of radioactivity is hindered. In order to reach this canister quality it is said that the canister must be tight at the deposition, resist the expected chemical environment in the repository and withstand the mechanical loads expected there.

We will point out later that the chain of operations suggested by SKB to bring the canister from production to its final place in the repository does not guarantee its integrity. We will later give serious comments on mechanical strength, QA system and corrosion resistance.

The final design of the canister is a result of a stepwise development resulting from both theoretical analyses and practical tests. We feel that it is important to still keep all doors open and also to be prepared to perform a fundamentally different design. Arguments for this will be given in later parts of this report.

The canister is supposed to withstand all known corrosion processes and remain intact in the repository during at least 100,000 years. It shall also maintain its integrity under the different types of physical loads generated at a depth of 700 meters in crystalline rock. Some main objections will be presented later in cases where we feel that SKB has not taken all important scenarios under consideration and also made conclusions that we feel are debatable. We also think that a serious discussion should be made of the way of handling the unknown scenarios. A general trend is to account for this by using safety factors. Those are often given without presenting a qualification, which leaves a strong feeling of uncertainty.

A QA system to assure the canister integrity from mechanical and corrosion points of view valid for all steps from canister production to the very final deposition in the repository should be thoroughly discussed. At present such a QA system is indicated only for the production and encapsulation steps. Little, if any, is said about QA of transportation and handling at the repository site.

Copper is selected as the corrosion barrier of the canister. This selection is based on "judgements" of copper lifetime in the repository. We think that the foundations for these judgements are not very well presented and the lifetime of the canister will depend dramatically on how the canister is influenced by the different production and handling steps as well as by the repository environment.

Development and design of the canister

Introduction

Werme [2] details the design premises for the canister. He points out that the requirements for safety during encapsulation, transport and emplacement may be assisted by protective measures but safety after final disposal depends on integrity of the canister through life. Life is not defined but Werme says that no known corrosion process should be able to violate the canister for 100,000 years. This appears as a departure from earlier statements when lives of up to 1,000,000 years have been proposed. Further, reference to known corrosion processes is very limited. We believe that the expression should be "no known process which may occur in the repository environment", and the lifetime specification is for others to comment on.

Chemical Considerations

There appears to be a conflict in the requirements presented by Werme [2]. In section 5.1.1 he indicates that sulphate reducing (SRB) are non-viable in bentonite when the density exceeds 1800 kgm^{-3} or higher and concludes that such bacteria therefore do not present a problem. Examination of the reference paper [9] confirms that SRB are destroyed in bentonite when the water activity is less than 1 (i.e. it is unsaturated). Under his experimental conditions (bentonite compaction pressure unspecified, applied hydrostatic pressure 2 MPa) such an activity was produced when the density exceeded 1800 kgm^{-3}.

In appendix 1 in [9], the assumption is made that the bentonite in the repository will be saturated and in order to control the swelling pressure it will be necessary to use a bentonite with a dry density of 1590 kgm^{-3}. From the information supplied in the two papers [2, 9] it is not possible to determine whether or not there is an overlap between the two conditions (dry density less than 1590 kgm^{-3} and density greater than 1800 kgm^{-3}).

It does seem that Pedersen [9] is saying that under water saturated conditions SRB will thrive and that Werme is saying that water saturated conditions will exist. Pedersen also says that under his experimental conditions bentonite of density 1800 kgm^{-3} is unsaturated.

We consider that the evidence supporting the assertion that the SRB will not survive in the repository environment is inconclusive and the assertion is also inconsistent with the statement that the bentonite will be water saturated. It also seems possible that the degree of compaction of bentonite necessary to suppress SRB would result in unacceptable bentonite swelling pressures. We consider that this area needs further examination.

Mechanical Integrity

Cast iron

Werme [2], section 5.1.2 refers to mechanical conditions in the repository. It is stated that the design hydrostatic pressure is 7 MPa to which must be added pressure from swelling of the bentonite. The latter has been set to 7 MPa although the evidence (in the Werme paper) suggests that this low value has not yet been achieved. Swelling pressure is clearly a complicated function of original moisture content, compaction pressure and the filling density around the canister. We believe that further work is required to determine the swelling pressure that may be expected when bentonite blocks are made by practical methods and this value should be used for the design calculations.

Werme states that calculations for a cast canister with internal support, according to the current concept, have been performed. The critical stress for failure based on ideal properties in the cast iron (Werme section 5.2.3) are given as 81 MPa for the BWR version and 114 MPa for the PWR version and these appear to offer good safety margins over the proposed design basis case (44 MPa). However all the stresses appearing in the cast iron liner will be transmitted through the copper overpack. The yield strength of the copper is given as 50 MPa and the calculations for the unsupported lid indicated tensile stresses exceeding 250 MPa for a pressure in the repository of 27 MPa. It seems probable that, on the lid, and near corners, the yield stress of the copper could be exceeded at very modest repository pressures, even for the current design of insert. This may seriously damage the system for lifting and retrieval, which is provided for in the design of the lid.

Measurements made on specimens cut from cast inserts indicate that the yield strength quoted as ideal for the iron is exceeded (Andersson [3] section 4.5.3). Yield strengths in the range 270 to 290 MPa are given at the cost of a reduction in ductility. The changes in ductility reported may be acceptable and the improved yield strengths provide an extra safety margin. It is our view that it will be necessary to carry out sufficient tests on material cut from cast inserts to give a confident determination of the range of material properties which occur in real castings and to redetermine the critical stresses using these figures. The variations in microstructure within single castings and between castings and the location, type and frequency of occurrence of casting defects should be determined as part of the same exercise. This information may be used in setting realistic safety factors.

We consider that it is necessary to pay further attention to the structural design in the context of the load cases described by Werme [2] (section 5.2.3.).

We agree that nodular iron is the most appropriate choice of material for the cast component and we recognise the benefit of the improvement in strength of the insert conferred by the supporting internal structure. This means that for the uniformly loaded pre-glaceation case the safety margins are substantial. (Critical pressure for failure 81MPa, design pressure 15 MPa x safety factor of 2.5=37.5 MPa). For the case during an ice age with an ice cap 3 km thick and with uniform pressure the total pressure, according to Werme is 44 MPa, the safety factor for this case is thus 81/44=1.84. This is less than the figure of 2.0 quoted by Savas [10] as a minimum in the reference work used by Werme.

We consider this to be inadequate and that it may be further degraded when a better estimate of bentonite swelling pressure is available.

