e pocket AA171 DEHAR MENT OF COMMERCE National Bureau of Standards Gaithersburg, Maryland 20899 POR-1

August 15, 1987

WM DOCKET CONTROL CENTER

LADR - WM-10(2) um-11(2) Um-16(0)

187 AUG 20 A11:04

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 Report for July 1987 (FIN-A-4171-7)

Dear Mr. Wick:

Enclosed is the July 1987 monthly progress report for the project "Evaluation and Compilation of DOE Waste Package Test Data" (FIN-A-4171-7). The financial information is reported separately.

Sincerely,

charles D. D. turan

Charles G. Interrante Program Manager Corrosion Group Metallurgy Division

Enclosures

Distribution: NMSS PM (1) Office of the Director, NMSS (Attn: PPAS) (1) WM Div. Director (1) WMEG Branch Chief (1) WMEG Branch Materials Engineering Sec. Leader (1) WM Docket Control Center (4) Office of Research (1) Office of Resource Mgmt. (1) Div. of Automated Services (ATTN: Director)

With Project 10,11,16 87232857 WM Project: WM-10, 11, 16 NM Record File: A4171 Dockat Ro PDR w/encl LPDR w/encl JB≤ (Return to WM, 623-SS) Joan-tic 8709080133 870815 WMRES EUSNBS PDR PDR (Return to WM, 623-SS) A-4171-7

Sec Hr. to Wick fm: Interrante 8-15-87 A4/171

Monthly Letter Report for July 1987

Published August 1987

(FIN-A-4171-7)

Performing Organization: National Bureau of Standards (NBS) Gaithersburg, MD 20899

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

Task 1 -- Review of Waste Package Data Base

STATUS OF REVIEWS

Appended to this report are the following 15 Draft Reviews not previously submitted. Comments by NRC and its contractors are solicited.

- RHO-BW-SA-391P, "Effect of Grande Ronde Basalt Groundwater Composition and Temperature on the Corrosion of Low-Carbon Steel in the Presence of Basalt-Packing," August 1985
- SD-BWI-TS-008, "Slow-Strain-Rate Testing of 9% Cr, 1% Mo Wrought Steel and ASTM A27 Cast Steel in Hanford Grande Ronde Groundwater," October 1984
- 3. RHO-BW-SA-509P, "Thermal Analysis of Waste Package Preliminary Reliability Assessment," March 1986
- 4. B047154, "BWIP General Corrosion Studies, Summary Report of Activities in FY-1984," October 1984
- 5. B023959, "Enviromechanical Testing of AISI 1020 Steel in Hanford Grande Ronde Groundwater," July 1983
- 6. HEDL-7560, "Test Plan for Long-Term Low-Temperature Oxidation of Spent Fuel Series 1", June 1986

- UCRL-94659, "Spent Fuel as a Waste Form Data Needs to Allow Long Term Performance Assessment Under Repository Disposal Conditions," V. M. Oversby, 1968
- 8. BMI/ONWI-597, "Buckling Design Criteria for Waste Package Disposal Containers in Mined Salt Repositories," December 1986
- 9. "Radiation Damage Studies on Natural Rock Salt from Various Geological Localities of Interest to the Radioactive Waste Disposal Program," Nuclear Technology, <u>60</u>, 231-243, February 1983
- BMI/ONWI-626, "ERG Review of the SRP Salt Irradiation Effects Program," November 1986
- PNL-SA-14029, "Corrosion of Iron-Base Waste Package Container Materials in Salt Environments," Nuclear Power Conference, Philadelphia, PA, July 20, 1986
- 12. "Effects of Composition on the Leach Behavior of West Valley HLW Glasses," X. Feng, et al., September 1986
- 13. WVDP-056, "Description of the West Valley Demonstration Project Reference High-Level Waste Form and Canister," July 1986
- 14. "Glass Performance in a Geologic Setting," Summer National Meeting of the American Institute of Chemical Engineers, 1986
- "Repository Simulation Tests," American Ceramic Society Annual Meeting, 1984

BWIP -- BASALT WASTE ISOLATION PROJECT

ŝ

The major deficiencies in the available data pertaining to a waste repository in basalt fall into four areas: (1) packing material breakdown (2) groundwater chemistry (in so far as it affects the corrosion and solubility of metals) (3) corrosion of the metallic waste container and (4) leaching of the glass waste form in a basalt environment.

Review has been initiated on RHO-BW-SA-554P, 1986, "Determination of Dissolved Gases in Basalt Groundwater in the Pasco Basin, Washington." Gas contents in the surrounding liquid are very important in controlling the metallic corrosion reactions which will occur. For a repository in the proposed location, this information is of primary importance to the determination of the expected lifetime of the metallic waste containers.

Due to its lack of localized corrosion behavior copper has received recent attention in regard to its possible use as the metallic waste container in a basalt repository. Review has been initiated this month on a report by the Rockwell Hanford Operations, "Feasibility Assessment of Copper-Base Waste Package Container Materials in a Repository in Basalt" which addresses this topic SD-BWI-TA-023 1986.

A third area in which more information has recently become available is that of the radionuclide barrier performance of the packing material. Review of RHO-BWI-LD-43, 1981, "Sorption of Selected Radionuclides on Secondary Minerals Associated with the Columbia River Basalts," was also initiated this month.

BWIP -- Review is continuing on the following reports.

