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William H Bowyer
Hans-Peter Hermansson

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Comments on "SKB FUD-program 95" Focused on Canister Integrity and Corrosion

William H Bowyer [1]
Hans-Peter Hermansson [2]

[1] Meadow End Farm, Surrey, GU102DB, England
[2] Studsvik Material AB, S-611 82 Nyköping, Sweden

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List of contents

The numbered part of this list of contents follows the numbering of the main headings in "SKB-program 95" [1]. In this way a point by point comparison is possible. It should be noted, however, that chapters six and seven in [1] are here commented in an integrated manner under chapter seven.

Comments to:

Abstract

According to Swedish law 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 1995 and is called the "SKB FUD-program 95" (FUD: Research, Development & Demonstration). The work presented in this report was commissioned by SKI and is a result of reading the "SKB FUD-program 95" with focus on canister production, integrity and corrosion.

We find the programme very difficult to follow owing to the lack of detail in chapter seven. In our opinion this will make the work difficult to monitor by SKI or SKB. We also feel that the interpretation of information already available is overoptimistic. As a consequence the difficulties ahead are understated and the programme is converging 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 timescale 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 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 for the canister system that have to be demonstrated not to be harmful.

We feel there are many areas which need further evaluation, i.a. effects of non uniform loading and creep, effects of departure from circularity, welding, quality control, effects of radiolysis, corrosion properties, etc.

We also feel that insufficient emphasis has been placed on the further development on high power electron beam welding, machining, casting of the insert, testing and overall handling.

We consider that more information should be provided on the detail and timing of the development plan for the trial fabrication programme of the canister, test programme, determination of quality standards and development of non destructive testing procedures.

Background

According to Swedish law (SFS 1992:1536) the 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 1995. SKB is the utility 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 integrity and corrosion.

The "SKB FUD-program 92"

The previous "FUD-program" [2] and the comments issued on that are important also for FUD 95 [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 programme and the formulation of the new.

In the previous comments on canister integrity and corrosion [3] some important conclusions were made, i.a.:

- The R&D programme is an impressive document.

- The amount of information is, however, too large to comprehend by reviewers in a short time. This is still valid in FUD 95. 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 FUD 92 [2,3] to follow how decisions were made relating to programme formulation etc. This is also still valid in FUD 95.

- 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 is our feeling that R is declining too rapidly in FUD 95.

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

- In FUD 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 FUD 92. This statement is still valid and the writers can trace a tendency of neglect in FUD 95.

- The question of alternate mechanisms for transportation of radionuclides to the biosphere were not considered to be fully understood in FUD 92. This statement is still valid in FUD 95 and there is a tendency to take the problems too easy also in this case.

Summary

The SKB "FUD-program 95"

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, excisting 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 range safety and for the canister and encapsulation technology. These parts are followed by descriptions of the R&D programme for the canister and encapsulation, deep repository, safety analyses, supporting R&D, the programme for the Äspö-lab, for alternative methods and for decommissioning of nuclear installations.

Finally there is a plan for the implementation and a discussion of uncertainty and economy.

Scope and time schedule of the present review

This task of commenting the SKB "FUD-program 95" was commissioned by SKI. It was intended that it should focus mainly on sections six and seven which are concerned with the canister for disposal of high-level nuclear waste and consequently other chapters are only dealt with very briefly.

The work has been carried out by reading relevant parts of the SKB "FUD-program 95" [1] and a selection of supporting, basic reports. This means that the whole programme [1] has been penetrated but special emphasis has been put on chapters 6 : "State of knowledge - Canister and encapsulation", 7: "Programme for canister and encapsulation", 8: "State of knowledge - Deep repository", 9: "Programme for deep repository" and chapter 11:"Programme for supporting R&D". Section 6 refers to the present state of knowledge and to the premises on which the future programme is based. Section 7 details the forward programme.

On Nov. 27–30 1995 the consultants worked together at Studsvik in order to prepare a manuscript of the present report.

An advanced manuscript was submitted to SKI on Dec. 31, 1995.

1 Introduction

In the present report the writers have followed the main numbering of SKB "FUD-program 95" [1] down to the level of two digits. The contract from SKI was focused on chapters 6 and 7 and consequently the main effort is made there but minor notes will also be found on other chapters.

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 safe-guards. 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 and technical solutions and design made inside Sweden but supported by foreign knowledge. Review by the authority will be for guidance and the information flow will be open to public.

2 Programme goal

The goal of the "SKB FUD-program 95" is, by fulfilling all environmental and security demands, to begin the deposition of a smaller part of the used fuel in a deep rock repository in the year 2008.

The fulfilment requires an encapsulation plant and a deep repository as well as complementation of the transportation system. The storage is supposed to be carried out according to the KBS 3 concept [4] 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 rock.

The writers feel that the goal is clear but they have reservations regarding the time-scale which will be discussed in the sections below.

3 Step-wise development and construction

The step-wise development and construction means that work will be carried out in a sequence permitting minor changes of direction after a step. For the deep repository this means steps such as studies of location, detailed investigations, construction step 1, preliminary operation, evaluation, final construction and operation. For the encapsulation plant it means location and planning, construction step 1, preliminary operation, evaluation, final construction and operation.

The writers think this is a good approach in agreement with comments on previous FUD-programmes.

4 Deep repository – Principles and requirements

This chapter defines the knowledge that is needed for the handling, treatment, transportation and storage of the waste. Furthermore the location and construction of plants, safety analyses and impact on environment as well as principles for radiation protection and safety and requirements of the barrier functions are treated.

It is the requirements of the barrier functions and especially those of the canister that is the focus of the present review.

5 State of knowledge – Long term safety

There are many parts of the knowledge basis for long term safety which needs to be strengthened. Some of them are discussed in this chapter and others mainly under heading 7 below.

Corrosion

The first, very striking impression on a reader of FUD 95 especially interested in corrosion is the total absence of the word "corrosion" in the list of contents. One very basic parameter of long term safety is of course canister corrosion but this parameter has not been granted any heading of its own on any level in the list of contents of (1). This phenomenon probably reveals very much of SKB's view on canister corrosion, that the parameter is well understood and not very much more remains to be done.

The authors think differently. The long term integrity of the canister is the main parameter of the total long term safety of the repository. The integrity has metallurgical and constructional (chapter 7) as well as corrosion (chapter 5 and 7) implications and we think that the knowledge base as well as the programme in these areas are unsatisfactory.

To begin with, it should be emphasised that the scientific basis for copper corrosion is limited to the simple system of water-copper at low temperatures, which is i.a. described in [5] period. Therefore there is a large need to extend this knowledge to much more complicated systems, involving both thermodynamics and kinetics, to be convincing. In the repository there are many elements and compounds present that could have an influence on canister corrosion as would different types of transportation processes that could have a long range effect.

Furthermore, the presence of different types of metallic materials will contribute to the risk of galvanic corrosion. The presence of cast steel and cast brass together with pure copper and zircalloy does give the impression of a multitude of possibilities to develop galvanic corrosion. We cannot see in the programme that these matters will be the subject of a study.

The presence of large amounts of cast iron in contact with nobler metals such as copper and zircaloy will also increase the risk of production of larger amounts of corrosion products. These could subsequently develop stresses in the canister and cause a rupture as well as in a later stage influence the outer environment of the canister. As a result, the buffer stability could be destroyed as well as its ability to isolate.

It is stated (p37) that only oxygen and sulphide can corrode copper. This is too strong a statement as chloride in abundance (deep rock water with high salinity) as well as complexing agents (alien material left after closure) also are able to contribute to copper corrosion in combination with much weaker oxidation agents than e.g. oxygen.

Localised corrosion could be caused by copper sulphide growth in a virtually reducing environment during development of whiskers [3]. Such a mechanism has been contradicted in [6] on grounds of low copper mobility in low temperature copper sulphide phases. However, very recent determinations of such low temperature mobilities of copper in copper sulphide clearly show that the high mobility of copper persists at low temperatures [7]. Therefore the whisker growth mechanism as suggested in [3] cannot be cancelled on grounds of low copper mobility.

Recent work [5,8] clearly shows that the corrosive situation for copper in the repository environment is much more complicated than predicted in KBS-3 [4]. Already the simple chemical system copper-water in the temperature range 0-150 °C is more complicated than was previously supposed. Analytical results and tools for thermodynamic calculations have been further developed since earlier work. This process has resulted in revision of the view of the chemical thermodynamics of the system Cu-H_2O as described in [5]. This work will be extended to more complicated chemical systems which more closely resemble the repository environment. It is expected that the results of this work will demonstrate that the situation of the copper in the repository is much more complicated than has previously been considered.