When the figures generated by Werme are used, the case for bentonite swelling on one side only has a safety factor of 1.9. Two further cases (cases 4 and 5) of uneven swelling in the bentonite place unacceptable loads on the canister. Consideration of the properties of bentonite and more careful analysis is said to demonstrate that the maximum tensile stresses are contained to values of less than 55 MPa, which could be quite acceptable. Whilst it is easy to justify the alternative calculation method and to believe the result, we are left with a feeling of discomfort. We know that the properties of the bentonite are uncertain. We do not know what values have been used and whether or not they are conservative. We know that the forces on the liner are transmitted through the copper but we have no indication of how the copper is responding.

Two further cases (6A and 6B) of uneven swelling in the bentonite do not challenge the strength of the liner. Four further cases are also considered.

It is our opinion that the results of these calculations should be treated as preliminary indications. Further calculations are required. Load cases should be selected to represent the extremes of behaviour, which are likely in the repository. For these calculations the system, rock, bentonite, copper, cast iron should be considered. Realistic properties or property ranges for the individual elements should be used and where appropriate creep strength rather than yield strength should be employed. Where properties are uncertain a range should be explored and the sensitivity to variation in the range should be examined.

Copper

We agree that if copper can be produced in a satisfactory condition and that if it is not susceptible to localised corrosion or microbial attack in the repository environment then it would be an ideal overpack material.

We note the grade of copper proposed and acknowledge that it is the best available grade for the function. We note that a grain size of < 350 µm is specified. The reason given is related to resolution in ultrasonic testing. No inspection standards are given and it is therefore not possible to judge whether or not satisfactory inspection performance can be achieved. Comparison with the performance of radiography, which is also undefined, is of no value. There are many objections to coarse grains and 350 µm is coarse. They will cause inspection problems and they accentuate any problem arising from grain boundary segregation of impurities. A further objection is that yield stress is inversely proportional to grain size. Werme [2] (section 6) refers to calculations that predict plastic deformation in the lid during handling. Whilst the assertion that this is acceptable for a fine-grained material is dubious, for a coarse grained material it is more so. Since relatively fine grains have now been achieved in extruded tubulars we strongly recommend that the grain size specification is revised to a lower value.

We continue to be uncertain concerning the creep ductility of the specified material in the repository environment. The SKB work on the effects of phosphorus on creep ductility has been carefully re-examined [11, 12, 13]. The most recently published work, Henderson and Werme [11], compares the results of creep tests on 30 specimens, which include 10 different types. The number of tests for each specimen type is rather small. The results suggest that at constant grain size, for extruded material tested at 215°C and 100 MPa, creep rates are reduced by a factor of 100 by the addition of 50 ppm phosphorus. Also phosphorus bearing material, processed by hot rolling and having a grain size of 115 μm (compared with 45μm for the extruded material) tested under the same conditions, has a creep rate which was 10 times faster than the extruded phosphorus bearing material. All of the specimens used in the above tests had 6 ppm sulphur and, whilst the phosphorus free material had lower fracture strains than the phosphorus bearing material. None had the very low fracture strains (circa 1-3 %), which have been the cause of concern. This could be because the phosphorus free material was very fine-grained (45μm). All the remaining specimens had less than 3 ppm sulphur and are therefore not relevant to the argument concerning poor creep ductility caused by segregation of sulphur or to the practical case. There is still no evidence that additions of phosphorus improve the creep strain to fracture of coarse-grained material. We consider that a dedicated exercise should be carried out to resolve this argument.

A very limited number of tests on material taken from welds are reported in the work referred to above. Specimens of phosphorus free material were cut so that the entire gauge length was weld metal. The specimen long axes were either along the weld direction or perpendicular to the weld direction. Specimens of phosphorus bearing material were cut to include a transverse section of the weld.

Chemical analysis revealed that phosphorus was not lost during welding, this is a surprising and an important result. Surprising because it is reasonable to expect the phosphorus to be lost either by volatilisation or by zone refining. Important because if neither of these mechanisms occur for phosphorus they may also not occur for other elements. If this is the case then zone refining is not responsible for the difficulties experienced in welding and there should be no problems related to high levels of impurities in the last metal to be welded. In view of the importance of this result we feel that it should be checked carefully, not only for phosphorus but also for the other impurities in the copper.

There are too few results to reveal any difference in creep properties, which might arise as a result of the addition of phosphorus in these weld materials. In addition the effects of different specimen types could mask any effects of phosphorus. Two specimens of the phosphorus free welds failed at unacceptably low fracture strains. The reason for this is not clear, as they are also very low in sulphur. We feel that at this stage the results of creep tests are too few and too disparate to allow firm conclusions to be drawn. There is however sufficient information to demonstrate a need for a comprehensive creep testing programme designed to improve our understanding of the creep properties of the overpack material both including and remote from the welds.

There is no doubt that the improvement in creep resistance conferred by phosphorus is desirable but there is some doubt concerning the likelihood of creep of the copper due to uneven swelling of the bentonite, any creep programme should also provide data which is relevant to this case.

The mechanical integrity of the copper overpack is also dependant on its thickness. We note that a reduction in thickness from 50 mm to 30 mm is under consideration. We agree that it is necessary to bear in mind the effects of a reduction in thickness on fabrication processes and handling in particular.

We do not agree that Electron Beam Welding is a proven process for the fixing of lids, at this time, or that it will necessarily be easier to fit lids to thinner canisters in the range under consideration, (i.e. down to 30 mm). We do agree that none of the other welding methods mentioned is proven for this application. In view of the difficulties that have been experienced in the development of Electron Beam Welding, we agree that all possible alternatives should be considered. The results from friction stir welding reported by Anderson [3] are very promising. We consider that this process has the right characteristics, for the canister sealing, in terms of its effect on microstructure.
If the copper overpack is reduced in thickness to 30 mm, it will be necessary to increase the thickness of the load bearing liner in order to provide adequate shielding. An important benefit from this could be an increase in the strength of the liner. A downside to using a thinner overpack is that it may not be capable of accommodating the handling loads.

Test Manufacturing of Copper Canisters with Cast Inserts

Introduction

Experience to date with test manufacturing has been described by Andersson [3]. In this section we will comment mainly on the statements in his report.

The copper overpack

Manufacture of tubulars

Three established manufacturing methods have been tried for the production of copper tubulars:

(a) roll forming,
(b) extrusion and
(c) pierce and draw.

In addition three novel methods;

(d) hot isostatic pressing (HIP),
(e) spray forming (Osprey) and
(f) elecrodeposition.

have been examined.

Whilst roll forming, extrusion and pierce and draw processing are established production methods, none has been developed for the production of tubes of the sizes of interest in copper. It is true that there are several manufacturers who have facilities for these processes on the scale required if they are considered as a collection. However there is only one who can do extrusion and one who can do pierce and draw. Both processes require very expensive plant and in both cases any production plant would be capable of producing the entire SKB annual requirement in a few weeks per year once the development programme had been conducted. For this reason it is unlikely that another producer will enter the market unless a substantial increase in demand arises from other sources. On the other hand roll forming is available from a number of sources. Non of the novel processes mentioned are available to produce full size tubulars today.

