



UNITED STATES DEPARTMENT OF COMMERCE
National Bureau of Standards
Gaithersburg, Maryland 20899

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May 15, 1986

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Mr. Everett A. Wick
Division of Waste Management
Office of Nuclear Materials Safety and Safeguards
U.S. Nuclear Regulatory Commission
Washington, DC 20555

Re: Monthly Letter Status Reports for April 1986 (FIN-A-4171-6)

Dear Mr. Wick:

Enclosed is the monthly progress report for the project "Evaluation and Compilation of DOE Waste Package Test Data" (FIN-A-4171-6). The financial information is reported separately.

On April 28th, a review of this work was given for the management of the NBS Institute for Materials Science and Engineering. This review included the following topics, each of which was only briefly discussed: A Summary of the DOE Mission; Constraints on Waste Repositories in the U.S.A.; Contractual Obligations; the NBS Budget; Staffing; Technical Issues for Salt, Basalt, and Tuff.

Sincerely,

Charles G. Interrante
Program Manager
Corrosion Group
Metallurgy Division

1 Enclosure

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Monthly Letter Report for April, 1986

Published May 1986

(FIN-A-4171-6)

Performing Organization: National Bureau of Standards
Gaithersburg, MD

Sponsor: Nuclear Regulatory Commission
Office of Nuclear Materials Safety and Safeguards
Silver Spring, MD 20910

TASK 1 - Review of Waste Package Data Base

WERB Reviews --

A preliminary set of procedures for review of this work by the Washington Editorial Review Board (WERB) has been suggested. This plan will have to be finalized in May and June of this year.

Instruction for Reviewers --

The set of instructions developed for reviewers has been refined so that a technical expert, who has had only a minimal interaction with this program, should be able to use them to furnish reviews of high quality. We do anticipate that a reviewer will require some interactions with our staff (mainly with the lead workers) to furnish details in the reviews that satisfy specific needs of the program.

These instructions are regarded as important to this work, as reviews will be made by various technical experts, some of whom have had no direct involvement with this program. This is especially important for anyone working as a subcontractor at a site remote from the NBS. A copy of our latest version is attached for comment by NRC and its contractors.

SRP

No reviews of SRP reports were initiated during this month. The following report is under consideration and will be reviewed when a suitable reviewer has been identified:

1. "Waste Package Reference Conceptual Designs for a Repository in Salt", Westinghouse Electric Corp., BMI/ONWI 517, Feb. 1986--
To be reviewed: 1. Appendix G is on Corrosion Analysis
2. Appendix H is on Structural Parametric Studies

2. "ERG Review of Salt Constitutive Law, Salt Stress Determinations and Salt Corrosion Modeling Studies" by Jo Ellen Balon., BMI/ONWI 592, March, 1986--
To be reviewed: 1. Section on Salt Corrosion

BWIP

Reviews were initiated this month on the following BWIP reports:

1. "Short-term Stress-Corrosion-Cracking Tests for A36 and A387-9 Steels in Simulated Hanford Ground Water", L.A. James., SD-BWI-TS-012, January, 1985.
2. "Technical Progress Report on BWIP Canister Materials Crack Growth Study for FY 1983", L.A. James and L.D. Blackburn., SD-BWI-TI-165, January 11, 1984.

"BWIP Crack Growth Studies" by L.A. James and L.D. Blackburn., SD-BWI-TI-120, January, 1983.

Review of the following report is essentially complete and ready for WERB review:

"Pitting Behavior of Low-Carbon Steel, J. B. Lumsden and R. L. Fish, prepared by Rockwell Science Center., BWI-TS-014, August, 1985.

NNWSI

Review of the following report is essentially complete and ready for WERB review:

"Derivation of a Waste Package Source Term for NNWSI from the Results of Laboratory Experiments," V.M. Oversby and C.N. Wilson, UCRL-92096, Materials Research Society, Symposium on the Scientific Basis for Nuclear Waste Management, Stockholm, Sweden, Sept 9-11, 1985.

Review was initiated this month on the following NNWSI report.

"Radionuclide Release from PWR Fuels in a Reference Tuff Repository Groundwater", C.N. Wilson and V.M. Oversby, UCRL-91464, March 1985.