ŝ

- RHO-BW-SA-316P, "Irradiation-Corrosion Evaluation of Metals for Nuclear Waste Package Applications in Grande Ronde Basalt Groundwater," November 1983
- 2. SD-BWI-TS-012, "Short-term Stress-Corrosion-Cracking Tests for A36 and A387-9 Steels in Simulated Hanford Groundwater," January 1985
- 3. SD-BWI-TI-165, "Technical Progress Report on BWIP Canister Materials Crack Growth Study for FY 1983," January 1984
- 4. RHO-BW-CR-148P, "REPREL Computer Code: User Guide," June 1985
- 5. RHO-BW-SA-560P, "Status of Environmentally Assisted Cracking Studies by the Basalt Waste Isolation Project," Symposium on Radioactive Waste Management '86, March 1986
- 6. "Electromechanical Testing of AISI 1020 Steel in Hanford Grande Ronde Groundwater," S. G. Pitman, July 1983
- 7. RHO-BW-LD-48, "Sorption Behavior of Selected Radionuclides on Columbia River Basalts," August 1986
- "Methods of Simulating Low Redox Potential (Eh) for a Basalt Repository," Materials Research Society Proceedings, 1984
- 9. "Control of Oxidation Potential for Basalt Repository Simulation Tests," Scientific Basis for Nuclear Waste Management, 1985
- 10. NUREG/CR-4309, ORNL-6199, "Valence Effects on Solubility and Sorption: The Solubility of Tc(IV) Oxides," March 1986
- BWIP -- Review has been initiated on the following reports.
- 1. RHO-BW-SA-554-P, "Determination of Dissolved Gases in Basalt Groundwater in the Pasco Basin, Washington," September 1986

- RHO-BWI-LD-43, "Sorption of Selected Radionuclides on Secondary Minerals Associated with the Columbia River Basalts," Informal Report, April 1981
- 3. SD-BWI-TA-023, "Feasibility Assessment of Copper-Base Waste Package Container Materials in a Repository in Basalt," September 1986

NNWSI -- NEVADA NUCLEAR WASTE STORAGE INVESTIGATIONS

Preparation of the NNWSI portion of the biannual report was begun during the month of July. Two new papers were received during this month on the subjects of copper and spent fuel.

Review was initiated on, "Long-Term Corrosion Behavior of Copper-Base Materials in a Gamma-Irradiated Environment," by W.H. Yunker and R.S. Glass. After fourteen months of exposure to gamma radiation in three moisture environments at elevated temperatures, copper and two of its alloys are showing some corrosion attack. Pitting, crevice corrosion, and uniform corrosion have been observed. [UCRL-94500, 1986]

Review was also initiated on, "Results from Cycles 1 and 2 of NNWSI Series 2 Spent Fuel Dissolution Tests," by C.N. Wilson discusses leach testing of spent fuel rod segments with and without defects in the radionuclides. Release rates from rods with cladding containing small (approx 200 μ m) laser drilled holes was not significantly greater than that observed for undefected rods. [HEDL-TME-85-22, 1987]

NNWSI -- Review is continuing on the following reports.

- UCRL-15723, "NNWSI Waste Form Test Method for Unsaturated Disposal Conditions," March 1985
- 2. UCRL-94708, "Carbon-14 in Waste Packages for Spent Fuel in a Tuff Repository," October 1986
- UCRL-94633, "Experimental Study of the Dissolution Spent Fuel at 85°C in Natural Groundwater," December 1986
- 4. UCRL-95962, "Hydrogen Speciation in Hydrated Layers on Nuclear Waste Glass," January 1987
- 5. UCRL-94658, "Integrated Testing of the SRL-165 Glass Waste Form," December 1986
- 6. UCRL-91258, "Leaching Savannah River Plant Nuclear Waste Glass in a Saturated Tuff Environment," November 1984
- 7. DP-MS-85-141, "Leaching Fully Radioactive SRP Nuclear Waste Glass in Tuff Groundwater in Stainless Steel Vessels," May 1986

- UCID-20895, "Application EQ3/6 to Modeling of Nuclear Waste Glass Behavior in a Tuff Repository," May 1986
- 9. UCRL-92891,"LWR Spent Fuel Characteristics Relevant to Performance as a Wasteform in a Potential Tuff Repository," June 1985
- 10. ANL-84-81, "NNWSI Phase II Materials Interaction Test Procedures and Preliminary Results," January 1985

NNWSI -- Review has been initiated on the following report.

- 1. UCRL-94500, "Long-Term Corrosion Behavior of Copper-Base Materials in a Gamma-Irradiated Environment," December 1986
- 2. HEDL-TME 85-22, "Results from Cycles 1 and 2 of NNWSI Series 2 Spent Fuel Dissolution Tests," May 1987

SRP -- SALT REPOSITORY PROJECT

There are nine reports in the review process for the Salt Repository Project (SRP). Four completed reviews are attached in draft format to this report. One of the two reports mentioned in the June Monthly Letter Report as being notable, PNL-SA-14029, has been reviewed. Two important conclusions reached in this review were: (1) Changing corrosion rate with exposure time and environmental fluctuation is a key factor needing further study to assure valid extrapolations to longer term exposures and anticipated environmental variations, and (2) potential effects of alternate wetting and drying or waterline exposure on waste canisters in brines should be considered in future experiments.

The authors of this report, R. E. Westerman et al., assume no effect of irradiation on corrosion rate and do not address potential effects of H_2 pressure buildup in sealed containers in reducing cathodic reactions.