In the following paragraphs each of the ”established” methods will be discussed in turn. For the time being the novel processes are not included in the future programme and we agree that this is appropriate.

Roll Forming

Roll Forming is a craft process depending on skill of the operators [3] (Page 13), the material used is copper plate of thickness 60-70 mm, length 5m and width 2m and this can only be prepared by a very small number of rolling mills in the world.

The ideal rolling process to produce a desirable microstructure of fine uniform grains has so far not been achieved. Usually the grain size is mixed and coarse. There are sound metallurgical reasons why this is so and it is unlikely that this route will ever achieve an ideal microstructure.

Results achieved by roll forming of copper plates are discussed on Page 27 in ref. [6]. Considerable variation in grain size in the plates is reported and it is claimed that the final four tubes were made from material meeting the grain size specification (<350µm).
No justification for this very coarse grain size has so far been given. Such large grains are usually undesirable. The standard ASTM method for measuring grain size is used by SKB contractors. It makes use of a comparator chart to give a grain size number that is subsequently converted through a look up chart to a grain diameter. This method does not give a true estimate of grain size variability.

Plates of the required thickness (60-70 mm) can not be reliably produced with uniform grain sizes. It is almost always possible to find some areas of fine grains and some areas of coarse grains. For this reason any reasonable grain size specification should also specify the method of measurement and the method of selecting and preparing samples. Usually mechanical property values and corrosion test results (which are influenced by segregation effects) are quoted for material having a grain size of up to 100 µm.
The mechanical properties such as yield stress and creep stress for given lives are affected by grain size with coarser grains giving lower property values. Further, grain boundary segregation effects are more severe as grain size is increased.

As a consequence of the manual processing to form roll formed halves the plate material is left in a non-uniform state of internal stress. This, together with the expansion characteristics of the material during welding, requires that the two halves of the tube be constrained in a very rigid frame during welding to prevent unacceptable distortion. This would otherwise completely prevent continuation of welding.

The constraint does not remove the stresses arising from processing, it simply contains them as internal stresses in the material. These internal stresses can cause the defects known as hot tearing or cold tearing in the zone close to the weld. Both these have been observed during manufacturing trials. Both arise as a result grain boundary weakness, which is associated with coarse grains and segregation of impurities. Cold tearing has occurred after removing the constraining jig and hot tearing has occurred during attempts to repair imperfect welds.

If a tubular is left in a condition where internal stresses are not relieved then it will distort during and after final machining. If it is subjected to a stress relieving treatment there is a possibility that it will undergo critical strain grain growth to produce very coarse grains near the surface. The critical strain at which this grain growth phenomenon occurs is between 5% and 10% which band contains the surface strain developed during roll forming. The critical temperature for initiation of critical strain grain growth is not known for the canister material. The choice of stress relieving temperatures by SKB is low in recognition of the possible problem. Whilst a number of roll formed tubulars have been made to date none has been used to produce satisfactory canisters. SKB have recognised that whilst it may be possible to develop the process to produce satisfactory canisters, it is not a process which is sufficiently reliable for serial production of tubes at this wall thickness. We agree with this conclusion.

Extrusion

Extrusion is introduced and described in [3] (Page 18) but no details are given on the degree of deformation used in the forging (upsetting) process, which is carried out prior to extrusion. We consider that this is very important information and that a standard procedure should be developed, because the extrusion ratio is less than the ideal value of 16. Thus it is important that the pre-forging should be adequate to break up the ingot structure.

Recent extrusion trials have been remarkably successful. We consider that grain size results (approximately 100µm grain diameter) are typical of those expected when a process is metallurgically well controlled. The good results suggest that the forging used in the upsetting process has been very beneficial. The processing temperature of 675°C is not low and there may still be scope for further reduction, which would give better control. There seems little doubt that this approach should be the method of choice for the present, but it may also be possible to obtain similar results by pierce and draw.

Pierce and Draw Processing

The pierce and draw process has some very attractive possibilities. In principle the first operation, upset forging is designed to remove the undesirable cast structure that occurs in the ingot (in the same way referred to for extrusion above). In order to achieve this, as a rule of thumb, the ingot is reduced in height by a factor of three and then restored to its original dimensions, both operations by hot forging. The associated deformation breaks up very large unfavourably shaped cast crystals throughout the ingot and recrystallisation should occur to produce a uniform equiaxed grain structure. However the very large size of the ingot results in very slow cooling rates in the ingot through the critical temperature zone with a consequent risk of severe grain growth, particularly in the centre of the ingot. This grain growth may or may not be a serious problem depending on the subsequent processing. Ideally the piercing operation will cause further improvement in grain structure, recrystallised grains will be heavily worked leading to further recrystallisation and the thinner walls resulting from piercing provide the potential for more rapid cooling. Careful control of the heating and cooling cycles in the processing to develop progressively thinner sections allows the possibility to produce a desirable structure in the finished walls by repeated deformation and recrystallisation whilst suppressing grain growth. So far trials have not been carried out to determine whether or not suitable heating and cooling cycles can be achieved. In the trial which has been undertaken temperatures were kept high in the interest of preventing damage to the equipment. It is understood that the grain size in the tubular section was too large but not so large as the grain size in the base.

In the future we might expect improvements in both the structures in the tubular and the base. However the original forging deformation is less than ideal and the structure in the material is unlikely to be adequately refined by this process alone. This may be of little consequence for the tubular where there is further opportunity for serious deformation, but the base will probably see very little further deformation and most of that which it does see will be close to the end of the processing cycle. Consequently it is less likely that a desirable structure may be developed in the base than in the tubular. It is possible that the process will not provide a suitable vessel with an integral base unless more attention is paid to the forging in the first operation and good control over temperature can be achieved in the further processing.

We agree that stress relieving should not be required on vessels or tubulars made by this route and we consider it to be a good candidate process for production of tubulars.

Manufacture of lids and bases

It appears that a satisfactory method for producing tubulars may be emerging but the problem of making and fitting bottoms still exists. The same arguments concerning control of structure in the tubulars apply to lids and bottoms. In order to produce a desirable structure in the component it is necessary to perform sufficient hot work on the stock material and to control the cooling. The problem is similar to that faced by manufacturers of railway wheels but more severe because copper does not provide the benefit of a phase change during cooling as steel does. In the railway wheel case, twelve sided or circular ingots are used and the processing includes a number of forging operations to change the shape from a tall ingot to a flat disc with a thick rim.

As with all other components it will be necessary to demonstrate that the final process for production of lids and bottoms delivers a satisfactory structure throughout the component. This means setting standards for the grain size and shape and distribution through the thickness and over the entire area. In setting the standard it should be recognised that the lid will be heavily stressed (for copper) during handling and possibly during service.