TASK 2 - Identification of Additional Data Required and Identification of Tests to Generate the Data

NBS lead workers are continuing their studies concerning the types of additional data (and the types of verification tests) needed to demonstrate that the DOE waste package designs will meet the performance objectives of 10 CFR 60. Our current objective is to develop a list of laboratory studies that will include both those to be conducted by the DOE as well as those to be conducted by the NBS under Task 3. A preliminary list of tests to be conducted at the NBS is being developed for joint NRC/NBS consideration before testing is initiated.

TASK 4 - General Technical Assistance

On April 13th and 14th, U. Bertocci and N. Pugh represented the NBS for the NRC at a Workshop/Seminar on Copper-Base Waste Package Container Materials, which was held in Houston. Their report on the meeting is appended to this report.

5/1/86

Instructions to Reviewers

After carefully reading the document to be reviewed please fill out (1) the Waste Package Data Review Form and (2) the Key Word Checklists. Note that these two items are interrelated, with the relationship indicated in the "Key Word Checklist Guidelines". The reviewer should first fill out one of these items (depending on his or her preference) and then using that information, complete the second item. In order to complete the Waste Package Data Review Form, please refer to the "Waste Package Data Review Form Guidelines". In these guidelines, the elements to be included under each major heading of the form are described along with an example. In general, the review form should be completed in an abbreviated (outline) format. A major exception to this generalization is that under the "General Comments" heading, complete sentences (using proper grammatical rules) should be used.

If there are areas covered in the document that are outside the expertise of the reviewer and which need to be assessed, please indicate this on a cover letter when returning the review. This is especially true if the reviewer feels the major conclusions of the paper depend upon the uncertainties in the data and, therefore, need a statistician to examine the data. Of course, in this latter case, a statement to this effect should also be included in the "General Comments" heading of the review form.

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Waste Package Data Review Form Guidelines

TYPE OF DATA:

- (1) Scope of the Report: e.g., Experimental, Theoretical, Literature Review, Data Analysis
- (2) Failure Mode or Phenomenon Studied: e.g., Corrosion, Creep, Fatigue, Leaching, Pitting, Hydrogen Embrittlement, Debonding, Dealloying

MATERIALS/COMPONENTS:

Description of the material studied (or component part, if specifically addressed as such; e.g., the screw-on type cap on a waste cylinder): e.g., 304L Stainless Steel, Brass, Zircolloy Cladding, Welds in 316 Stainless Steel, Packing Material, Basalt.

TEST CONDITIONS:

Includes (1) the State of the material being tested, and (2) the Environment of the material being tested, e.g.:

- (1) Cold Worked or Annealed 304L Stainless Steel, Thermo-mechanical History of the material (or component) being studied
- (2) Aqueous environment, Radioactive surrounding, Electrolytes or corrosive agents present, Temperature and pressure (externally applied or not) during the test

METHODS OF DATA COLLECTION/ANALYSIS:

Includes Data Measurement Methods and Types of data measured, as well as Data Analysis Techniques, e.g.:

Electron microscopy, weight loss vs. time, slow strain rate tensile test, x-ray diffraction, differential thermal analysis, A.C. electrical resistivity using a Wheatstone bridge, mass spectroscopic chemical analysis of the corrosive environment, Latin Hypercube method, Monte Carlo techniques

AMOUNT OF DATA:

Includes the number of tables and graphs of data together with their titles and axes (indicating the range in values), e.g.:

5 tables of temperature and time data for five molten glass pouring operations, each table including the data from ten sensor locations. The temperatures ranged from 1100 °C to 0 °C over a time period of 24 hours.

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UNCERTAINTIES IN DATA:

Included here are error bars and uncertainties in the data as stated by the author. This also includes qualitative statements by the author on the reliability of the data, e.g.:

Temperatures carry an accuracy of +5 *C while the times are reported to within +15 sec. It was felt that under real glass pouring operations (without well controlled crucible cooling) the temperature-time curves will be shifted to somewhat higher temperatures than shown here.

DEFICIENCIES/LIMITATIONS IN DATABASE:

Includes statements by the author on the applicability of the data, e.g.:

Extrapolation of the temperature-time (time < 24 hrs) data presented here to times in excess of 100 years should not be performed. The data presented here is useful only for indicating trends and qualitative parameter relationships, not for the purpose of presenting absolute values.