Two new reports, PNL-5650 and ONWI-384, have been identified for critical review. The first, "Spent Fuel and UO_2 Source Term Evaluation Results: FY-1984 Annual Report," will provide additional information on static leach tests of spent fuel in salt brines. These tests were conducted during FY 1984 at the Pacific Northwest Laboratory, Richland, Washington. The second report, "A Sensitivity Study of Brine Transport Into a Borehole Containing a Commercial High-Level Waste Canister," by Joe L. Ratigan of RE/SPEC Inc., will provide more information on brine inflows into a typical commercial high-level waste canister borehole in addition to identifying the relevant properties whose uncertainty contributes most significantly to the range of the predicted inflow.

During this month, preparation of input for the forthcoming draft biannual report was prepared.

SRP -- Review is continuing on the following reports.

- 1. BMI/ONWI-612, "The Effects of Stabilizers on the Heat Transfer Characteristics of a Nuclear Waste Canister," July 1986
- 2. DOE/CH-21, "Systems Engineering Management Plan for the Salt Repository Project," August 1986
- 3. UCRL-53726, "Reference Waste Package Environment Report," October 1986
- 4. BMI/ONWI-611, "ERG Review of Waste Package Container Materials Selection and Corrosion," July 1986
- 5. BMI/ONWI-583, "Waste Package Materials Testing for a Salt Repository: 1983 Status Summary Report," September 1986
- BMI/ONWI-490, "Waste Package Materials Testing for a Salt Repository: 1982 Status Report," August 1983
- PNL-3484, "Investigation of Metallic, Ceramic, and Polymeric Materials for Engineered Barrier Applications in Nuclear-Waste Packages," October 1980
- BNL-29909, "Radiation Damage Studies on Synthetic NaCl Crystals and Natural Rock Salt for Radioactive Waste Disposal Applications," Technology of High-Level Nuclear Waste Disposal, Vol. 1, 1981
- 9. PNL-5426, "Corrosion and Environmental-Mechanical Characterization of Iron-Base Nuclear Waste Package Structural Barrier Materials Annual Report -- FY 1984," March 1986
- SRP -- Review has been initiated on the following reports.
- 1. ONWI-384, "A Sensitivity Study of Brine Transport Into a Borehole Containing a Commercial High-Level Waste Canister," February 1987
- PNL-5650, "FY-1984 Annual Report: Spent Fuel and UO₂ Source Term Evaluation Results," February 1986

WASTE FORM DEGRADATION

Glass

ĸ

During July, several draft reviews of literature reports on glass leaching studies were completed, and a number of technical issues were identified.

Papers reviewed recently include some in which repository conditions are simulated in laboratory leaching studies. Simulations include, for example, rock cups used to hold the leachant and the glass sample. Pieces of the canister metal may also be added. These refinements make the tests potentially more reliable as indicators of behavior in real repositories. Repository simulation tests can, in principle, incorporate flow, but this variable has not been used in the simulation studies reviewed to date.

Glass leaching studies reviewed thus far have shown that the leaching rate of glass waste forms is lower in repository simulation tests than in other laboratory tests. This implies that the leaching rate will also be lower in a repository environment. However, more detailed data on the hydrology and geology of the repository site are needed to determine how well the simulation corresponds to actual repository conditions. This issue of site characterization remains critical.

Review of some key papers on the role of the redox potential, Eh, in glass leaching were completed through the draft stage. Background documents from Swedish research on this subject were also studied, and experts on Eh-pH (Pourbaix) diagrams were consulted to evaluate the work being carried out in the nuclear waste program. It was concluded (subject to modification as more papers on the subject are reviewed) that because the experimental methodology and the application of the concept are potential concerns, the data should be interpreted carefully. However, the basic qualitative conclusion of the work, i.e., that solutions of a more reducing nature are less aggressive in leaching the glass, should be valid.

WASTE FORM DEGRADATION -- Review is continuing on the following reports.

- "Chemical Durability Studies on Glass Compositions Pertaining to Waste Immobilization at West Valley," A. Barkatt, et al., Waste Management '86: Waste Isolation in the U.S. Technical Programs and Public Education, March 1986
- "Long Term Leach Behavior of West Valley HLW Glasses," P. B. Macedo, et al., ANS Spectrum, 1986
- 3. "Leach Mechanisms of Borosilicate Glass Defense Waste Forms -- Effects of Composition," A. Barkatt, et al., Waste Management '86: Waste Isolation in the U.S. Technical Programs and Public Education, March 1986
- 4. "Chemical Determination of West Valley Waste Form Products,"
 D. M. Oldman, J. R. Stimmel, and J. H. Marlow, March 1987
- 5. "Startup and Initial Experimental Results for the West Valley Vitrification Demonstration Project," Waste Management '86: Waste Isolation in the U.S. Technical Programs and Public Education, Volume 2 High-Level Waste, March 1986
- 6. "Method for Showing Compliance with High-Level Waste Acceptance Specifications," Waste Management '86: Waste Isolation in the U.S. Technical Programs and Public Education, Volume 2 High-Level Waste, March 1986

- 7. "Solubility Tests on Borosilicate Glasses for West Valley Waste Immobilization, High-Level and Transuranic Waste Management," X. Feng and A. Barkatt, ANS Transactions, 1986
- 8. PNL-5157, "Final Report of the Defense High-Level Waste Leaching Mechanisms Program," August 1984
- 9. "Physical Chemistry of Glass Surfaces," J. Non-Cryst. Solids, 1978
- 10. DP-MS-83-135, "Process Technology for Vitrification of Defense High-Level Waste at the Savannah River Plant," Paper for presentation in the proceedings of the American Nuclear Society Meeting on Fuel Reprocessing and Waste Management, August 1984
- 11. DP-MS-86-96, "Process and Mechanical Development for the Savannah River TRU Waste Facility," Paper proposed for presentation at the American Nuclear Society International Meeting, Spectrum '86, September 1986

OTHER REPORTS/TECHNICAL PAPERS -- Review is continuing on the following report judged to have related scientific value sufficient to warrant its review (as reference material).