It will therefore be necessary to demonstrate that the standards proposed are adequate and that they are met. The demonstration will require a quantitative metallographic survey of a number of components and measurement of the effects of structure on mechanical properties.

Joining of lids and bases to tubulars

SKB has been developing the Electron Beam Welding (EBW) technique with The Welding Institute (TWI) for many years [3] (Page 12). However, it is not yet developed to a usable state either for seam welding (when the roll forming process is used for tubulars) or lid welding.

The seam welding uses high vacuum technology, which is very well established, even for copper. However it is unusual to require welds of 5 m length in material of thickness 50 mm. It has been reported and it has been observed by SKB that the welding process, when it is applied to high purity copper works well initially but after a certain length of weld has been made, the electron beam becomes unstable. Flashover occurs in the electron gun and the welding process is interrupted. The interruption may be of very short duration but it may also recur after very short intervals. When flashovers occur repeatedly it is necessary to stop welding in the interest of safety. Interruption of the weld under these conditions leads to a defect, which on most occasions to date have been impossible to repair. It has been proposed but not demonstrated that the flashover occurs as a result of a build up of impurities in the weldpool. The cause is suggested to be zone refining but this is not yet demonstrated.

We consider that in addition to the empirical approach to process improvement it is necessary to carry out a detailed programme to determine why the process fails after long lengths of good weld have been produced.

For welding of lids and bases there are two further problems. The first is that the welds are ”blind” (i.e. they are only open to the welder side). They are therefore susceptible to weld root defect (shrinkage porosity in the root of the weld) which may be severe. The second is that the width of a normal electron beam weld is so narrow that it is difficult to follow the intended weld line. This is exacerbated by the fact that the weld is slightly curved in the depth direction (the reason for which is not understood). This curvature of the weld is more serious in effect for deep welds where it takes the weld off the intended weld line at a distance below the surface.

The reduced pressure welding process has been devised to overcome these problems. It uses very much higher power than the high vacuum welding process. It produces broad weld pool in the shape of a teardrop with the narrow end at the surface. It has been under development for many years in the joint SKB TWI programme. So far no fully satisfactory welds have been made. Recently new improved equipment has been built to overcome the remaining problems.

The new equipment has been installed at Oskarshamn. A substantial development programme and learning phase may still be required and it may or may not resolve the problems that remain. We consider that a detailed programme for understanding and solving the remaining problems should now be undertaken.

For the first time we are advised [3] (Page 34) that high vacuum electron beam welding has been tried on bottoms for two canisters. The result is surprising. Flashovers were observed and in one case this led to the need for a repair. In the past repair attempts have failed both with high vacuum welds and reduced pressure welds. Ultrasonic tests carried out on these welds are reported. No weld root defects have been detected except where the weld is running out at the end. It is known that the ultrasonic inspection method used at TWI is limited in its effectiveness on these materials but weld root defect can be very large and it should be detected if it is present. No comment is made on the effects of using a narrow weld. It seems that the state of knowledge has not yet progressed to the point where a firm development programme to solve problems one by one may be defined.

Andersson [3] (Page 52) describes a possible alternative welding process, friction stir welding. The results of developments at TWI, which are given, are very encouraging. If it can be fully developed for the purpose of welding lids and bottoms it will provide a substantial improvement over EBW in practicality, in quality of the structure developed in the final weld and in soundness of the finished weld. It is fundamentally a much more favourable process and we consider that further development should be prominent in the future programme.

Cast ingots for copper canisters

Very large copper ingots are required in connection with production of tubulars for extrusion and for the pierce and draw process [3] (Page 18). They are being continuously cast at 1 m diameter. It will be necessary to identify and understand the defects, which occur in the ingots and demonstrate that any such defects, which would affect quality in the finished component, are identified at an early stage and rectified or rejected. The obvious defects are unsoundness, surface cracks, centre line (star) cracks, segregation and cold shuts (surface defects arising from interruption to the supply of molten metal during casting). In the material of choice unsoundness and star cracks may occur, but they should not cause a problem if they are not surface breaking. This is because they should be healed satisfactorily during forging. Similarly it seems unlikely that segregation in the ingot should be a problem in such a pure material which is to be subject to homogenisation during further processing (notably in the forging steps). Surface cracks and cold shuts are more likely to cause difficulties as associated oxides will be included as defects in the finished products. It is necessary to have an inspection standard for ingots to ensure that casting defects do not lead to unacceptable defects in finished components.

Ingots for seamless tube manufacture by pierce and draw are not surface dressed. Ingots for extruded tube are dressed to remove 2 cm from the diameter. This dressing is to remove casting defects. If pierce and draw is adopted for the canister it is necessary to know what happens to the material containing the defects.

Inspection/non-destructive testing

Acceptance criteria for NDT of the copper canister are not available [3] (Page 28). At this time the effectiveness of the test methods is also unknown. In order to be safe it is normal to develop designs that assume that the largest defects that would go undetected are present. This has not been done in this case. At some point it will be necessary to define either the type of defect which may occur and the severity which can be accepted in the light of sound technical criteria, or the severity of defect which may be reliably detected.

For the first case it will be necessary to develop adequate test procedures and for the second it will be necessary to re-examine the designs to ensure that they are tolerant to defects which may be present. Dye penetrant and X-ray inspections are mentioned but at present there is no information on the effectiveness of either. If these are the only methods available then some systematic work is required to qualify them for the task. Ultrasonic testing is also being planned, but there is no evidence at present of a technically based inspection standard or of the effectiveness of ultrasonic inspection in the material of interest. The forward research programme should include detailed work to address all these points.

There is a suggestion that thinner copper overpacks are under consideration, and this would simplify the process. Grain size problems would be less severe, welding would be easier and production of ingots and plates would be simplified. All these problems would not completely disappear but they would be less difficult to control.

The cast liner

Attempts to produce cast steel inserts are referred to [3] (Page 37). Whilst the mechanical test results on cast steel insert material may be satisfactory the quality of the castings is not. The reasons for the poor casting results are well understood and they are very unlikely to be overcome. The change to nodular cast iron was entirely appropriate and the results with this material are very promising.

The nodular iron castings are top poured from a tundish through a feeder system matched to the shape of the top of the casting. Uphill rather than direct casting was tried first. The reason for using uphill casting first is that it is known to provide more uniform filling of the mould and reduce the likelihood of defects such as cold shuts, slag inclusions and shrinkage cavities through inadequate feeding. It is clear that uphill casting met with limited success and somewhat surprising that direct casting should have been so much more effective. The future programme will need to define casting conditions closely and to demonstrate that the defects referred to above, which are common in top poured products may be reliably avoided.