In [8] the possible mechanisms for corrosion of the copper/iron canister in the repository environment are carefully explored. The main conclusions are that general corrosion is not going to constitute any problem, but different types of localised corrosion probably will. Examples are pitting, bimetallic corrosion, crevice corrosion, hydrogen embrittlement and microbially induced corrosion.

The production and transportation procedures could cause problems with the canister from the corrosion point of view. There are several events that would influence later corrosion behaviour. These will be treated in detail in chapter 7, but some examples of such influences are:

1. A important to take away that film by machining to avoid later problems with corrosion. It is also important to show that the machining process itself does not create a favourable starting point for later corrosion attacks.

2. The local degradation of a noble film by mechanical action in the chain of handling events.

3. The implantation of sulphide grains in the surface during the process of placing the canister in the disposition hole.

We have not seen any discussion of these problems indicated in the programme. They could be important, however, as points 1 and 2 could initiate pitting corrosion and point 3 could initiate localised sulphide corrosion.

Buffer

The present situation for the buffer as a mixture of bentonite and crushed rock deserves some comments.

First of all the permeability of the filler system depends on the relations between crushed rock and bentonite. There is going to be only 10-20 % of bentonite in the filler system. This puts a large emphasis on filler quality control as the risk for inhomogeneities and subsequent good conduction of water and gas will increase. Inhomogeneities and their consequences could also be caused by a too large size distribution of the grains in the crushed rock.

The handling of materials (bentonite and crushed rock, etc.) would supply a material that is saturated with air at the moment of usage. Beside the saturation with oxygen there would also be an oxidation of i.a. Fe(II) to Fe(III) and other materials that could be oxidised (sulphide to polysulfides, etc.) present at rock surfaces and in the bentonite. The latter oxidised material could later act as electron acceptors at sulphide corrosion in reducing (=free of oxygen, low potential) environment.

We have not seen any lengthy discussion about the influence of canister corrosion on the surrounding buffer. The influence could be mechanical if a large amount of corrosion products are formed locally. The influence could also be of a chemical nature as corrosion products could influence the chemical stability of the buffer materials.

It is also said that the buffer would act as a filter for colloids. Perhaps this is the case, but the zeta potential of the bentonite and silicates of the crushed rock would be heavily negative at the prevailing elevated pH values. Particles as well as many ions would also be negatively charged. Consequently there would be no filtration by electrostatic actions, only by mechanical. Perhaps this is sufficient to hinder colloidal transportation but the situation should be considered.

Concerning chemistry it is said that all processes of importance for release, transportation and retardation of radionuclides have been identified. It is obviously premature to say so. We are still not convinced about how the exact chemical environment in the repository is going to develop as a function of time. What would be the influence of temperature gradients and subsequent chemical gradients on the canister environment? Will there be geogas/melting methane ice mechanisms of transportation? The action of different types of corrosion is not fully evaluated either. Furthermore it is not finally decided which kind of canister would be used and the kind of properties it would possess and consequently the final corrosion properties are unknown today. The influence of corrosion processes and the corrosion products are not yet fully understood.

The actions of geogas and the possible formation of methane ice in the repository during glaciations were discussed in the comments to FUD-92 [3]. Those effects are neglected in the present programme FUD-95. This is serious as gas transportation could act as a short-cut of the geological barrier.

6 State of knowledge – Canister and encapsulation

Chapter 6 presents the experience and knowledge base on which the programme presented in chapter 7 depends. This knowledge base is drawn from literature and from experience in the earlier parts of the SKB programme. Whilst the achievements of the programme to date are substantial we find that they are seriously overstated with the consequence that the difficulties which will occur in the future programme are seriously understated. Indeed the lack of detail in the future programme leads the reader to the false impression that these difficulties do not exist. Since our detailed criticisms of the information presented in section 6 is only relevant where it impacts the programme presented in section 7, they are presented in section 7 in connection with the programme components which they influence.

7 Programme for canister and encapsulation

Introduction

We find that section 7 would benefit from more details. At present it is difficult to read without constant reference back to chapter 6. There is also an absence of information concerning the times at which particular intermediate goals might be reached. This will make the progress in the programme difficult to monitor, either by SKI or SKB. Several of the assertions (in section 6) on which the programme is based are unsound and these are detailed later in this section.

In view of the close dependence of chapter 7 on chapter 6 the two are discussed together in the following sections.

In order to help the reader the first subsections of section 7 (i.e. 7.1, 7.2 etc.) in this review follow the section numbers in the SKB FUD-program 95 [1].

7.1–7.3 Summary of sections

In the first 3 sections of chapter 7 there are descriptions of prerequisites and goals, a discussion of alternate locations and programmes and design of the encapsulation plant as it is linked to the canister design work.

There is also a general time schedule describing the relations in time of the different main parts of the work from now on up to the start of operation of the first part of the repository in the year 2008.

A general impression coming up when reading the details of work already performed and the programme to come, is that this time schedule is very tight. We have a general doubt that the time schedule will be met.

7.4 Development and design of canister

7.4.1 Introduction

This section lists and discusses the detailed criteria which must be established before final sizing and design of the canister can be undertaken.

They include considerations of the following main areas.

Long term safety and performance in the deep repository.

This chapter is divided into:

- Initial Integrity
- Strength
- Corrosion resistance
- Heat transfer
- Radiation dose
- Criticality
- Chemical impact and
- Mechanical impact

Reliability of fabrication and handling.

This chapter is divided into:

- Fabrication and inspection of unfilled canisters
- Transport to the encapsulation plant and arrival inspection
- Handling of canisters in the encapsulation plant
- Sealing and post weld machining

Sizing and design of the canister.

This section starts by pointing out that it is not until the final design criteria are established that the process of final design can begin. It is divided into:

- Material selection and tests
- Sizing
- Criticality calculations
- Detailed design of the lid
- Chemical environment of the canister
- Alternative designs

Each of these subjects are commented individually in the following.

7.4.2 Long term safety and performance in the deep repository

7.4.2.1 Initial Integrity

The statement "the probability of undetected defects that could lead to canister failure will be analysed on the basis of selected methods" is taken to mean that selected probabilistic methods will be used to decide the probability of the occurrence of defects which could lead to canister failure. It is not clear what kind of defects are to be considered or which probabilistic methods will be employed. It will be necessary for SKB to provide details on both these points. SKI will need to be satisfied both that the list of defect types is comprehensive and that the method of analysis is valid.

The requirement that the canisters must be "fabricated, sealed and inspected with methods that guarantee that less than 0.1% of the finished canisters will contain undetected defects which could entail initial leakage or that could lead to early failure of the canister", is very challenging. Non of the process so far proposed for manufacture, sealing or inspection have been shown to even approach this standard of performance. It is very useful, however, to have this criterion against which to measure progress towards the desired performance. It is assumed that canister failure is defined by any event which may cause leakage to occur.

7.4.2.2 Corrosion resistance

It is stated in [1] that the corrosion properties of copper are well known. In our opinion this statement is too strong and corrosion constitutes a dangerous threat to canister integrity.

Corrosion properties of copper are well known for normal conditions. In the repository copper is going to interact with an environment containing a multitude of elements and compounds. Oxygen will be present for some introductory time. The consequences of it being introduced again at later stages i.a. because effects of glaciation are not considered in sufficient depth. Localised corrosion could occur, perhaps followed by local sulphide corrosion when oxygen is consumed. Sulphur in combination with electron acceptors such as Fe(III), polysulfides and others can cause attacks to continue after oxygen has disappeared.

The presence of oxidants as related to effects of radiolysis is an important matter. There is an implication in section 6.2.3 that the required thickness of the overpack has a role in limiting the level of radiolysis outside the container in order to prevent acceleration of the corrosion rate of the copper. This is important because review of the whole subject of corrosion could lead to a relaxation (or an increase) of the thickness requirement for the overpack.

There is a discussion of radiolysis outside the canister in [9], valid for the original canister with a 200 mm copper overpack. The results imply that radiolysis in this case would not contribute to outside corrosion of this (200mm) copper canister. There are some calculations made also for thinner walls.