 "Aging Degradation of Cast Stainless Steel," O. K. Chopra and H. M. Chung, October 1986

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 verification tests needed to demonstrate that the DOE waste package designs will meet the performance objectives of 10 CFR 60.

TASK 3 -- Laboratory Testing

.

Title of Study: Evaluation of Methods for Detection of Stress Corrosion Crack Propagation in Fracture Mechanics Samples Principal Investigator: Charles Interrante

The majority of the work on the Evaluation of Methods for Detection of Stress Corrosion Crack Propagation in Fracture Mechanics Samples experiment for the month of July was a continuation of the investigations cited in June's report. These were primarily in the areas of instrumentation and software applications. One of the major areas of concern during this period was the interfacing of the mini-computer with a digital oscilloscope. The purpose of this is to allow the mini-computer to constantly monitor the condition of the oscilloscope and transfer the corresponding acoustic data. Several complications arose with the software and work on resolving the problems is continuing. Another area of on-going work for this project, asks the question can the required tasks be performed in a more efficient manner. Our review of commercially available data acquisition and control systems is continuing.

ĩ

Last month, several materials were reviewed for their susceptibility to the intergranular form of stress corrosion cracking. The information gathered on these steels was discussed with various consultants particularly those with the corrosion group at Carpenter Technology Corp. in Reading, Pennsylvania. Their opinions confirmed our results for the choices of both materials and environments. Work on preparation of the fracture mechanics specimens from these materials and on the environmental aspects should begin next month.

In addition to the ferritic steel tests already planned, consideration is being given to the use of austenitic steels in either the partially or completely sensitized condition. In both cases, intergranular fracture models are to be studied first and transgranular fracture will be included latter.

Title of Study: Effect of Resistivity and Transport on Corrosion of Waste Package Materials Principal Investigator: Edward Escalante

During this month, specimens were machined from sheet stock, cleaned, and weighed. The corrosion cell has been designed to allow diffusion measurements to be made in addition to the corrosion rate measurements. This required placing an inert metal electrode in the cell along with the corrosion specimen. Configuration of these electrodes is important because current, applied during the experiment, must be uniform over the surface of both electrodes. Thus, they are positioned in such a way that minimizes current shielding during measurements. In addition, a special clear plastic cover was made to hold the electrodes in position. Improvements on the equipment and software are continuing.

Title of Study: Pitting Corrosion of Steel Used for Nuclear Waste Storage Principal Investigator: Anna C. Fraker

The work on this project is on schedule with the work statements listed in the proposal. The work statements and status of work conducted in July 1987 are given below. Work conducted in previous months was reported in June.

1. Obtain steel, set up equipment and environment (three months from start). Completed.

2. Determine the pitting potential, using stimulation techniques in simulated Grande Ronde No. 4 water at 95°C, (three months from start).

A stimulation test designed to determine pitting potential was run on A-27 steel in Grande Ronde No. 4 water at room temperature. The A-27 steel did

not passivate (develop a protective film on the surface) initially or during the test. The stimulation test does not apply to metals which do not passivate, and metals which do not passivate should not be subject to pitting. Lumsden indicated in the paper, "Pitting Behavior of Low Carbon Steel, BWI-TS-014, Aug., 1985" that pitting would not be expected to occur under repository conditions. The pitting potentials listed in this paper were those associated with a rapid rise in current at breakdown (rapid rise in current indicating increased metal corrosion and surface film instability). Indications are that the data of Lumsden regarding pitting potentials are correct. Additional tests will be run and further analysis of the data and specimens will be made.

3. Determine polarization behavior and pitting potential using cyclic polarization methods in simulated GR- 4 water at 95°C (12 months from start).

Specimen preparation and other preparations are underway to conduct the anodic and cathodic polarization measurements in GR-4 water at 95°C.

Microscopic observations of specimens anodically polarized in GR-4 water at 22°C do not seem to show pits at 200X magnification. More specimens need to be studied at higher magnification to check this finding. There is a thick film on the surface, and under this film, the specimen appears to be etched.

Title of study: Corrosion Behavior of Zircalloy Nuclear Fuel Cladding Principal Investigator: Anna C. Fraker

The work on this project is on schedule with the work statements listed in the proposal. The work statements and status are given below.

1. Obtain materials and testing environment (three months from start). Completed

Zircaloy-2 cladding was received from the General Electric Company, Vallecitos Nuclear Center, Pleasanton, California.

The Zircaloy-4 material, from Babcock and Wilcox, was received in the oxidized and nonoxidized conditions. Testing of the oxidized material will be limited since preoxidizing the cladding is not being done any more.

The J-13 water has been prepared.

2. Prepare a brief literature survey on Zircaloy corrosion (nine months from start).

This is in progress.

1

3. Anodic polarization curves for Zircaloy in J-13 water at 95 C (12 months from start).

Specimens of Zr-2 and Zr-4 are being prepared.