The mechanical test results for cast on bars made with the nodular iron castings simply indicate the material was in specification. They do not indicate that the casting is satisfactory or in specification on soundness, structure or properties. These depend on casting practice as well as composition. It is noted that the test bar from casting I13 is

out of specification on ductility but no reason is given. The composition appears to be satisfactory and the casting procedure has not been questioned. Either the cast on test bar is unsound or it has developed a structure typical of more rapid cooling.
The cooling curves given for one casting are typical of what may be expected for the material. It should be recognised that these are cooling rates on the surface and may not be representative of cooling rates in the interior. The microstructure demonstrated is from a test bar, and it is a good example of what may be achieved with the particular material under ideal conditions. So far there is no evidence that it was achieved in the castings.

Test results from bars cut from casting I7 show variations of up to 10% in yield strength and ultimate strength compared with the cast on test bar from the same cast. They also show a decrease in ductility of more than 50%. This is an indication that the structure achieved in the part of the casting from which test bars were taken is different to the structure of the cast on test bar. The point must be made that variations in structure and properties of the cast material will vary according to position in the casting and to the casting procedure. It will be necessary to carry out a sufficient number of tests to understand the variations in properties that occur through the casting and if necessary how these variations may be controlled. The final stressing calculations for the canister must be based on mechanical properties that may be reliably achieved in the finally specified casting practice**,** not on the properties determined from cast on test bars.
No ultimate compressive strength values are given for the tests on any material but compressive yield strengths are given for bars from casting I7. It is likely that the compressive strength is quite close to the compressive yield strength and that the material is brittle in compression (bearing in mind the low tensile ductility and the different mode of deformation, which occurs in compression).

It is important to know the compressive as well as the tensile failure stresses for the purpose of stressing calculations and the given numbers show that the compressive strength may be only 66% of the tensile strength.

Cast inserts have been tested with die penetrants and ultrasound. It is necessary to know much more about the testing before a statement that the material is homogeneous can be made. It is very doubtful that ultrasound can be used to examine anything but the outer surface of the casting. It certainly can not give information on the material between the steel profiles used as cores. Tests will be required which demonstrate the effectiveness of the ultrasonic method on these castings before claims that the material is homogeneous can be substantiated. This will require both non-destructive and destructive testing on the same castings and positive demonstrations that when defects are present they can be detected.

During development the castings have been modified to include cooling channels in the heavy sections [3] (Page 44). These channels are filled with sand. Their presence reduces the section of the material that is to be cooled and therefore the total amount of heat that needs to be removed. They will reduce the tendency for shrinkage cavities to occur in the material in these areas and they will influence the microstructure of the nodular iron. The future programme should include surveys of the structure of the iron and of porosity throughout the sections of the casting. The influence of the variations that are finally accepted on the mechanical properties of the canister should also be recognised.

Corrosion and chemistry

Introduction

The largest emphasis is placed on the canister to isolate the waste in the final repository. Therefore also a heavy proof burden exists to demonstrate the ability of the canister to fulfil the isolation function at virtually all times up to 100 000 years.

Copper will be exposed for the risk of both general and different kinds of localised corrosion in the repository. The complex mechanical and chemical environment with high pressures varying in time and location and with oxygen, chloride, sulphur and carbon bearing compounds present will generate a risk of different types of attack that are going to prevail during different periods of time.

There will certainly be an introductory period with an oxidising environment followed by a reducing. There is, however, no guarantee that the oxidising conditions will not return at later stages, for example during glaceation events. At such events the pressure field will have a complex gradient, which could cause surprising directions and velocities of the ground water flow, resulting in intrusion of oxygenated water into the repository and subsequent replenishment of the oxidising conditions. During such periods, oxygen will provide the driving force for corrosion to proceed. Different mechanisms can prevail during different periods of time but also simultaneously in different geometrical parts of the non-homogeneous pressure, heat and chemistry fields that could form in the repository. Unfortunately a non-homogeneous environment is a good starting point for corrosion.

There will probably be long reducing periods between oxidising ones. During such periods corrosion processes are generally considered to come to a halt. We think, however, that several mechanisms for i.a. sulphide based attacks should be considered under reducing conditions.

Different periods of the repository lifetime are going to be exposed to different transportation mechanisms for the groundwater. As a consequence the reacting chemical species transported by the water will be transported with different velocities. This is valid both on a global and a local scale. A couple of mechanisms for such variable transportation mechanisms could be mentioned, but others are probably also possible. Normal ground water flow during normal conditions is a first. Changes of flow gradients due to different pressure gradients during glaceation periods is a second. Antropogenically caused changes form a third. Gas evolution due to processes in the repository as well as a flow of substances that at some stage might evolve geogas is a fourth possible mechanism for transportation. A fifth is the presence of temperature gradients.

In the following the subjects on this page will be discussed more in detail.

Status of the canister at deposition

All procedures of production, filling, transportation and deposition of the canister are crucial for its later integrity throughout the storage period in the repository. We have found that there are risks of metallurgical and mechanical faults in the canister due to any of those processes that can influence the corrosion integrity at a later stage. Also buckling of the copper onto the iron insert due to the pressure field of the closed repository and also changes of that field could cause subsequent corrosion related problems.

The QA system sketched by SKB doesn't seem to take care of all these effects, especially not in the later steps of the handling chain. One example is a handling fault in a canister induced during transportation to and deposition into the repository. Such a fault could cause later corrosion related problems and also be of a systematic, repetitive nature. This means that corrosion related damages at a later stage could appear more or less simultaneously in a large number of canisters. We feel that this is a serious scenario that should be further evaluated.

Chemical environment in the repository

As already mentioned there are several chemical environments around and also inside the canister that can be foreseen. Beside those there is always a residual probability of chemical environments that cannot be foreseen at present.

In all cases the environment is framed by the presence of the surrounding rock and of the bentonite that could be inhomogeniously composed, wetted, heated and mechanically loaded by an an-isotropic pressure field.

The first type of chemical environment is oxidising and containing high concentrations of chloride. It is going to dominate the scene a shorter or longer time after closure of the repository. The second is probably reducing, still containing chloride but also possibly sulphide by action of bacteria and/or presence of pyrite. Both types of chemical environment will also contain other chemical species like sulphate, carbonate, and matter with an organic origin and also antropogenic substances. Oxidising and reducing environments could replace each other during specific periods of time like glaceation.