With radiolysis of water there is a complicated set of reactions taking place, which for low LET irradiation could be summarised by the simplified, unbalanced formula:

$H_2O + \gamma \rightarrow H_2, H_2O_2, OH, H, H^+, OH^-, e^-_{aq}$

It should be emphasised that H_2O_2 is a marker for all long lived oxidants that could appear in different relative concentrations as a result of the radiolysis. The oxidative power of the process is a result of the larger mobility of hydrogen, which will migrate away from the neighbourhood of the canister and leave the oxidants there, able to participate in corrosion processes. It is self explanatory that it is important to show that the radiolytic production of oxidants is still low enough for the new types of canisters suggested in FUD-95 (1), otherwise the environment of the canister could be corrosive on an oxidative basis.

In order to evaluate the effects of radiolysis, the shielding capacity of the present canister should be compared to the capacity of the 200 mm copper canister discussed in [9]. The present canister contains 50 mm copper and 100 mm iron or 100 mm brass. New calculations have to be performed before any conclusions can be drawn about possible influence on oxidant concentration.

7.4.2.3 Strength

In addition to the general statements made in section 6.2.3 it is also necessary to consider the effects of non uniform loading and of eccentricity of the canister on plastic collapse. The latter will need to be done in the light of manufacturing tolerances which can be achieved in the finally selected process. The work of Savas [10] considered a steel canister of diameter of 785 mm, a wall thickness of 50 mm and a yield stress of 329 MPa. The tolerance on roundness was ±3.5 mm on the diameter. It was demonstrated that a uniform pressure of 45 MPa would cause plastic collapse of the canister which was on the margin of the roundness specification. This load level should be compared with the 20-30 MPa which it is suggested should arise in a future ice age. The roundness tolerance seems very large compared with practical limits but the assumption of uniform stress may be questionable, it is important that check calculations are included in the design process for the practical case.

Non uniform loading could lead to continuing creep in the copper overpack and, if it is a real possibility, account will need to be taken of this in the choice of material and of the condition of the material. If the presently proposed candidate is adopted it will be necessary to demonstrate that it has adequate creep strength and creep ductility when it is in the states that will arise as a result of the manufacturing procedures.

The pressure gradient around the repository could be substantially disturbed as the ice front passes the repository area. The gradient could cause altered directions of ground water flow with consequences for buffer and canister integrity. The altered flow could e.g. cause the introduction of oxygenated water and water with high salinity into the repository with subsequent corrosion effects.

7.4.2.4 Heat transfer

The heat transfer information should be used to demonstrate the worst case time temperature profile in the material of the copper overpack.This information should be used in conjunction with the worst case loading situation and the worst case structural condition to demonstrate that unsatisfactory structural degradation of the copper does not occur (this includes failure by excessive steady state creep, failure as a result of reduced creep strain to fracture and failure as a result of corrosion associated with alloy concentration in grain boundaries).

There would be at least 2 effects of elevated temperature on corrosion. Firstly an increase in temperature normally increases reaction and migration rates. Secondly, elevated temperature might cause the creation of a separated environment in the neighbourhood of the container by the action of heat induced mass transfer of some components but less or none of others. Such a separated environment would be more dangerous from the corrosion point of view.

7.4.2.5 Radiation dose

As already commented above, radiolysis outside the canister will produce oxidants that would cause corrosion.

Inside the canister this effect is much stronger if enough water is present especially in combination with nitrogen. Therefore it is very important to exclude water from the inside.

In both cases the problem would in some way be proportional to the dose.

7.4.2.6 Criticality

The Oklo phenomenon clearly shows that there is a certain probability to reach criticality if fissile material is available by the result of other mechanisms in a situation where transportation processes are restricted, as is supposed to be the case in bentonite. The probability should, however, be very low for criticality to develop in the repository environment.

7.4.2.7 Chemical impact

Sensitivity in relation to corrosion is commented on elsewhere.

7.4.2.8 Mechanical impact

Mechanical interactions between canister and bentonite mostly concerns the bentonite interaction on the canister surface. The passive film on the canister can be mechanically damaged by the bentonite either during the process of deposition or at a later state if there are movements in the supporting bentonite. As bentonite contains pyrite, sulphide could also be incorporated in the surface film on the copper as a result of the mechanical interaction. In both cases the interaction could result in a later initiation of corrosion, if other factors are "favourable" for a corrosion process to start.

The mechanical action by the canister on the bentonite is not further commented at this stage.

The effect of mechanical impact on the canister prior to deposition will need to be considered. Experience at Wyman Gordon and VSEL has already shown that the copper overpack is very susceptible to mechanical damage by impact events. The three obvious effects of this are, effective reduction in the thickness of the available corrosion barrier layer, rupture of any surface film which may be present and creation of localised areas of high surface strain. All three could reduce the corrosion life of the overpack. It will therefore be necessary to establish the degree of surface damage which may be tolerated prior to deposition.

7.4.3 Reliability of fabrication and handling

7.4.3.1 Fabrication and inspection of unfilled canisters

Section 7.1 recognises that it is necessary to determine the acceptable limitations on:

- Microstructure of the material
- Porosity and surface finish
- Strength properties
- Inspection of fabrication welds for both the load bearing liner and for the overpack

These characteristics are linked to each other, to the manufacturing and testing procedures and to the requirements of the application. Inevitably in such a situation it is difficult to totally specify one parameter until the others are known. Since no one of them is tied down it is difficult to determine the starting point for any one of the series. It may be that when the manufacturing possibilities for the load bearing inner canister are clearly defined it will be possible to settle on the size of the canister and from there build up the quality standards for the vessel as a whole. Acceptable standards on porosity in the overpack will be based more on corrosion considerations than on strength but it will be necessary to consider their effects on creep characteristics (principally failure strain) in the uniform and non uniform loading cases. The possibility of non uniform loading would change previous conclusions that pores and structural defects deep in the material of the overpack are of little significance. Clearly large scale creep effects are possible in the non-uniform loading case and such effects would be influenced by deep defects. It is important therefore to determine the extent of any non uniform loading which might appear and to take account of it when quality standards are established. The required microstructural and strength properties for both vessels will depend on the precise alloys selected. Structural condition, including grain size will need to be specified as will degree of any segregation of impurities which may be present to grain boundaries. The impurity contents which are specified will need to take account of the grain size in the finished product. Surface finish will be important from the point of view of corrosion as well as non-destructive testing. Non destructive testing will need to be capable of detecting cracks, either surface breaking or buried, as well as pores in the material.

SKI will need to be satisfied that satisfactory criteria are established and that the test methods for checking against these criteria are available.

No proposals have yet appeared concerning the design of the canister for the necessary lifts. This is not a straightforward matter when the outer vessel is of almost pure copper. The attachments will have to be sufficiently robust to carry the lifting forces without undergoing significant plastic deformation and their design must be such that they do not introduce crevices and other surface alterations which could form sites for accelerated localised corrosion. It may be necessary to carry out corrosion tests on the proposed designs as part of the selection process, and these may have to be done after simulated or real loading cycles have been imposed.

7.4.3.2 Transport to the encapsulation plant and arrival inspection

It is very likely that special handling equipment will need to be designed so that the canister may be handled and transported without damage. This may be costly but there are no technological barriers to be overcome. Handling in the plant during and after filling but before welding will require the development of dedicated equipment. If the problems of inspecting the canister after manufacture and after welding the lids are overcome, inspection after transport should present no difficulties.

The full chain of production, transportation and handling as well as deposition in the repository will influence the mechanical as well as corrosion properties of the canister.

Each of the suggested methods of production will produce a canister material with different mechanical and corrosive properties . All the factors of microstructure, porosity, surface conditions and the status of the weld are of outmost importance for mechanical as well as corrosion integrity.

The mechanical state of the canister is important for initiating subsequent corrosion. Localised corrosion could be initiated e.g. by the presence of a noble surface film formed in the production sequence and that has been locally destroyed by bad handling in later stages. The presence of small sulphide grains implanted in the surface by contact with pyrite of the bentonite could also act as initiators of local sulphide corrosion in later stages. If such corrosion communicated with pores or crevices in the copper, (for instance with pores associated with the end of the electron beam welds) a minor corrosion attack would be transformed into an attack which could rapidly penetrate the copper overpack.

Handling procedures can thus in all stages influence the later integrity of the canister.

7.4.3.3 Handling of canister in the encapsulation plant

See comments under 7.4.3.2.

7.4.3.4 Sealing and post weld machining

There are outstanding problems related to sealing and post weld machining of both the inner and the outer canisters.