APPLICABILITY OF DATA TO LICENSING:

Indicated here is the licensing issue addressed by this paper. It is either (a) a specific Listed licensing Issue in an NRC Site Characterization Plan (ISTP) or (b) a new issue not yet identified in an ISTP.

The ranking of the paper is determined as follows: The "Key Data" box is marked if the paper contains data that is of sufficient quality that it must be considered by NRC in an evaluation of a license application. Such a paper must meet at least one of the following criteria: (1) it is an in-depth review of the pertinent literature, (2) it contains data that is found to be especially significant after being assessed for scientific merit and quality, or (3) it contains data with such a small uncertainty that it must be considered in a performance evaluation of a license application. If the paper does not meet any of the above three criteria, it is indicated as "Supporting Data". Reviewer's comments on the listing of the document may be included with the appropriate Issue Listing in subcategory (a) or (b).

GENERAL COMMENTS:

The reviewer's general comments on the document. This category is wide open as far as content. It contains information the reviewer did not enter into any of the above categories, but which is considered important for the reader to know. It would also be in this section that the reviewer would put any of his comments on the deficiencies and uncertainties in the data and analysis. e.g.:

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This is a very comprehensive review of the literature on the temperature sensitization of stainless steels. Even though it neglects the definitive work of Bertocci, Shull, Kaufman, and Escalante [Phys. Rev. J13, (1879), pp. 15-358] in this area (presumably because of the difficulty in locating this document), this review still considers a sufficiently large number of other investigations to provide a good understanding of the present status of the field. The one discordant note here, however, is that it would have been a much more useful review if stainless steel types 301, 303, 304, 316, and 440C had also been addressed.

DATA SOURCE:

Full document reference. This section will be completed for the reviewer before he/she receives the document.

KEY WORDS:

These are already entered, as they are included in the entries of the above categories.

DATE REVIEWED:

The date the document review was completed.

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Key Word Checklist Guidelines

The following is a listing of the major headings of the Waste Package Data Review Form indicating (both by title and list numbers) which key word lists may be filled out by the reviewer from the information that appears in that review form heading.

TYPE OF DATA: Scope (1), Failure Mode (13)

MATERIALS/COMPONENTS: Material Studied (6, 7)

TEST CONDITIONS: (a) State of the Material (5, 8, 12)
(b) Tests (3, 4, 9, 10)...

METHOD OF DATA COLLECTION: Measurement Type (11)
Model (2)

AMOUNT OF DATA: Number of tables, graphs, and titling
numerical results and their axes and
ranges

UNCERTAINTIES IN DATA:

DEFICIENCIES/LIMITATIONS IN DATABASE:

APPLICABILITY OF DATA TO LICENSING:

- (a) Relationship to Waste Package Performance Issues Already Identified
- (b) New Licensing Issue

GENERAL COMMENTS:

DATA SOURCE:

KEY WORDS: From List of Elements

1. Scope of Work

- 1. data analysis
- 1. experimental data
- 1. literature review
- 1. planned work
- 1. theory
- 1. other _____

2. Model/Methodology

- 2. Latin Hypercube
- 2. Monte Carlo
- 2. PDF (probability distribution functions)
- 2. sampling
- 2. scoping test
- 2. other _____

3. General Environment

- 3. J-13 water
- 3. basalt
- 3. field site
- 3. granite
- 3. laboratory
- 3. radiation field (alpha)
- 3. radiation field (gamma)
- 3. salt
- 3. simulated field site
- 3. tuff
- 3. other _____

4. Water Present

- 4. J-13 water
- 4. PH
- 4. basalt composition
- 4. brine
- 4. deionized
- 4. flow rate
- 4. granite composition
- 4. redox condition
- 4. salt concentration
- 4. significant dissolved species concentration
- 4. tuff composition
- 4. other _____

5. Other Materials Present in the Environment

- 5. Cl
- 5. Cu
- 5. Fe
- 5. J-13 water
- 5. Keller's reagent
- 5. Ni
- 5. sulfur ions
- 5. other _____

6. Material Studied (General Type)

- 6. brass
- 6. bronze
- 6. cast iron
- 6. cladding
- 6. copper base
- 6. electrolyte
- 6. general environment
- 6. nickel base
- 6. packing
- 6. radionuclide
- 6. stainless steel
- 6. steel
- 6. titanium base
- 6. water
- 6. weld
- 6. zircaloy
- 6. zirconium base
- 6. other _____