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

(a) Organization Producing Data

Rockwell International, Richland, Washington

(b) Author(s), Reference, Reference Availability

R. P. Anantatmula, "Effects of Grande Ronde Basalt Groundwater Composition and Temperature on the Corrosion of Low-Carbon Steel in the Presence of Basalt-Bentonite Packing," (RHO-BW-SA-391 P), August 1985

DATE REVIEWED: 1/28/87; Revised 4/30/87

TYPE OF DATA

Experimental data. Weight loss corrosion tests. Statistical analysis.

MATERIALS/COMPONENTS

1020 AISI Steel packed in 75% basalt, 25% bentonite, in the presence of solutions containing various amounts of NaCl, NaF, Na_2SO_4 and Na_2CO_3

MATERIALS/COMPONENTS

Steel as received (hot-rolled). T = 100 °C and 250 °C in pressure vessels. No oxygen.

METHODS OF DATA COLLECTION/ANALYSIS

Weight loss after 4 weeks. Analysis by the Plackett-Burman statistical method.

AMOUNT OF DATA

1 Table. Weight losses for various solution compositions and temperatures.

UNCERTAINTIES IN DATA

95% confidence level.

DEFICIENCIES/LIMITATIONS IN DATABASE

Not addressed.

KEYWORDS

3

experimental data, Plackett-Burman statistical design, weight change, laboratory, deionized, Cl, F^- , SO_4^{--} , CO_3^{--} , basalt, bentonite, high temperature, carbon steel, 1020 carbon steel, hot-rolled, chloride, basalt, bentonite

COMMENTS

It is an extremely limited test, so that its relevance is minor. Can be taken as indicating that groundwater composition is not too important under reducing conditions.

The conclusion "if resaturation of the repository occurs at a higher temperature (200 to 250°C) the effect on corrosion of low-carbon steel in the presence of the anions within the concentration range tested will be negligible" is not justified by the data. [page 6, paragraph 2]. Lack of statistical significance does not imply absence of a real effect.

<u>APPLICABILITY OF DATA TO LICENSING</u> [Ranking: key data (), supporting (X)]

(a) Relationship to Waste Package Performance Issues Already Identified

This document address BWIP ISTP issues 2.2.4, what are the corrosion modes of the waste package container and 2.2.4.3, how does the packing material affect the corrosion rates?

- (b) New Licensing Issues
- (c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

2

(a) Organization Producing Data:

Pacific Northwest Laboratory, Richland, Washington 99352

(b) Author(s), References, Reference Availability:

S. G. Pitman, "Slow-Strain-Rate Testing of 9%Cr, 1%Mo Wrought Steel and ASTM A27 Cast Steel in Hanford Grande Ronde Groundwater", SD-BWI-TS-008, August 1984.

DATE REVIEWED: 2/26/87; Revised 7/31/87

TYPE OF DATA

- 1. Scope: Experimental Results
- 2. Failure Mode: Environmental Induced Cracking; Stress Corrosion Cracking

MATERIALS/COMPONENTS

2 Candidate Container Alloys

9Cr-1-Mo Steel (ASTM A387 Gr. 9) (normalized at 900°C for 52 min, air cooled, tempered at 720°C)

ASTM A27 Cast Steel (tested as cast)

The specific compositions of the alloys are given in tables 1 and 2. The specimens were procured and fabricated according to MD-82-1, Rev. 0. The third candidate alloy was the subject of another report (see ref).

TEST CONDITIONS

Slow Strain Rate Tests

Standard Proc: MD-82-7, "Slow-Strain-Rate Studies, Unirradiated" Strain Rates: 1×10^{-4} , 1×10^{-6} and 2×10^{-7} /s Autoclave environment (150°C) Simulated Grande Ronde Groundwater Flowing (9 ml/h), Through crushed basalt then autoclave Deaerated with either pure Ar or Ar-20%O₂ Dissolved O₂ levels: Ar = <1ppm and Ar-20%O₂ = 8ppm O₂

METHODS OF DATA COLLECTION/ANALYSIS

Load and Displacement measurements

- Yield Strength
- Ultimate Tensile Strength

Direct Measurement of Sample

- Reduction in area
 - Strain to Failure

AMOUNT OF DATA

6 Tables:

2

- 1. Composition of 9Cr-1Mo Steel
- 2. Compostion of ASTM A27 Steel
- 3. Composition of Umtanum Flow Basalt
- 4. Composition of Hanford Grande Ronde (basalt) Groundwater
- 5. Results of SSR Tests on 9Cr-1Mo Steel at 150°C
- 6. Results of SSR Tests on ASTM A27 Steel at 150°C

5 Figures:

- 1. Reduction in Area (0 to 70%) vs. Displacement Rate $(10^{-7} \text{ to } 10^{-3})$ for 9Cr-1Mo Steel at 150°C in air and groundwater.
- 2. Load (0 to 4500 lbs.) vs. Displacement (0 to 0.5 in) for 9Cr-1Mo steel at 150°C in groundwater.
- 3. Fractography of 9Cr-1Mo steel tested at 1×10^{-6} in/s in groundwater.
- 4. Reduction in Area (0 to 70%) vs. Displacement Rate (le-7 to le-3) for A27 , Steel 150°C in air and groundwater.
- 5. SEM Fractography of A27 Steel tested at 150°C in groundwater showing ductile "MVC" fracture.