Corrosion in an oxidising environment

During oxidising conditions in a highly concentrated chloride solution, which will prevail for example during the first time after closure of the repository, there will be general corrosion of copper in a situation of rather homogenous environmental conditions. Such a homogeneous attack is probably not disastrous for copper integrity, as transportation of reacting matter probably is slow. It is interesting to see that calculations demonstrate [14] that there is no protecting, passivating layer on copper in highly saline media over a range of potentials at 50-100 ^{o}C. If the diagram in figure 1, which is calculated for 25 ^{o}C, is compared with those for higher temperatures presented in [14] it can be seen that the immunity limit for copper descends as a function of temperature at high chloride concentrations. This means a risk that copper is neither noble, nor passive at least at higher temperatures in the repository environment containing large concentrations of chloride. We are concerned that copper in such an environment is protected from serious general corrosion only by keeping transportation rates of reacting species very low. What would happen if transportation rates were increased in an oxidising period for the environment? We think it is essential that this situation be addressed in the programme at least for cases of higher transportation rates of reacting species above the immunity limit.

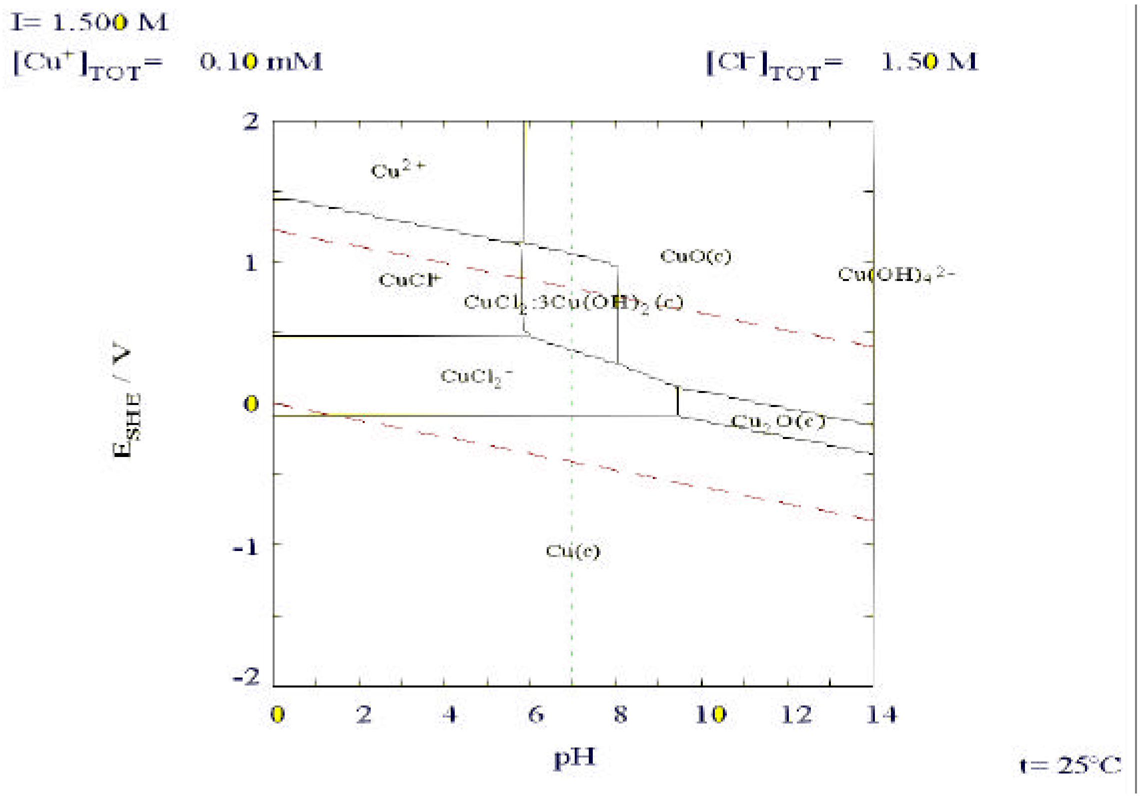

Figure 1 Pourbaix diagram for the system Cu-Cl-H-O at indicated conditions. The diagram is calculated using I Puigdomenec`s programs and database [15].

However, different types of localised corrosion that can occur as a result of any inhomogeneity in the system are probably more dangerous than the general corrosion described above. The inhomogeneity could be of a chemical nature (concentration gradients) or mechanical (pressure load gradient), as a temperature gradient (different heat conducting conditions from the canister surface) and also as materials faults and inhomogeneities in the copper metal itself caused by inadequate manufacture, transportation and handling processes.

All of those cases can be disastrous to canister integrity if they prove to be able to cause a rapid breakthrough of the copper layer. Pitting of copper in an oxidising environment is well known from several applications and environments and could also appear in the repository during the oxidising periods. Localised attacks could be a dangerous reality at the right combination of environment chemistry and material faults as cracks, pores, welding faults etc.

A localised attack could result in exposing the underlying iron insert. In an oxidising environment such an event could be followed by galvanic corrosion as well. The exposed iron insert would then be the dissolving, anodic electrode. A bad situation of a very large copper cathode, short-circuited with a very small iron anode would develop quickly and could generate an opening to the fuel. Only a retarded transportation of reacting species would hinder such a situation to develop dangerously. We recommend that this scenario be better evaluated.

A combination of a mechanical fault, material stress and local corrosion in an oxidising environment would be especially dangerous as it could be multiplied. As already mentioned there could be systematic and undetected mechanical damages on many canisters due to handling procedure faults and an incomplete QA system. We therefore think it is essential to further evaluate these combined risks and also to extend the QA system to cover all of the chain down to the final closure of the repository. It should also be considered to place indicators in the closed repository to detect early breaks as they could be of a multiple nature.

After some time a variant of the combined attack could show up. Severe buckling of the copper could occur, resulting in mechanical loads, local diminishing of the thickness as well as cracking of the copper layer. Localised corrosion would complete the disaster of such events especially in an oxidising environment during a glaceation period where shorter times to breakthrough would be required in those cases mechanical breakthrough were not already the case. Subsequent galvanic corrosion and destruction of the nodular iron insert exposing the fuel could then follow in a replenished oxidising chemical environment if transportation rates are high enough. This problem could be of a global nature and cause simultaneous break of many canisters.

Corrosion in a reducing environment

Copper sulphides are formed on the copper surface at lower potentials in environments containing sulphide and also high chloride concentrations, see figure 2. The sulphide film formed could be of a rather complex nature consisting of several phases. The local micro composition of the sulphide could under such circumstances depend very much on local micro conditions in the bentonite close to the metal surface. For example would variations in pyrite concentration probably have an influence on the local copper sulphide composition. A slight variation in the potential over the copper surface would also have a high influence on the composition of the copper sulphide film. Calculations as those accounted for in figures 1 and 2 also show that the immunity of copper decreases as the sulphide concentration increases. This results in a complicated situation with low immunity and a complex copper sulphide film of variable and disputable ability to passivate. We feel that such a system should not be left without further study, i.a. of the protective ability of the copper sulphide passive film within the span of possible environments. What would for example happen if there were a transfer from reducing conditions with a bad copper sulphide film into oxidising in combination with increased water flow. This could happen at the beginning of a glaceation period.