7. Material Studied (Standard Designation)

- 7. 304 stainless steel
- 7. 304L stainless steel
- 7. 308L weld filler wire
- 7. 316L stainless steel
- 7. AISI 317L
- 7. AISI 321
- 7. AISI 347
- 7. AISI 1020
- 7. AISI 1025
- 7. J-13 steam
- 7. J-13 water
- 7. bentonite
- 7. deaerated distilled water
- 7. distilled water
- 7. grey cast iron
- 7. high-nickel alloy 825
- 7. nodular cast iron
- 7. zircaloy-4
- 7. other _____

8. Material Condition Prior to Tests

- 8. case hardened
- 8. cast
- 8. cold worked
- 8. irradiated
- 8. magnetized
- 8. mill annealed
- 8. prestressed
- 8. sensitized
- 8. sintered
- 8. solution treated
- 8. stress relieved
- 8. textured
- 8. welded
- 8. wrought
- 8. other _____

9. Electrolytes

- 9. J-13 water
- 9. acetic
- 9. alkaline
- 9. aerated
- 9. chloride
- 9. deaerated distilled water
- 9. irradiated
- 9. neutral
- 9. other _____

10. Radionuclides and Materials Containing Them

- 10. Co60
- 10. Np237
- 10. Pu239
- 10. commercial high level waste (CHLW)
- 10. defense high level waste (DHLW)
- 10. spent fuel (power reactors)
- 10. spent fuel (water reactors)
- 10. other _____

11. Measurements

- 11. | adsorption
- 11. | electrochemical
- 11. | microscopy
- 11. | neutron diffraction
- 11. | slow strain rate
- 11. | sorption
- 11. | spectroscopy
- 11. | surface film
- 11. | tensile test
- 11. | thermal history
- 11. | visual examination
- 11. | weight change
- 11. | x ray diffraction
- 11. | other _____

12. Mechanical and Thermophysical Properties

- 12. | bent beam tests
- 12. | creep strength
- 12. | density
- 12. | elongation
- 12. | heat (conduction)
- 12. | heat (convection)
- 12. | heat (radiative)
- 12. | heat capacity
- 12. | hydrostatic head
- 12. | lithostatic pressure
- 12. | modulus of elasticity
- 12. | stress-strain
- 12. | tensile strength
- 12. | thermal conductivity
- 12. | thermal expansion
- 12. | yield strength
- 12. | other _____

13. Failure Modes or Phenomena Studied

- 13. buckling
 - 13. corrosion (crevice)
 - 13. corrosion (general)
 - 13. corrosion (intergranular)
 - 13. corrosion (local)
 - 13. corrosion (microbial)
 - 13. corrosion (pitting)
 - 13. corrosion (stray current)
 - 13. corrosion (stress cracking - SCC)
 - 12. creep
 - 13. creep buckling
 - 13. dealloying
 - 13. debonding
 - 13. deformation (elastic)
 - 13. deformation (plastic)
 - 13. degradation (spent fuel)
 - 13. devitrification (glass)
 - 13. diagenetic-like changes
 - 13. fatigue (corrosion)
 - 13. fatigue (high cycle)
 - 13. fatigue (low cycle)
 - 13. fatigue (thermal)
 - 13. fracture (brittle)
 - 13. fretting
 - 13. galvanic
 - 13. hydration (glass)
 - 13. hydrogen attack (CH₃ formation)
 - 13. hydrogen embrittlement
 - 13. leaching (radiation enhancement)
 - 13. leaching (spent fuel)
 - 13. matrix dissolution (glass)
 - 13. passivity
 - 13. poisoning (chemical)
 - 13. radiation effects
 - 13. relaxation (thermal)
 - 13. rupture (ductile)
 - 13. rupture (stress)
 - 13. sensitization
 - 13. spalling
 - 13. thermal instability
 - 13. other
-

Workshop on Copper-Base Waste-Package Container Materials
Houston, Texas, March 13-14, 1986

Comments of E. N. Pugh

Stress Corrosion Cracking (SCC)