UNCERTAINTIES IN DATA

"The basalt rock and groundwater employed in these tests were the BWIP reference compositions at time of usage. Current (FY-1985) reference compositions are GR-4 groundwater and Cohassett flow basalt. Differences in composition between Cohassett and Umtanum basalt are slight and no difference in corrosion behavior are expected to result."

DEFICIENCIES/LIMITATIONS IN DATABASE

"It is recognized that the groundwater flow rate, oxygen concentration levels, specimen environment and imposed stresses may not be identical to those anticipated for the waste container in a repository. However, for the stated purpose of assessing relative susceptibility of different canister materials to environmental assisted cracking, the test conditions are thought to be sufficient."

KEYWORDS

experimental data, supporting data, microscopy (SEM), tensile testing, visual examination, slow strain rate, laboratory testing, basalt groundwater, high temperature, steel (A27 and A387), cast steel, elongation, tensile strength, yield strength, corrosion (SCC), hydrogen embrittlement

COMMENTS

1. The author assumes that hydrogen embrittlement and SCC are separate and distinctly different phenomena.

2. The author assumes that, since a two week pre-exposure had no effect on the SSR test ductility at 1×10^{-4} /s, the failure mechanism is SCC. (Presumably, what the authors means is that the mechanism of SCC was some mechanism other than hydrogen embrittlement.) However, no reduced ductility was found for tests conducted in the environment at this strain rate. If this strain rate was too fast to cause a reduction in the ductility in the environment, then why should it be slow enough to test the hydrogen embrittlement hypothesis. If hydrogen embrittlement is responsible for the observed crack propagation, then hydrogen diffusion could be the rate limiting process and it would not change its rate with the testing environment. There is insufficient evidence to conclude that hydrogen embrittlement is not responsible for the observed crack propagation.

3. The author concludes that SCC is not occurring based on the presence of dimples on the fracture surface (even though numerous secondary cracks and a reduced ductility were observed). Mechanisms have been proposed for hydrogen embrittlement and SCC which would be consistent with a dimple morphology. As a result, it is premature to conclude that there is not environmental contribution to crack propagation especially when a reduction in the ductility and secondary cracks are observed.

RELATED HLW REPORTS

S. G. Pitman, Environmental Testing of AISI 1020 Steel in Hanford Grande Ronde Groundwater, SD-BWI-TI-152, Pacific Northwest Lab., Richland, WA.

<u>APPLICABILITY OF DATA TO LICENSING</u> [Ranking: key data (), supporting data (x)

(a) Relationship to Waste Package Performance Issues Already Identified

This paper addresses BWIP ISTP issues 2.2.4.1, the corrosion rates for various corrosion modes of the waste package container, and 2.2.4.2, the effect of radiation on the corrosion behavior of the waste container.

- (b) New Licensing Issues
- (c) General Comments

3

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

È

(a) Organization Producing Data

Rockwell Hanford Operation

(b) Author (s), Reference, Reference Availability, Date

Shu-Chien Yung, Robert T. Toyooka and Tristram B. McCall, Thermal Analysis of Waste Package Preliminary Reliability Assessment, RHO-BW-SA-509P, presented at: Symposium on Radioactive Waste Management '86, Tucson, Arizona, March 2-6, 1986.

DATE REVIEWED: 2/5/87; Revised 6/30/87

TYPE OF DATA

A three-dimensional integrated model was used to simulate and predict thermal conditions for the BWIP waste package and its environment. Axial temperature gradients in the waste form and container are predicted. The predicted temperature histories of the waste package container are employed to assess the container lifetimes. Calculations are made from a theoretical model describing the time dependence of the temperature at various places in the repository.

MATERIALS/COMPONENTS

Consolidated spent fuel (CSF), Intact spent fuel assembly (SFA), West Valley High-Level Waste (WVHLW)

TEST CONDITIONS

The assumption of the model are:

- The repository and its environment are in a homogeneous basalt flow and extend vertically to the boundaries of the problems investigated.
- (2) All waste packages are simultaneously emplaced in the basalt repository.
- (3) All components of the waste package are made of isotropic and homogeneous materials.
- (4) Materials properties remain constant in each computer run.

(5) The heat of vaporization and condensation of groundwater in the host rock and convective transport of heat are not considered.

METHODS OF DATA COLLECTION/ANALYSIS

Calculation of temperatures as a function of time and position via the three-dimensional integrated model, Heating 5.

AMOUNT OF DATA

One Table and 10 Figures:

TABLE:

 Important Temperatures from HEATING 5 computation results -Design-allowable temperatures and peak calculated temperatures for consolidated spent fuel, spent fuel assembly and West Valley High Level Waste.

FIGURES:

- 1. Diagram Illustrating Waste Package Terminology.
- 2. Underground Repository Layout.
- 3. Integrated Three Dimensional Thermal Model Based on Assumption of Geometric Symmetry and Waste Package Simultaneous Emplacement.
- 4. Detailed XZ Plane Dimensions of the Integrated Three-Dimensional Model.
- 5. Temperature Histories in and Near Consolidated Spent Fuel Waste Package in the Nominal Case, Calculated by the Integrated Thermal Model, for 1-10000 years, in some points: waste (axial center), container (end), borehole (inner surface), basalt (1.6m from waste center), emplacement room center, temperature range: 50-300°C.
- Axial Temperature Profiles in Consolidated Spent Fuel Waste Form Midplane, 1, 5, 10, 25, 50 years after emplacement. For distance along the waste form of 0-4m in the temperature range of 100-300°C.
- 7. Axial Temperature Profiles in Container for Consolidated Spent Fuel Waste Form, 1, 5, 10, 25, 50 years after emplacement. For distances along the waste form of 0-4m and temperature range of 100-300°C.
- Temperature Profiles in the Near Field of the Consolidated Spent Fuel Waste Package, 1, 5, 10, 25, 50 years after emplacement. For distances from waste package center of 0-2m and temperature range of 50-250°C.