It will be necessary to seal the inner canister in an inert gas environment and to ensure that the seal does not allow significant outgassing of the container during welding of the copper lid. If a mechanical seal is used outgassing is likely and this could interfere with the proposed electron beam welding. If welding is used a process must be devised which is compatible with the material of the inner vessel and does not require preheating or post weld heat treatment and which enables inspection of the weld on completion. These considerations will no doubt be included in the selection process for the inner canister and SKI will need to be aware of the strategies employed and of their impact on the security of the container.

Sealing of the copper lid probably presents greater problems than sealing the inner lid. There is as yet no proven electron beam welding process for serial production of canister lid welds. Some promising results have been achieved by TWI at their Cambridge Laboratories, but a considerable amount of further development is required and success is by no means guaranteed.

When the process is fully developed it will be necessary to produce a smooth machined surface over the weld in the interest of good corrosion properties and for inspection purposes. No attempts have yet been made to address this point. Experience at VSEL (Vickers Shipbuilding and Engineering, Ltd) on early trials has shown that machining of the copper vessel is difficult. It is not possible to take light cuts. An adequate machining allowance must therefore be provided for heavy cuts to be used in cleaning up the surface of the weld. This has implications for the design of the vessel and it will require that the depth of the weld should be increased.

The machining operation after lid welding may require the development of a special lathe. Machining of the canister with its axis horizontal would be very difficult and could involve an undue amount of handling after filling. Under these circumstances machining with the axis vertical would be more desirable. A carrousel type lathe could be used, it would be important to support the job close to the tool to avoid bending loads and this would have to be recognised in the design of the lathe. In view of the significant heating which arises during machining it will also be necessary to provide cooling in the hot cell.

The surface finish of the copper canister has implications on the later corrosion resistance. All ways of production can create a thin surface film. At the extrusion process a lubricant would be used which probably is going to decompose. The lubricant itself or the decomposition products could react with or be incorporated in the surface of the copper. This altered surface layer could be even nobler from the electrochemical point of view than copper itself. In the repository environment the nobler surface would constitute a very large area compared with that of the "fresh" copper area produced by handling damage. This situation could be dangerous from the localised corrosion point of view. To avoid this situation developing it would be necessary to remove the altered surface area e.g. by machining or a proper chemical treatment (decontamination?). It should be pointed out, however, that the application of an unsuitable decontamination method would also imply later corrosion risks. SKI will need to be assured that this possibility has been properly dealt with.

7.4.4 Sizing and design of the canister

7.4.4.1 *Materials selection and test*

The programme is for further mechanical testing, including creep testing to the extent required to:

(1) verify the outcome of trial fabrication and
(2) qualify materials from alternative fabrication methods.

Such testing is required but first it is necessary to have trial fabrications which are representative of what might be achieved in serial production. The trial fabrications which have been produced to date do not include an inner vessel which has been manufactured by an acceptable route (that is because significant preheating was used before welding the lid), nor do they include an overpack which has a satisfactory grain size to enable proper inspection.

There is an indication in section 7.5 that further trial fabrications will be produced. The indications are that these will include cast inserts and overpacks made from tube extruded at 600 °C or tube fabricated from sheet.

Detailed analysis of the microstructures achieved in the proposed inner vessel and the overpack will be required and this will include variations in microstructure within castings, variations of microstructure within extruded tubes or plates used for fabrication of the overpack and variations in microstructure due to welding of both vessels. It will be necessary to estimate the effects of these variations on stability of the structure during service, on corrosion performance and on mechanical properties, including creep with special reference to creep strain to failure. The indications in section 7.4.2 is that SKB may intend to do all this but that it is too early to present the detail. The detail is important and it should be required in due course. It is very important that the mechanical as well as the other testing is carried out on material having a microstructure which is similar to that which will arise in serial production. When the results of all these tests are available, together with the information from criticality studies, SKB will be in a position to make the final design calculations.

7.4.4.2 Sizing

This activity depends on the results of all materials property and processing activities. When they are completed SKB will carry out the final sizing calculations. SKI will be aware of all the work leading up to this point and will need to see the final size calculations.

7.4.4.3 Criticality calculations

There is a certain probability to reach criticality if fissile material is available by the result of other mechanisms in a situation where transportation processes are restricted. SKI will need to see that this probability has been realistically assessed and reduced to an acceptable level by the design process.

7.4.4.4 Chemical environment within the canister

The chemical environment within the canister is determined by physical factors such as geometry, temperature, pressure and radiation field and by the chemically active components present. The latter are first of all whatever construction materials are used, e.g. steel/cast iron/cast bronze, zircaloy, inconel and fuel. The outside copper should also be included in this set as mechanically induced or corrosive processes could cause physical contact between copper and the inner system. Important chemical components of the inner system are also water, nitrogen and oxygen and the components of the fuel itself, i.a. caesium and iodine.

The scene is thus set and an immediate impression is that it is complicated with many potential actors. There is an awareness of this in the programme and it is stated as a necessity in [1] that the water contents should be very small and the air (oxygen and nitrogen) replaced by an inert gas (argon?). Such precautions are also completely necessary as the formation of nitric acid by radiolysis in combination with the presence of oxygen, all in a water-phase, would be disastrous on its own merits. If such a chemical system also is exposed to the action of bimetallic cell combinations (zircaloy/cast iron, inconel/cast iron, copper/cast iron) the corrosive effects could be very much increased.

We feel that it is necessary to study this situation more than has already been done. Studies carried out to date are founded on a discussion with very simple systems as the starting point. The theoretical (thermodynamical) background should be better evaluated, taking all components into consideration. It is also necessary to perform experiments to find out where the safety limits are for the complex system concerning radiation fields, concentrations of chemical components and bimetallic element combinations.

7.4.4.5 Chemical environment of the canister

The chemical environment outside of the canister is constituted by the buffer, containing bentonite, filler, water and initially also nitrogen and oxygen. From a chemical point of view this mixture is very complex, containing a lot of elements and compounds dissolved in the water or bonded into the mineral particles of the bentonite. The bentonite contains pyrite which is a source of sulphur which can preferably react as sulphide with the copper in the container. There is also chlorine present as chloride in the bentonite water. The chloride would act as a complexing agent and support copper dissolution at least at situations of high chloride concentrations. Also fulvic and humic acids present in the water could act as complexing agents.

Another source of chemicals would be material left by mistake in the repository. Such substances would i.a. be mineral oil and gasoline. Decomposition products of such materials could also act as complexing agents and contribute to copper dissolution.

The main chemical parameters as pH and E_h would behave in a way that pH probably stays very constant ($pH_{25} \approx 8$) for very long times because of the buffering action of bentonite. E_h would stay high for some time and then decline to rather low values as oxygen is consumed when oxidising i.a. Fe(II) to Fe(III) and sulphides to polysulphides. It should be pointed out, however, that the electron accepting capacity of the repository is not altered as a result of the consumption of oxygen. The electron acceptors are there to participate in future reactions driven by other forces but needing an electron sink to proceed. One example of this is the possible formation of copper sulphide.

Part of the chemical environment problem is the alterations in environment that can take place. Examples are the already mentioned change of E_h, change of water flow and chemical concentrations, formation of corrosion products and changes in temperature and pressure.

Some of the changes are mainly inherent in the system, for instance the E_h change and temperature change in the close vicinity of the canister. Changes can also be caused by other factors such as the actions of an ice-age, the formation of corrosion products and the appearance of gases (i.a. produced by melting methane ice).

In this context it is important that the canister be designed not to have a chemical influence on its own environment. For example there are no obvious studies of canister corrosion influences on the buffer. This type of influence could be of at least two kinds. The first is a mechanical influence by an abundance of corrosion products causing the buffer to crack and create at least local pathways of transportation. The other is chemical influence, which means that the corrosion products either react with the buffer material or are adsorbed in a way which changes the properties of the buffer in a negative way. Corrosion products could interfere with the bentonite by being absorbed, changing its chemical as well as mechanical properties. Such actions could result in higher transportation rates for water, gas and radionuclides through the buffer.

Interaction of temperature gradients will modify the chemical composition of the canister environment in the long run. It could be foreseen that there is a stronger radial temperature gradient combined with a weaker axial. The gradients could support transportation phenomena, e.g. supply oxidants and remove reaction products, and thus keep corrosion processes going on. It could also cause corrosion products to be transported and subsequently react with the buffer. The temperature gradient could also create and support concentration gradients that would subsequently create elctrochemical concentration cells.