Several speakers ruled out the occurrence of SCC in OFHC copper but their evidence was not convincing. Mattsson (SKB) discounted SCC on the basis of the assumption that only NO_2^- ions will cause this form of attack and that groundwaters contain less than the critical concentration required to cause cracking. Based on slow strain rate (SSR) tests by R. N. Parkins in aqueous NaNO_2 , the latter concentration was considered to be 69 mg/L. Similarly King (AECL/WNRE) ruled out SCC on the basis of the absence of NO_2^- in the Canadian environment. However, the role of NO_2^- in the cracking process is not understood, and it is clearly premature to consider that NO_2^- has a unique role. Other species may be found which also cause cracking. There has been little systematic experimental study of the effects of chemical species on SCC of copper and, in the absence of a theoretical understanding of the SCC process, such an experimental study would seem appropriate. The identification of a critical concentration of NO_2^- for SCC based on rather limited testing would also seem naive.

Anantamula (BWIP/RHD) ruled out SCC of OFHC copper (and Cu-10Ni) on more rational grounds. He reported that SSR (PNL) and Fracture Mechanics (WHC) tests have so far detected no susceptibility in simulated groundwater. He argued that SCC is unlikely in copper and its alloys because of stress relaxation, particularly in the unalloyed metal and particularly at the higher temperatures. This is a reasonable suggestion since failures of copper in aqueous NaNO_2 and more recently in aqueous NH_3 (unpublished work by K. Sieradzki at BNL) have been observed only in SSR tests. The severity of the SSR tests is well known. Thus it would be worthwhile to conduct SSR tests on OFHC copper in aqueous NO_2^- solutions and establish conditions where failure is rapid, and then conduct tests on the same material under the same environmental conditions but under both constant load and constant deflection. [Suggest these tests for preliminary experiments at NBS. We have already studied behavior under SSR conditions].

Werme (SKF) made an interesting comment on the occurrence of bentonite swelling in the Swedish design which he felt could cause creep failure at temperatures $< 100^\circ\text{C}$. He discounted SCC under these "SSR conditions" on the basis of the absence of a critical concentration of NO_2^- , but, as indicated above, this is not a safe assumption.

E-pH Diagrams

In his presentations, Mattsson made effective use of Pourbaix diagrams to represent environmental conditions in the Swedish environment. He used Fe- H_2O and Cu- H_2O -S, employing the latter to discuss the effects of S on pitting. It seemed that the use of these diagrams might profitably be extended to the US environments: they might provide an useful method of describing equilibrium in complex solutions. [This is a possible role for the Corrosion Data Center at NBS].

Comments of U. Bertocci

Introduction

The workshop consisted of a review of the work being done to study copper and several of Cu-base alloys as containers for high level nuclear waste. Reports came from three countries, Sweden, where Cu has already been chosen as the container material, the USA, where Cu and Cu-base alloys are being considered for three geological environments (basalt, tuff and crystalline rock), and Canada, where Cu is being considered for use in granite rocks but where the studies are still as a very preliminary stage.

Sweden

In the case of the Swedish program the pure Cu containers to be buried in a hole drilled in granite and backfilled with bentonite, are expected to last at least 10^6 years. The reasons for this belief are based on three points: 1) the radioactive waste will be cooled for a period of about 40 years before being buried, so as to decrease substantially temperature and γ -field; 2) the thickness of the containers is such that the intensity of the γ radiation outside the canister is low, leading to limited radiolysis; 3) the amount of oxidizing agents, reaching the container is so low that even assuming that all of it reacts with the Cu, only a small fraction of it would be corroded.

The uncertainties concerning the soundness of the Swedish plants, as far as corrosion is concerned, stem from two things:

- a) Reduction of sulfates to sulfides and their reaction to form Cu_2S . This reaction, although thermodynamically possible, has never been observed to occur, unless catalyzed by bacteria. However, if this reaction were to occur at a rate significant for the very long time involved, it would increase the amount of oxidizer to be reckoned with.
- b) Pitting - If the corrosion of Cu were to occur in a non-uniform manner, penetration of the attack would be much more than that calculated for uniform corrosion. At the present time, a value of 25 is taken as a worst case for the ratio between pit depth and uniform attack. This value is very uncertain, however, and it is believed to be conservative.