- 9. Isoterms in the XZ Plane through Consolidated Spent Fuel Waste Package Center, 1, 5, 25, 50 years after Emplacement. For distances from waste package center of 0-3m and temperature range of 100-200°C.
- 10. Far Field Temperature Profiles in YZ Plane through Center of Consolidated Spent Fuel Waste Package, 50, 100, 500, and 1000 years after emplacement. For depths of 0-2000m below ground surface and temperature range of 0-250°C.

UNCERTAINTIES IN DATA

The decay power and the thermal conductivity of the waste form and packing have been varied in their estimated ranges. It was determined that the conductivities of the waste form and the packing are the most influential parameters affecting the temperature profiles. The ranges considered in the study showed that a temperature difference of more than 70°C may result from the uncertainty of the conductivities.

DEFICIENCIES/LIMITATIONS IN DATABASE

Neither the heat of vaporization and condensation of ground water in the host rock nor the convective transport of heat were considered in the model utilized here.

KEY WORDS

model/metodology; waste form; physical properties; data analysis; three dimensional integrated model; thermal analysis simulation; air; basalt; commercial high level waste (CHLW); spent fuel; thermal profiles; thermal instability

COMMENTS

This report is only a theoretical evaluation without any experimental results to validate the results. The modeling approach that was taken is significant in that it simultaneously provides both the thermal fields of the waste package and its near-field and far-field environment.

The fact that the heat of vaporization and condensation of the groundwater in the host rock was not considered could be very important in case of significant groundwater flow through the repository site.

RELATED HLW REPORTS

BMI/ONWI-517 BMI/ONWI-612

<u>APPLICABILITY OF DATA TO LICENSING</u> [Ranking: key data (), supporting data (x)]

(a) Relationship to Waste Package Performance Issues Already Identified

This document addresses basalt ISTP issue No. 2.3.7 (how does the waste form design accomodate all potential waste package conditions) by calculating the thermal history of the waste package and repository area due to the waste form. Three different waste forms were studied.

- (b) New Licensing Issues
- (c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

(a) Organization Producing Data:

Corrosion Technology Section, Westinghouse Hanford Co., P. O. Box 1978, Richland, Washington 99352.

(b) Author(s), Reference, Reference Availability:

F. Brehm, J. M. Lutton, C. L. Rivera, H. P. Maffei, A. P. Bohringer, D. D. Paine and L. A. Pingel, "BWIP General Corrosion Studies, Summary Report of Activities in FY-1984", B047154, 1984.

DATE REVIEWED: 2/5/87; Revised 6/18/87

TYPE OF DATA

Experimental

MATERIALS/COMPONENTS

Test coupons of cast and wrought AISI 1020 steel, Fe-9Cr-1Mo steel, Cupronickel 90-10, weldments of these materials, OFHC copper and phosphorusdeoxidized copper. AISI 1020 steel with artificial pits. Ferrallium 255, Hastelloy C-276, Inconel 600 and Inconel 625 were obtained as alternate materials with higher corrosion resistance, and these will be used for future testing.

TEST CONDITIONS

An air-steam chamber (ovens with exposure boxes to operate at 150°C, 200°C, 250°C and 300°C) to simulate the environment of the preclosure period; post closure anoxic, high temperature environment (200°C, 1250 psig) simulated by static pressure vessels (Parr type) and autoclaves containing basaltbentonite packing, Grande Ronde #4 synthetic ground water and corrosion specimens. Anion effects (specific amounts of Cl, F, S04, or CO3 added to test solution at 100°C and 200°C) on 48 specimens of 9Cr-1M0-Fe, Packing material used in the tests was a 3:1 basalt:bentonite mixture.

METHODS OF DATA COLLECTION/ANALYSIS

Weight loss determinations; electrode potential measurements, pit count, pit diameter and pit area measurements

AMOUNT OF DATA

The eight figures are listed as follows.

- Fig. 1. Operator installing a four vessel rack in oven. Two other racks can be seen, one on each level. Negative 8401740-13CN
- Fig. 2. Air/Steam test with atmosphere chamber installed. The cover flange of the box has been removed to show the coupon racks in place. Negative 8401740-19CN
- Fig. 3. Autoclave flow diagram
- Fig. 4. Riffle sample splitter
- Fig. 5. BWIP General corrosion test specimen summary
- Fig. 6. Average corrosion rates (and standard deviations)
- Fig. 7. BWIP Corrosion test-100°, anion effect on corrosion of 9Cr-1Mo-Fe
- Fig. 8. BWIP Corrosion test-200°C, anion effect on corrosion of 9Cr-1Mo-Fe.
- There are three numbered tables which are;
- Table 1. Composition of actual and synthetic Grande Ronde 4 solutions,
- Table 2. Reagents required for preparation of one liter basic stock solution, and
- Table 3. Reagents for preparation of one liter of stock solution. There is one table with no number which lists "Compiled Data for Wrought AISI Steel Coupons"

UNCERTAINITIES IN DATA

Not addressed by authors.

DEFICIENCIES/LIMITATIONS IN DATABASE

Data collected and reported in the anionic effects tests do not give accurate analyses of corrosion rates due to limitations of the analytical balance which was used for weighing the specimens.