It could thus be concluded that the chemical environment of the canister is a dynamic system which could alter with time in different ways, by itself or by outer forces. The resulting environment can support corrosion processes on the copper. The details and time constants are still to be evaluated.

7.4.4.6 Alternative designs

This section suggests that the writers of SKB "FUD-program 95" are more concerned about meeting the property requirements of the inner canister than of the overpack. If there are no remaining problems with the choice of copper or copper alloy for the overpack but problems of manufacturing a satisfactory load bearing structure persists, the use of the 100 mm thick copper canister would seem to be the only route from the present range of options. However the problems of producing a copper vessel with twice the wall thickness would be much more than twice the problems of producing one with the present preferred thickness. Whilst there are certainly problems related to producing the cast inner container it seems unlikely that they would be less than those associated with the 100 mm copper option (unless of course one of the alternative options, HIP, Spray forming or Electrodeposition are developed). This does not seem like a realistic alternative at this stage and it should be a matter of concern that no alternative strategy is available, if SKB are uncertain about the prospect of developing a suitable load bearing structure by fabrication or casting in a ferrous alloy.

7.5 Development of production technology

7.5.1 Introduction

It is claimed in the second paragraph of section 7.5 in [1] that "Two methods of fabrication of copper cylinders have been tested and evaluated", and that "These trials show that copper cylinders of the planned dimensions can be fabricated with the necessary quality.". This has not yet been demonstrated in practice. To date there is no evidence that the required quality can be achieved or even assured by non destructive evaluation if it were achieved.

Referring to the copper canister section 6.4 asserts " In recent years, trial fabrication has been carried out with the first two methods, (tube extrusion and pressing/rolling) which are commercially available in full size". This is misleading. An extrusion press exists which will process a copper ingot to a full size tubular, but at the time of writing only one full size copper canister has been fabricated and that is known to be unsatisfactory on grounds of quality. The press in question could enter into serial production but at present there is no indication that a satisfactory structure can be achieved by this process. Similarly, of four full size plates prepared by rolling, two were out of specification on composition and grain size. Of the other two one was out of specification on grain size and the other had a mixed grain size which would make its acceptance doubtful. There is still no evidence that the available equipment is suitable to produce adequate material or that it could be modified to produce adequate material.

It has been demonstrated that the preliminary design dimensions can be achieved in a copper steel canister and it is now proposed that future work will be concentrated on development of the fabrication and inspection technology so that canisters with suitable and quality assured properties can be fabricated industrially. This is an appropriate strategy but the difficulties which need to be overcome should not be understated.

In section 7.5 the options available as fabrication routes are dealt with sequentially, for the sake of convenience, comments on each will be presented in the same sequence.

7.5.2 Cast inserts

The inserts which have been considered to date have been fabricated in steel and the difficulty related to welding of lids was referred to in section 7.4.3.4. There is also the problem of filling the free space inside a fabricated canister which has raised doubts concerning the risk of criticality. It is now proposed that cast rather than fabricated liners should be used and the cast liner has become the reference case. It is referred to extensively in section 6.2.4 of the programme document. It is said that this is a slight modification. It is not slight it is Major. The fabrication route uses technology which is established and though difficult it has been shown to be feasible. Unfortunately the criticality considerations may rule it out in the originally proposed form.

Section 6.3.2 asserts that sufficient information is in the literature for the material selection of a steel inner vessel. This assertion is not valid if a casting rather than a fabrication route is employed. This is because of the uncertainty surrounding the structure which would be developed in a casting. It will be necessary to provide the design calculations, the literature references and most probably the experimental evidence that the properties used in the calculations are realised in the castings. Experimental values would be required from all the most highly stressed areas of the casting from a sufficient number of castings to be statistically significant.

Section 6.3.2 refers to investigations of corrosion of the steel canister following penetration of the copper, the results of these investigations may not be valid if the liner is made by casting.

It is not acceptable to state that "calculations show that in the event of a defect in the copper shell, corrosion will take place over a large portion of the surface of the steel. The corrosion products formed will not create such stresses in the copper that the defect will be widened". If this is a potential problem it is necessary to reference the calculations and to demonstrate by experiment that the assertion is correct. It could also be argued that there is a large noble copper surface in contact with a very small iron surface at the bottom of the crack/pore in the copper. The corrosion of iron would proceed on the small surface supported by the action of the large noble copper surface. This could lead to rapid penetration of the inner vessel.

There is no evidence that a satisfactory container can be cast in steel. Informed opinion in the UK is that such a component cast in steel would be affected by severe shrinkage stresses causing unsoundness and probably shrinkage cracks and that control of microstructure would be extremely difficult. Cast iron is a better candidate since it has an effective coefficient of thermal expansion which is less than half that of steel coupled with a lower solidification temperature (shrinkage allowances on steel castings are typically 2%, whilst on nodular iron they are typically 0.7%). This leads to a reduced tendency to form shrinkage cracks and cavities on casting. The choice of a spheroidal graphite iron is appropriate from the point of view of toughness. It would, however, be necessary to check that a consistent spheroidal graphite structure would be achieved throughout a casting of such size. There is also considerable uncertainty regarding control of cooling stresses to acceptable levels in a casting of this size. If it proves possible to make the casting, SKI will need assurance that it is crack free, that the required structure has been achieved and that the internal stresses are not so high that the component will fail during processing or early in its service life.

Cast bronze is the third candidate for the load bearing component. There is no problem in casting bronze components of this size and considerable experience is available both in alloy selection and in casting technology. The coefficients of thermal expansion of the bronzes are higher than either the steels or irons (iron $12\text{x}10^{-6}\,{}^{\circ}\text{C}^{-1}$, bronzes $(16\rightarrow18)\text{x}10^{-6}\,{}^{\circ}\text{C}^{-1}$) but internal strains arising from cooling in a cast bronze component would be less than in the steel or iron case owing to the lower melting points of the bronzes. Many bronzes exhibit a ductility trough at 400 to 600 °C and it would be necessary to select the alloy carefully to avoid this becoming a problem during cooling after casting.

The problem of feeding the casting would be difficult for all three choices and it would be necessary to devise a procedure to ensure soundness and to develop a non destructive test procedure to check it. The choice of a copper alloy may be more favourable from a casting point of view but it should be recognised that the best cast copper alloys are likely to have a proof stress similar to that of a mild steel. This could lead to a need for the dimensions to be reconsidered if a copper alloy were selected. The dimensions of a cast inner component would in any case have to be reconsidered from the point of view of structural stability owing to their lower strength and stiffness properties.

It has been suggested that the inner component might be cast in two pieces which are subsequently welded together. If such a procedure is made to work it will be necessary to consider the effect of the welds on the properties of the structure with particular reference to the effects of the heat affected zones close to the welds. Tests will be required to demonstrate that adequate strength is retained and that the toughness of the structure is adequate.

The need for filling the space in the canister is clear but alternative ways to the integral cast inner canister could be explored, for instance the use of cast building blocks with shaped channels which could be inserted in the fabricated canister. Whilst a cast component could be the most economic solution in the absence of technical problems it is very likely that the technical problems of manufacture and quality assurance will lead to major cost penalties which would far outweigh the extra cost of casting and inserting separators. The problem of assuring the quality of the integral cast components with a reliability of 99.9% is very challenging and it would be necessary to demonstrate that adequate quality assurance procedures are available.

For the inner container it has been shown [10] that the effect of small departures from a truly round profile can have serious effects in terms of plastic collapse even when a high strength low alloy steel is used. The acceptable tolerance on roundness of alternative materials will also need to be checked before they can be accepted for use. The toughness of cast materials could be a problem particularly in the presence of internal stresses and if the target structure is not achieved. It is also necessary to consider the performance of the inner canister when the loading is unbalanced and tensile and shear strength properties become important. The use of cast material will reduce the tolerance of the container to unbalanced stresses or to departures from a truly circular section unless the dimensions are changed.

7.5.3 Rolled and welded or extruded copper tubes

Section 6.3.1 states that the choice of copper as the canister material is made on corrosion grounds and that the addition of 50 ppm phosphorus has the dual effect of limiting the development of coarse grains and preventing the development of low creep ductility. The argument for choosing pure copper is very strong in the absence of technological problems such as how to make it pure enough to avoid low creep ductility and how to make it with a sufficiently fine grain size to dilute the impurities in a large volume of material and render it inspectable by ultrasonic methods.