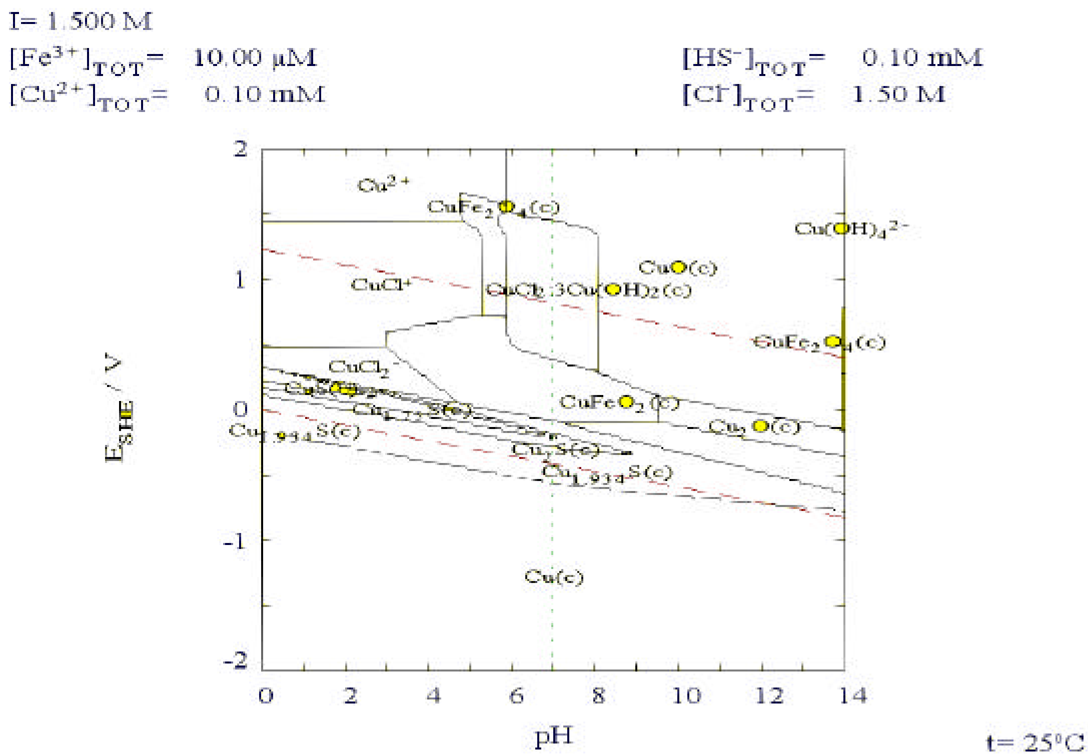

Figure 2 Pourbaix diagram for the system Cu-Fe-Cl-S-H-O at indicated conditions. The diagram was calculated using I Puigdomenec`s programs and database [15].

Another type of corrosion attack of a localised nature that appears on copper in a sulphide environment is whisker growth. The phenomenon has been discussed earlier [6] and recent experiments clearly demonstrate that it is possible to grow whiskers on copper. However growth of whiskers in a non-oxidising environment has been observed [16], further investigations are necessary to judge the importance of such a mechanism. For example it should be investigated if whisker growth really gives local attacks causing pits in the general sense. The high mobility of copper in sulphides would indicate that the metal could be taken from a larger surface area than locally below the growing whisker. We think that this remains to be demonstrated in order to dismiss whisker growth as a potentially dangerous corrosion mechanism.

It is obvious that sulphide and chloride strongly influences copper stability in several ways. To avoid surprises we recommend that the Cu-Fe-Cl-S-H-O system be investigated thoroughly both experimentally and theoretically. Experiments should for example be performed to prove the supposed innocence of whisker growth and to evaluate details of the transfer from reducing to oxidising conditions. This is important, as objects formed on the copper surface during reducing conditions could act as starting points for local corrosion during subsequent oxidising conditions. Theoretical calculations should be performed at least for the system Cu-Fe-Cl-S-H-O in order to describe as good as possible the thermodynamical framework for copper stability in the span of foreseen repository environments.

Assorted corrosion related topics

In some cases "known corrosion processes" are mentioned in [1] and the underlying reports. Which are those? They should be explained and discussed as quantitatively as possible with a firm evaluation of the risks.

In many cases of judging corrosion processes, "security or safety factors" are used in the discussion. Such factors should be quantified in each case and not stated as "sufficient" or in similar qualitative manners. A qualitative handling of security factors gives a feeling of high uncertainty to the aware reader and gives the ignorant reader a false impression of safety. The use of safety factors indicates a shortage of scientific facts.

The quality of bentonite in relation to the pyrite contents is not discussed in relation to later possible sulphide attacks. It has been pointed out earlier using figures 1 and 2 that both sulphide and chloride are going to lower the immunity level of copper and produce either a bad passivation film or none at all. Also bentonite quality is then a matter of concern for corrosion stability.

As general corrosion could be a problem at elevated transportation rates it is important to evaluate the possibility of seemingly odd transportation mechanisms, for example by the action of geogas.

There should be a system in operation for the monitoring of canisters during the first period of operation in order to selectively detect failing canisters for recovery. The hydrodynamic technique employing salt water/brine for recovery operations is, however, disputable from a couple of reasons. There is for example a risk for the future integrity of neighbour canisters as they could be exposed for an excess of aerated brine.

Conclusions

The conclusive part of this report contains 43 statements that are sorted under brief sub-headings.

Microbes, swelling pressure and safety factors

1. We consider that the evidence supporting the assertion that the SRB (sulphate reducing bacteria) will not survive in the repository environment is inconclusive, and the assertion is also inconsistent with the statement that the bentonite will be watersaturated. It also seems possible that the degree of compaction of bentonite necessary to suppress SRB would result in unacceptable bentonite swelling pressures. We consider that this area needs further examination.
2. We believe that the expression “no known process, which may occur in the repository environment,” should be capable of violating the canister should be used, rather than ”no known corrosion process”.
3. We also believe that the expected lifetime of the container should be clearly specified.
4. We believe that further work is required to determine the swelling pressure, which may be expected when bentonite blocks are made by practical methods and this value should be used for the design calculations.
5. We consider the safety factor on the strength of the canister for the glacial loaded case to be inadequate and that it will be further degraded when a better estimate of bentonite swelling pressure is available.