The Swedish program concerning corrosion, therefore, is aimed at reducing these two uncertainties: for point a) an upper value for the reaction rate is being sought: for point b), experiments are underway to determine the pitting ratio under more realistic conditions. If the experimental results show a smaller danger from pitting, the thickness of the canister walls may be decreased.

U.S.A.

The reports on the U.S. program were divided according to the geological environment. Of the three rock formations, both basalt and crystalline rock offer a reducing environment, while tuff is an oxidizing environment, where ambient air is supposed to permeate the rock.

The presentations of the various workers is not described here, because a report on this workshop will be published. Rather, comments are made on the most significant issues. Since the main difference in environmental conditions is whether they are oxidizing or reducing, the comments will be divided accordingly rather than by type of rock.

a) Reducing Environments

In both basalt and granite, the availability of oxidizers is quite low, so that rapid corrosion is very unlikely. However, γ -radiation fields and temperatures are significantly higher than those envisaged in Sweden, so that tests are being carried out at temperatures up to 300°C in the presence of steam and γ -radiation.

No result so far shows very serious problems, either in the form of general corrosion or pitting attack, either for Cu or for Cu 10%Ni alloy, but tests are continuing. From some perplexing results showing higher corrosion rates in the absence of oxygen than in air/steam, it appears that there may be competition between attack by sulfides and by oxygen. Electrochemical tests for pitting are also being carried out, under various conditions. Nothing very worrisome has yet been found.

From slow-strain tests carried out on Cu and Cu-10%Ni no susceptibility to stress corrosion cracking has been detected. A significant difference between basalt and granite is that the chloride concentration may be very high in the latter. Exact values are unknown, but some results of drilling

in Canada indicate the possibility that the fluid may be a very concentrated brine. However, oxidation of the copper to copper chloride requires either an oxidizer or sufficient acidity, and they may not be available. In any case, the work applicable to crystalline rock in this country is at too an early stage to draw any conclusions.

b) Oxidizing Environments

The repository site in tuff is above the water table, and the rock is porous, so that air and groundwater can easily percolate. It is expected that the canister will be at a temperature above the boiling point of water for the duration of the (1000 y) expected life of the canister. The survival of the container, therefore, will depend on its corrosion resistance and not on availability of reactants. Radiolysis in the presence of atmospheric nitrogen may give rise to a large number of compounds, whose effects are not well known. Therefore, the possibility of SCC cannot be easily discounted, also because there is no lithostatic pressure to counteract eventual tensile stresses. Enhancement of corrosion due to γ radiation has been reported in the literature.

The tests carried out so far have not shown any catastrophic effect, but on the contrary indicate low corrosion rates. The data available are not sufficient to eliminate all concerns, and the safe use of Cu-base materials in tuff is far from established.

Canada

The Canadian program is at an early stage, too early to contribute significantly to any conclusions as to the suitability of Cu as a container. The Canadians have still a few years, according to their timetable, before

establishing a conceptual design. The most interesting result reported concerns the composition of the "water" found after drilling in granite, which has shown increasing chloride concentration with borehole depth. One mol/L Cl^- has been found, up to a maximum of 282 g/L of dissolved low salts. In these conditions, and in the presence of γ -radiation the stability of Cu is open to question.

Mechanical Properties, Design and Fabrication

All participant groups presented some information concerning problems associated with the mechanical properties of Cu-base metals and with the fabrication of the canisters. A concern for the US groups is the lack of data at higher temperature (say around 300°C) particularly for creep. This concern is not shared by the Swedes, who plan to keep the containers at lower temperature. The Swedes have done some studies on fabrication, including welding of thick copper sections, and are encountering difficulties.

Conclusions

Cu and Cu-base alloys have not shown obvious defects after a fair amount of testing, so that for the moment they seem to be a suitable container material for high level waste, at least under reducing conditions, where corrosion resistance is really not important, since the corrosion rate is limited by the availability of reactants. In the case of USA conceptual designs, some doubts are related to the fairly high temperature and γ -field envisaged. Further testing is necessary to clarify some issues. In oxidizing conditions, the degree of uncertainty is much greater. Thorough examination of the behavior of the the materials under realistic conditions is necessary before Cu-base materials can be declared safe.

A reason for preferring Fe-base metals to Cu-base metals is their cost. However, the off-the-record opinion of many of the participants is that the cost of the material is such a small fraction of the overall costs that it may turn out to be insignificant.