Unfortunately in section 6.3.1 there appears to be some misunderstanding of the roles of sulphur and phosphorus on creep ductility and grain size and the interpretation of very limited experimental work [11] is flawed. Pure oxygen free copper has not been shown to have poor creep ductility, as far as the writers is aware, it has not been tested. Poor creep ductility has been observed in nominally oxygen free copper having low concentrations of sulphur which segregates to grain boundaries [11]. This effect is well known and it is accentuated when the grain size is large owing to the correspondingly low grain boundary volume to accommodate the segregate. In the same work it was observed that material containing phosphorus additions with less sulphur and finer grains did not exhibit low creep ductility for the limited test regime explored. This may be expected as a result of the lower sulphur content and the finer grain size alone, however material of the same fine grain size and sulphur content which was free of phosphorus did have a very low creep strain to fracture. This evidence supports the suggestion that the presence of phosphorus reduces the effect of sulphur in these very fine grained materials ($45\mu m$) but it does not demonstrate that this low sulphur, phosphorus bearing material would not suffer from low creep ductility if it were coarse grained. The important point to consider here is that in a material of grain size $250\mu m$ the grain boundary area - per unit volume is five times less than it is in a material of $45\mu m$. The concentration of impurities in the grain boundaries is correspondingly five times higher. Phosphorus might have a beneficial effect in the coarse grained material but it can not be assumed.

It is said in section 6.3.1 that the addition of phosphorus increases recrystallisation temperature and this is well established. The increase in recrystallisation temperature is coupled with a claim that this controls grain size. This can be the case during annealing of cold worked material and it can be important to controlling grain growth during service. The material proposed for canisters is not cold worked and annealed however. The grain size in that material is formed during hot working at temperatures where the phosphorus would have no effect. Phosphorus does not control final grain size in a hot worked product. In such products the grain size is established during hot working and during cooling from the hot working temperature. This is well above the recrystallisation temperature. The experience of SKB in production of an extruded tubular and in the procurement of copper plate for fabrication, where grain sizes of several millimetres have been observed, clearly demonstrate this point.

The sulphur content of these materials is unknown but there is likely to be a sulphur content at which the low creep ductility is a problem and this critical level will be a function of grain size. The relevant grain size may be influenced by the presence of phosphorus.

In section 6.3.2 it is asserted that there is a need to break down the ingot structure in order to make the material inspectable by ultrasonic methods. This is true but the main reason for breaking down the cast structure is to develop the required mechanical properties, the NDE point is secondary.

SKI will need to be assured that the problems of coarse and mixed grain sizes in the copper outer canister has been adequately dealt with. This is necessary in order to eliminate the possibility of effects like concentration of impurities such as sulphur in grain boundaries, which could lead to brittleness or accelerated galvanic or crevice corrosion. Also the presence of long grain boundaries at the surface is crucial as they could open up during processing (for example through hot tearing in the weld area). Furthermore the presence of hydrogen at levels likely to cause hydrogen embrittlement is an important problem. It is likely that some or all of these are of no consequence in the greatest proportion of cases. It is most unlikely that none of them are of any importance in more than 0.1% of cases.

The phosphorus, oxygen and hydrogen contents proposed are generally recognised as satisfactory as specified and the sulphur content is acceptable in the absence of coarse grains or alloy concentration effects near welds. The desired grain size of 250μm has not yet been achieved in a product of this size and alloy composition and there is no good evidence to indicate that it can be achieved on a routine basis. Even if there were, there is no evidence that material of this composition and grain size is free of the low creep ductility problem. It is possible that if the present copper alloy is accepted as the overpack material coarser grain sizes than 250μm may have to be tolerated. If this becomes the case it will be necessary to reconsider the impurity levels which are acceptable and to develop improved ultrasonic inspection procedures.

There is not as yet any systematic work reported on alloy concentration, particularly with respect to copper, in the region of electron beam welds. Since work supported by SKB and referred to above has strongly suggested that low creep ductility is related to coarse grain sizes [11] and sulphur segregation in these materials it should be investigated more thoroughly.

The benefits of using chromium, zirconium or tin bearing alloys has been mentioned by one of us (WHB) on a number of occasions and it appears that they are generally accepted. In the cases of chromium and zirconium it is pointed out that such alloys are more difficult and therefore more costly to produce. This is certainly the case although in the view of one of us (WHB) they should not be totally discounted until a successful alternative has been demonstrated. The possible use of a tin bearing alloy appears to be under further consideration and it could have benefits in the processing and handling of the canister through its solid solution strengthening effect. It is rightly pointed out that it has the effect of increasing the recrystallisation temperature but it should be born in mind that this does not necessarily emerge as a reduced grain size in a hot worked product for the reasons presented earlier when phosphorus was discussed. The zirconium or chromium bearing material can be made to exert their grain refining effect during hot working. They therefore offer the possibility of a fine grained product in the section size required for the SKB programme.

There is no problem controlling grain size in cold rolled and annealed sheet or in hot rolled or extruded products where a big enough reduction is taken and cooling is controlled. Unfortunately there is serious doubt that adequate reductions may be taken when the required material thickness is to be produced.

Section 6.4.1 implies that a suitable full size canister has been created. This is not the case. One full size vessel has been produced and it contains massive welding defects which have proved not to be repairable. Even in the absence of the welding defects it is not established that it would be suitable or that the fabrication method is applicable to serial production. The particular canister shown in 6.3 may represent a good starting point for the development of a fabrication process if the production and metallurgical problems can be solved.

Points of concern at this stage are:

(1) the lid of the inner vessel was welded in place after preheating to a temperature of 250 °C. It is necessary to show that it is acceptable to preheat the canister top in this way when it is filled or to have a practice which does not involve preheating. If the lid is mechanically fixed in place it is necessary to demonstrate that it does not leak to release species which will interfere with the electron beam welding of the copper lid.

(2) the outer copper canister was produced from an extruded tubular by welding on a top and a bottom. The tubular had a very coarse grain size with grains of several millimetres in diameter in some areas. It may or it may not be possible to produce a tubular having the target grain size by this route. There is no evidence to suggest that it is possible at this stage. It is certainly possible to use a lower starting temperature for extrusion but the lower the starting temperature the greater will be the extrusion force required and the self heating during extrusion. It is necessary to demonstrate at an early stage that a suit

able grain size and structure can be produced since. If this is not achieved the process is unsuitable.

(3) a mysterious defect was present all over the surface of the inside of the extruded product. It was similar to the marks which appear on the surface of a continuously cast billet and which mark the positions of the mould during the casting process. The defect suggests that melting may have taken place during extrusion. It is necessary to know whether this defect persists in future extrusion trials and if so ,what effect it has on the structure of the extrusion.

(4) the tubular for the copper canister was neither round or straight as a result of distortions which occurred during cooling or handling. It proved possible to machine a straight and round tubular from it but this involved a protracted process of measuring and jigging in order to remove variable amounts of material around any diameter and at any position along the length. In this case the machining allowances were generous. If serial production is contemplated, it is necessary to determine the machining allowances which must be made and to accept the cost of this and of the measuring and jigging operation which will be specific to each individual canister.

(5) the material was extremely difficult to machine owing to its very soft condition and this is unlikely to change even if the other problems are overcome.

(6) the welds were unsound. The base weld had very serious defects which were not repairable, both the lid and the base had smaller defects which were shown up by ultrasonic testing. The seriousness of these defects is not known. It is known that the ultrasonic testing method does miss many defects and that this will be accentuated by the coarse grains.

(7) the claim that Electron Beam Welding, EBW has been shown to be a feasible method for fabricating canisters (section 6.5.4) is premature. It has been shown using a prototype equipment that specimen welds of the type required for the manufacture of canisters can sometimes be made. The welds which have been made are all imperfect and would be unlikely to be accepted for any engineering application. The special needs of the canister situation will allow large defects to be tolerated and some of the welds made have been free of such large defects. The question of the actual size of defects which may be accepted in any location needs to be addressed at an early stage. There is no evidence that these welding defects can be or have been systematically avoided in the work to date. There is some evidence that the welding process may be operated close to its practical limit and that this is the cause of some quality problems and some equipment failures in practice. This possibility should be realistically addressed at an early stage.

(8) the production of large copper ingots is referred to. Outokompu are known to have made one or two at around one metre in diameter. No details of the process or of the quality of the product are yet available. The quality of the ingot and its size will be important to the quality of the product extruded from it. Production and extrusion of the ingot in the short time scales of the programme is a very considerable achievement. If

the extrusion route is adopted it will be necessary to explore the production technology and the quality of the ingot as cast and the effect of ingot quality on the quality of the extrusion.