Insert and canister load

6. We agree that nodular iron is the most appropriate choice of material for the cast component, and we recognise the benefit of the improvement in strength of the insert conferred by the supporting internal structure.
7. It is our view that it will be necessary to carry out sufficient tests on material cut from cast inserts to give a confident determination of the range of material properties which occur in real castings and to redetermine the critical stresses using these figures.
8. We consider that the variations in microstructure within single castings and between castings, and the location, type and frequency of occurrence of casting defects should be determined. This information may be used in setting realistic safety factors.
9. It is our opinion that the results of stressing calculations given by Werme for various load cases should be treated as preliminary indications. Further calculations are required. Load cases should be selected to represent the extremes of behaviour, which are likely in the repository. For these calculations the system, rock, bentonite, copper, and cast iron should be considered. The entire volume of the canister should be explored and realistic properties or property ranges for the individual elements should be used and where appropriate, creep strength rather than yield strength

should be employed. Where properties are uncertain a range should be explored and the sensitivity to variation in the range should be examined.

Copper properties

10. We agree that if copper can be produced in a satisfactory condition and that if it is not susceptible to localised corrosion or microbial attack in the repository environment then it would be an ideal overpack material.
11. Werme (section 6) refers to plastic deformation in the lid during handling as safe, but we do not agree with this. Further if the lid material is coarse grained, the yield stress will be lower and the deformation more extensive.
12. Since relatively fine grains have now been achieved in extruded tubulars we strongly recommend that the grain size specification is revised to a lower value.
13. We agree that addition of 50 ppm phosphorus to OF copper improves substantially its creep resistance and that this is beneficial in the proposed application. We consider that the assertion that it also removes the tendency to low fracture strain, particularly in coarse-grained material, is not demonstrated and it remains a matter for concern.
14. There is still no evidence that additions of phosphorus improve the creep strain to fracture of coarse-grained material. We consider that a dedicated exercise should be carried out to resolve this argument.
15. We consider that the results of creep tests reported to date present a very confused picture of the creep properties of the phosphorus bearing material with and without welds. It is desirable that a systematic exercise should be conducted to improve our understanding of the creep behaviour of the OFP material under conditions likely to arise in the repository.
16. Chemical analysis revealed that phosphorus was not lost during welding, a surprising and important result. If this is confirmed, then zone refining is not responsible for the difficulties experienced in welding and there should be no problems related to high levels of impurities in the last metal to be welded. In view of the importance of this result we feel that it should be checked carefully, not only for phosphorus but also for the other impurities in the copper.

Canister manufacture

17. We agree that roll forming is not an industrial process suitable for serial production of copper tubulars.
18. We agree that extrusion has produced very promising results for the production of tubulars and that it should be a major component in the future development programme. We consider that attention should be paid to the effects of the pre-forging operations and the extrusion temperature on the properties and structures of the tubulars.
19. We consider that the pierce and draw process for manufacture of copper tubulars has potential for further development and should be included in the future programme. It

may be possible to produce a tubular with an integral bottom of suitable metallurgical structure by paying attention to the pre-forging operations and control of temperature during pre-forging and the pierce and draw operations.

20. We agree that HIP and electrodeposition are less promising than the other methods (18 &19 above) and that they should take a low priority in the future programme.
21. We consider that the Osprey process still has further potential to explore. If it can be used for titanium alloys it should work for copper. We believe that the possibility of using an inert gas atomiser in the process should be explored.
22. We believe that all process developments should be targeted at a mean grain size of 100µm in the copper components
23. We consider that if fine grain sizes can not be achieved, the method of selecting specimens and measuring grain size should be selected to ensure that the results quoted present a full and true picture of the structure.

Welding

24. We believe that a research programme is required to determine why electron beam welding fails by flashover after considerable lengths of good weld have been made. There should also be a detailed programme to qualify the process for use on the canister or reject and find something else.
25. We consider that friction stir welding is, in principal, a much better process than electron beam welding for the canister purpose and we believe that all efforts should be made to develop it for this purpose.
26. We consider that, as part of the quality assurance programme, work should be carried out to determine the nature, size and location of defects which may be tolerated in the copper overpack, the lid and base of the overpack and the welds which join them together. We further consider that a programme should be put in hand to define the effectiveness of available non-destructive test methods in locating and sizing all defects of interest. A similar programme is required for the insert.

Corrosion

27. The largest emphasis is placed on the canister to isolate the waste in the final repository. Therefore also a heavy proof burden exists to demonstrate the ability of the canister to fulfil the isolation function at virtually all times up to 100, 000 years.
28. Copper will be exposed to both general and different kinds of localised corrosion in the repository. The complex mechanical, chemical and microbial environment with high pressures varying in time and location and with oxygen, chloride, sulphur and carbon bearing compounds present will cause different types of attacks that are going to prevail during different time periods.
29. There will be an introductory period with oxidising conditions followed by a reducing period. There is no guarantee that the oxidising conditions will not return at a later stage, for example during glaceation.

30. At glaceation the pressure fields would be strongly inhomogeneous, which could cause surprising directions of ground water flow to occur, resulting in intrusion of oxygenated water into the repository. During such periods, oxygen will again provide the fundaments for the driving force of the corrosion processes.
31. There will probably be long, reducing periods between the oxidising. During such periods all corrosion processes are supposed to come to a halt. We think that possible mechanisms for sulphide based localised attacks should be considered during such periods.
32. Different periods of the repository lifetime are going to be exposed to different transportation mechanisms for groundwater and thus also for the reacting chemical species. This is valid both on a global and a local scale.
33. The procedures of production, handling and treatment of the canister throughout the processes of filling, transportation and deposition are crucial for its later, corrosion-related integrity throughout the storage period in the repository. There is a risk that due to systematically induced faults, many canisters may have later problems.
34. During oxidising conditions in a chloride solution there will be general corrosion of the copper. Such an attack would be serious if the transportation rates of reacting species were high enough.
35. More dangerous are different types of localised corrosion that can occur as a result of any inhomogeneity in the system.
36. All of those cases can be disastrous to the canister integrity as they could cause a rapid break through of the copper layer exposing the underlying iron insert. In an oxidising environment such an event would be followed by galvanic corrosion as well.
37. We think that the importance of an inhomogeneous environment should be better evaluated.
38. A combination of a mechanical fault, material stress and local corrosion in oxidising environment would be especially dangerous and could cause early problems in many canisters. Therefore it would be advisable to introduce an early warning system in the repository that could tell in a selective way about early failures of the canisters.
39. In some cases SKB talk about "known corrosion processes". Which are those? They should be discussed in a quantitative way.
40. In many cases also in connection to the judging of corrosion processes, security factors are used. Such factors should be quantified in each case and not stated as "sufficient" or in similar qualitative manners.
41. The hydro-dynamical technique employing salt water/brine for retrieval operations is disputable.
42. We are concerned about lacking guidelines for quality control and assurance covering the whole chain of handling events especially the steps of transportation and final disposal. We fear that this could i.a. cause global corrosion related problems later on in the repository.
43. We think that the corrosion part of the series of RD&D Programmes has diminished too rapidly.

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