(9) material for the production of roll formed cylinders has been produced at a number of locations and at thicknesses between 60 and 70 mm. The choice of a fabrication route with two longitudinal welds rather than one has increased the number of producers capable of carrying out the rolling as well as simplifying the roll forming process. Grain sizes of 180 to 360μm have been quoted for these sheets. It is acknowledged that the rolling reductions used were less than ideal. Unfortunately it is not at all certain that ideal reductions can be achieved by rolling alone. A combination of rolling and forging may be feasible but this effectively constitutes the development of a new process. The rolling reduction is significant owing to its effects on final grain size and structure. In the cases of the sheets produced for this programme the rollers took great care to target fine grain structures. Care should be taken in interpretation of the grain size figures quoted. It is customary in production to quote an average grain size. When nominally inadequate rolling reductions have been used it is common to achieve a mixed grain size with some very coarse grains present. It is important in these cases to quote the range of grain sizes as well as the average.

Mixed grain sizes may be more favourable than all coarse grain sizes from the point of view of grain boundary volume. If they are to be used it is necessary to ascertain the stability and corrosion resistance of the structures under simulated operating conditions since mixed grain sizes are considerably less stable than uniform grain sizes.

(10) it is stated that both roll forming and press bending meet the precision requirements for electron beam welding and machining of the finished cylinders. The roll forming process used for this work is a craft operation depending on the skill of the operator. The expression "within a few millimetres of tolerance" is used in section 6.4 to describe the straightness and roundness of a fabricated tube. This is difficult to interpret. Does it mean that the dimensions were within the tolerance limit or does it mean that they were only a few millimetres outside the allowance. Allowing a higher tolerance will mean using a thicker plate which in turn will accentuate the problem of holding the required grain size. The first tubular prepared by roll forming and welding distorted considerably during welding. This varied both radially and longitudinally to such an extent that the proposed tubular could not be machined from it. It is not known how much of the loss of tolerance was due to welding and how much is due to the original plate rolling. It is clear, however, that the process is problematical. The craft nature of the rolling is a matter for concern and there is no obvious way to automate the process and remove the craft element.

The welding introduces distortions which may be controlled by welding in a constraining jig. Unfortunately such a procedure would result in an undefined internal stress system. Since the constraints prevent permanent distortions of the product it is reasonable to suppose that the internal stresses introduced by the constraints will exceed the proof stress of the material and that plastic deformation will have occurred. If this method is used it will be necessary to decide whether these residual stresses are acceptable or whether a stress relieving treatment will be required. It will also be necessary to explore the possibility of hot tearing associated with the welding.

The concerns expressed in this section add to the general feeling that the time allowed for the whole programme is inadequate.

7.6 Development of sealing technology

In sections 6.3.2 and 7.6, weldability and Non Destructive Testing, NDT are dismissed in a few words. It should on the contrary be made clear that neither the welding nor the NDT problems are solved.

7.6.1 Welder

It is known that the need to weld into a blind joint creates very serious problems for the electron beam welder. This is because very high powers are necessary to weld in a low pressure rather than a vacuum system. The power which has been used is 80 kW (220 KV at 360 ma) and this is ten times higher than the power required for a standard weld. Such power levels have never before been used in systems other than high vacuum systems and the technology is at present being operated on its limit. It is not known at this stage whether the limit is fundamental or technological and therefore it is not known whether or not the limit can be extended.

In brief the reason for the concern is that all Electron Beam Welders are prone to flash-over, that is a discharge between the electron gun and the support structure. As well as interrupting the welding, flash-over can have serious consequences in terms of damage to the equipment (a 5A discharge at 220 KV corresponds to 1.1 MW). The tendency to flash-over increases with beam power and with the gas pressure in the system. The strategy for managing flash-over is to detect it when it happens and ramp down the beam voltage very quickly. In order to maintain the stable weld pool it is ramped up again equally quickly when the discharge ceases. Clearly repeated discharges may happen and when they do the beam is repeatedly interrupted and restarted. The control system is designed to close the whole system down for safety reasons if discharges continue after a set number of repetitions of the interruption process. The problem with this strategy is that it does not prevent flash-overs happening. It is not clear what effect the flash-overs have on the materials of the system but it seems likely that damage is cumulative. It is therefore important to examine the durability of the materials in the system under operational conditions. The process of ramping the current down and up again causes severe transients which may be wire born or air born, such transients have caused failures in the control systems. It is necessary to screen against these effects in an operational system. Shutting down the system during the progress of a weld causes massive defects which would be very difficult to repair. Failing to shut down could be much more serious. There is no alternative strategy for managing flash-over at present. It is therefore important to know what the effects of high power discharges are on the materials of the EBW system and that adequate screening methods are available to protect the control systems.

7.6.2 Non destructive testing

Development of satisfactory NDT procedures is complicated by the fact that the types of defect which need to be detected have not been fully defined. In section 6.3 reference 6/43 is used to confirm the extreme resistance of the almost pure copper material to crack growth. The results given indicate that the container is not at risk of mechanical failure arising from cracks or pores in the structure which would escape detection by very basic test methods. Tests carried out at 20 °C and 100 °C indicate that the structure is likely to be safe against this type of failure from the time of manufacture through its service life. The results of work reported in ref. 6/43 suggest that the material is also safe against cold tearing in the heat affected zone of welds under the influence of welding stresses. Severe welding stresses are still likely and it is to be expected for reasons given earlier that they will exceed the proof stress of the material. The point is rightly made, however, that near surface or surface breaking defects could give rise to crevice or other types of localised corrosion. At this stage all reasonable steps to ensure the integrity of the material appear to have been taken. At a later stage it will be necessary to examine material from representative production canisters to demonstrate that it is not susceptible to stress corrosion cracking and that it is free of surface cracks resulting from hot tearing in the heat affected zones.

Investigations of creep crack growth are mentioned and it is stated that so far no growth has been observed. This is encouraging. In the long term it will be necessary to understand the factors which lead to low creep ductility and to ensure that the material and its processing are specified to avoid them.

Ultrasonic inspection has been studied by Day in the canister context and FUD-95 refers to his work (refs. 6/44 & 6/45). Section 6.3 states "The pulse-echo technique was able to detect defects in the weld down to 2 mm in diameter with a signal to noise ratio of 6 dB" and "detectability for defects was not always directly related to size." This can be misleading, it disguises the fact that a defect measuring 21 mm^2 in area was not detected.

It is claimed that detectability of deeper lying defects is fully adequate by both digital radiography and ultrasonic inspection. This is based on the assertion that a defect would have to be very large indeed before it would cause a problem rather on a conviction that deep defects are easy to detect. Fracture toughness work (ref. 6/34) indicates that the acceptable size for defects which will not become exposed by corrosion is very large indeed. Work on development of test methods continues and this is appropriate. It is also appropriate to note that test methods will be required for material with the grain size which will be realised in serial production, this could well be coarser than the 250μm which has been considered to date.

Section 7.6 states that rigour in process inspection of the metal containers and post sealing inspection of the canister will be carried out. It also says that methods will be determined, and if necessary, developed simultaneously with methods for fabrication and sealing of the canister and that this will take place over the next few years. This is not unreasonable but it would be useful to see some of the detail of the proposed development plan, and in particular the preferred options at this stage.

7.6.3 Alternative technologies

Alternative methods for production of the copper canister is referred to in section 7.5 and detailed in section 6.4.2. Alternative methods for joining the copper lid are discussed in section 6.5.2. Alternative technologies which are considered for the production of the copper canister are Hot Isostatic Pressing (HIP), electrodeposition and spray forming.

The references to hot isostatic pressing (section 6.4.2) suggest that this could be a viable route providing that the problem of internal stresses in the copper could be resolved. The stresses would be very uniform and a method for removing them is given in ref. 6/32. Refs. 6/33 - 6/36 are in Swedish. They are reported to show that in large scale trials good mechanical properties were achieved but that the material suffered from "hydrogen sickness". This is a potentially serious problem. In the absence of the hydrogen embrittlement problem hot isostatic pressing offers a very attractive route. It would no doubt have its problems and it would require dedicated plant. It is difficult to see how it could be more difficult or more expensive than the current candidates and in the absence of hydrogen related problems it provides a technically far superior product. In the opinion of the writers it is worthy of deeper consideration. The possibility of hydrogen related problems could be checked on a scaled down model which could be made in existing plant.

Electro deposition is also is a candidate worthy of deeper consideration and the first step must be to understand the properties of material made in model experiments.

Spray forming is a fairly new process in which atomised material is deposited on a former in the atomising chamber before the atomised droplets are solidified. Intuitively one feels that a spray formed product may need to be consolidated by hipping but the experience at Sandvik should throw light on that. If hipping is required it would be necessary to justify spray forming against the straight powder metallurgical route. Such a justification may be possible in terms of economics and of elimination of hydrogen related problems.

All the processes which form an overpack directly on the inner liner are worthy of consideration because they all offer a possibility of providing a pure copper overpack in a fine grained form with no problems related with integration with or collapsing onto the inner vessel.

Section 6.5.2 considers alternative welding methods. It dismisses diffusion bonding and brazing too easily on the basis that they require long soaking times at high temperature. There is no doubt that if a satisfactory electron beam welding process is available it is a good choice for the encapsulation programme, if only because it does not introduce effects due to alloying elements which have not so far been investigated. At present however there is still some doubt that a satisfactory electron beam process will be available. It is appropriate to persevere with the EBW process but alternatives are worthy of investigation and they may be necessary. A clearer understanding of the costs and benefits of the alternatives may lead to them being preferred.

Friction welding is the current preferred alternative and the small scale trials have given promising results. The power required for a full scale equipment is estimated in ref. 6/42 at 5 MW. This could be viewed as a disadvantage in the light of the engineering which would be involved. In view of the relatively low cost of testing friction welding at larger scale using equipment at Saab Scania in Luleå (ref. 6/42) it is surprising that it has not been done.

The papers quoted on diffusion bonding are in Swedish and I (WHB) have not therefore read them. Diffusion bonding without an interlayer is attractive because no uncertain effects of alloying are introduced. The disadvantage is that long diffusion times may be necessary. Transient liquid phase bonding is a form of diffusion bonding which is close to brazing. It has the advantage that it requires lower temperatures and shorter times but it does introduce the uncertainty associated with alloying. In view of the great benefit of the simplicity which it offers, it is in the opinion of the writer, worthy of closer scrutiny.

There is a suggestion that copper has been selected as the overpack because studies have shown that such an overpack would probably survive for millions of years and that this provides a considerable margin of safety, which is needed because of the considerable uncertainty in the premises. It should always be remembered that the uncertainties related to the durability of copper in the manufactured state could erode that margin of safety to unacceptable levels. There is a danger that the shortcomings of the copper overpack which arise from manufacturing procedures may be accepted because they are within the “considerable margin of safety“ which has been suggested. Such erosion of the safety margin could easily result in a product with a much lower residual safety margin than that which may be achieved by using other candidate materials (such as stainless steel) which present fewer manufacturing problems.

Conclusions and recommendations

1 We have found the programme difficult to review because of the very small amount of detail in section seven.

2 Section seven presents only the vaguest of outlines to the programme, with no detailed actions and no timescales for meeting specific objective. This will make it very difficult to monitor progress.

3 The objectives of the programme are given against the background of the information given in chapter six. We feel that the programme is converging too quickly, that the difficulty of meeting the objectives is seriously understated and this may be a result of the overoptimistic interpretation of information which is presented in chapter six.

4 We agree with the observations in chapter six 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 agree that alternative technologies such as Hot Isostatic Pressing (HIP), Electrodeposition, Spray Deposition (Osprey Process) merit consideration for production of the outer canister, in particular we feel that the Osprey process followed by HIP is worthy of investigation. We also agree that alternative joining processes such as friction welding, diffusion bonding or brazing may provide satisfactory processes for joining the canister lid but that further research and development would be required to establish this. A single canister having the materials of the reference case in the overpack and the dimensions of the reference case overall has now been produced. In view of the uncertainty of performance and the certainty of difficulty which exists with the reference case we are surprised that greater prominence is not given to the alternatives in the programme.

5 We disagree with the assertions in chapter six, that

(1) Suitable full size canisters have been created.
(2) Production technology is available for both canisters at full size.
(3) Copper canisters can be produced with adequate quality.
(4) The grain size of hot worked copper can be controlled by additions of phosphorus.
(5) A copper canister with a lead infill is a reasonable alternative canister to the reference case.
(6) Electron Beam Welding has been shown to be a feasible production method.
(7) The proposed change to a cast insert is a minor modification to the reference case.
(8) Defects down to 2 mm in diameter can be detected by ultrasonic methods in the copper canister.

6 We do not feel that is has been sufficiently recognised that materials properties which are to be used in design procedures must be derived from material which has been processed according to the proposed production procedures and is in the same metallurgical condition that would be achieved in production. Consequently we consider that further research is required on the metallurgical structures which will be achieved in production and on their properties. In particular more research is required on the combined effects of sulphur content, phosphorus content and grain size on creep properties and on the levels of residual elements which may be permitted as a function of grain size when corrosion properties are considered. It is also necessary to have better detailed knowledge of the factors controlling the grain size of heavy copper plate after processing by hot working methods and on the stability of uniform and mixed grain size material under repository conditions.

7 We feel that the presence of several types of metallic materials will contribute to the risk of galvanic corrosion. We cannot see in the programme that these matters will be properly investigated. The presence of large amounts of cast iron in contact with noble metals such as copper and zircaloy will increase the risk of a production of large amounts of corrosion products that could cause a spectrum of problems. The anticipated influence of corrosion products on the surrounding filling is an example.

8 Different types of localised rather than general corrosion would constitute problems. Localised corrosion could be initiated by several mechanical as well as chemical reasons and the mechanical state of the canister and its outer surface is important for the initiation of subsequent corrosion. Local corrosion could also be the cause of copper sulphide growth causing development of whiskers.

9 It is stated that only oxygen and sulphide can corrode copper. This is a too strong statement as chloride as well as complexing agents can contribute. When oxygen has disappeared, chemical agents like Fe(III), polysulfides, etc. could act as electron acceptors at sulphide corrosion in reducing environment. Furthermore, the presence of oxidants as related to effects of radiolysis should be re-evaluated in relation to the new canister concept.

10 It should be emphasised that the scientific basis for copper corrosion is limited to the simple system of water-copper at low temperatures. The repository environment is far more complicated. As an example, interaction of temperature gradients could cause a differentiated chemical environment creating subsequent electrochemical concentration cells of corrosion.

11 There is going to be only 10-20 % of bentonite in the filler system. This puts a large emphasis on filler quality control as the risk for inhomogeneities and subsequent good conduction of water and gas will increase. This is very important in conjunction with an ice age as the pressure and thereby transportation pattern could be very different from the normal case. The buffer would act as a filter for colloids. However there may be no electrostatic filtration in the buffer by electrostatic actions, only by mechanical. Perhaps this is sufficient to hinder colloidal transportation through the buffer but the situation should be considered more carefully.

12 Areas which need further evaluation include:

(1) The possibility of non uniform loading and its effect on the canister, including creep of the overpack.
(2) The effects of departure from circularity on the mechanical stability of the canister in service.
(3) The feasibility of routine electron beam welding using very high power levels in a coarse vacuum environment.
(4) Quality control of very large continuously cast copper ingots.
(5) The effects of radiolysis on the required thickness of the overpack.
(6) The corrosion properties of the canister in the repository environment, inside and outside the canister.
(7) The effects of residual stress on stability of microstructure and mechanical and corrosion properties of the overpack.

13 We do not feel that sufficient emphasis has been placed on the further development which is required in connection with:

(1) High power electron beam welding.
(2) Machining of the overpack material before and after lid sealing.
(3) Production of the insert by casting.
(4) Sealing of the lid of the insert.
(5) Production technology for the overpack.
(6) Non destructive testing of the entire system.
(7) The system for lifting the finished canister.

14 We consider that more information should be provided on the detail and timing of the development plan for the canister taking into account the following:

(1) The trial fabrication programme.
(2) The test programme for canisters produced in the trials.
(3) The determination of quality standards for the canister.
(4) The development of non destructive testing procedures.
(5) The proposed methods for determination of the probability of meeting defects of selected types.

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STATENS KÄRNKRAFTINSPEKTION
Swedish Nuclear Power Inspectorate

Postadress/Postal address	**Telefon/Telephone**	**Telefax**	**Telex**
SKI S-106 58 STOCKHOLM	Nat 08-698 84 00 Int +46 8 698 84 00	Nat 08-661 90 86 Int +46 8 661 90 86	11961 SWEATOM S