KEYWORDS

Planned work, experimental data, measurements, simulated groundwater, basalt, bentonite, high temperature, steel, cast and wrought 1020 and 1025 carbon steels, weld, corrosion, pitting, chloride, fluoride, sulfate, carbonate, preclosure, postclosure

COMMENTS

Equipment was wet up and tested for the conduct of three types of tests on candidate waste package materials and in some cases, their weldments. Candidate materials and alternative materials were obtained. These tests simulated repository environmental conditions before closure, after closure and a third test determined the effects of anionic species. Some pitting studies were conducted, but results were not conclusive. This type of simulated testing is valuable in producing data on the durability of materials under the environments of the repository before and after closure. These tests provide an indication of reactivity of these materials on a short term basis. Tests were conducted to check out the equipment and additional tests were reported to be in progress and results should be available by now. Additional electrochemical measurements should be carried out to supplement these data and to identify the corrosion processes.

RELATED HLW REPORTS

Westinghouse Hanford Company reports on corrosion studies in years preceding and following the period of this report.

<u>APPLICABILITY OF DATA TO LICENSING</u> [Ranking: key data (), supporting (X)]

(a) Relationship to Waste Package Performance Issues Already Identified

2.2.4 regarding potential corrosion failure modes for the waste package container, 2.2.4.1 dealing with corrosion rates, 2.2.4.3 dealing with effects of packing materials on corrosion

- (b) New Licensing Issues
- (c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

_ _ .

(a) Organization Producing Data

Pacific Northwest Laboratory

(b) Author(s), Reference, Reference Availability

Pitman, S. G., "Enviromechnical testing of AISI 1020 steel in Hanford Grande Ronde Groundwater", B023959, July 1983

DATE REVIEWED: 5/19/87; Revised 7/7/87

TYPE OF DATA

Experimental, environmentally assisted cracking

MATERIALS/COMPONENTS

1020 steel waste canister

TEST CONDITIONS

Hot-rolled wrught 1020 steel. Slow-strain-rate tests in air and Grande Ronde groundwater, 150°C, refreshed autoclave system

METHODS OF DATA COLLECTION/ANALYSIS

Slow-strain-rate tensile test with LT and TL orientations SEM observations

AMOUNT OF DATA

Tables listing compositions of 1020-steel test material, Umtatum flow basalt and Hanford Grande Ronde basalt groundwater

Table giving detailed results of 20 slow-strain-rate tests in $150^{\circ}C$ groundwater, $150^{\circ}C$ air and $20^{\circ}C$ air. Strain rates of 10^{-4} and $2 \ge 10^{-7}/s$, with 1020-steel samples tested on longitudinal and transverse orientations.

Three figures plotting data from Table (above): Figure 3 - Yield and ultimate strength (35 to 70 ksi) versus strain rate $(10^{-4} \text{ and } 2 \times 10^{-7}/\text{s})$ for 1020 steel (LT orientation) in air and groundwater.

Figure 4 - Similar to Figure 3. Reduction in area (45 to 65%) and elongation (25 to 30%) plotted as function of strain rate.

Figure 5 - Similar to Figure 4. Data for 1020 steel tested in TL orientation. 20 to 60% reduction in area and 15 to 25% elongation.

Optical micrograph of 1020 steel structure, 100x and 250x

Macrograph of slow-strain-rate specimen immediately after testing. Specimen strained to failure in 150°C groundwater at 2 x 10^{-7} /s; 21 day test.

Fifteen SEM photographs of fracture surfaces of slow strain rate test specimens. Includes illustration of pitting attack.

UNCERTAINTIES IN DATA

Author states that groundwater used in this study differs somewhat from composition now being considered as the reference Grande Ronde groundwater but is unlikely that differences affect environmentally assisted cracking kinetics. Specific groundwater composition differences not described in report.

DEFICIENCIES/LIMITATIONS IN DATABASE

Author states that definitive conclusions concerning structural barrier material selection and design are beyond scope of document.

KEYWORDS

experimental data, corrosion, simulated field, simulated groundwater, Cl, ambient temperature, high temperature, carbon steel, 1020 carbon steel, hot worked, slow strain rate, groundwater, elongation, tensile strength, yield strength, cracking (environmentally assisted)

COMMENTS

Author does not state thickness of hot rolled steel plate used to prepare test specimens. There are significant structural differences in hot rolled plate in the thicknesses required for canister construction as compared with thinner gauge plate. These structural differences may affect EAC test results. Data provide clear indication of environmentally assisted cracking for 1020 steel tested at low strain rate $(2 \times 10^{-7}/s)$ in Grande Ronde groundwater. Further studies are needed to relate conclusion to structural barrier material selection.

RELATED_HLW_REPORTS

۰**ه**. ۱

> <u>APPLICABILITY OF DATA TO LICENSING</u> [Ranking: key data (), supporting (X)]

(a) Relationship to Waste Package Performance Issues Already Identified

This paper addresses BWIP ISTP issue 2.2.4, what are the various corrosion modes for the waste package container?

- (b) New Licensing Issues
- (c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE:

(a) Organization Producing Data:

Hanford Engineering Development Laboratory, P. O. Box 1970, Richland, WA 99352

(b) Author(s), Reference, Reference Availability:

Einziger, R. E., "Test Plan for Long Term, Low-Temperature Oxidation of Spent Fuel, Series 1", HEDL-7560, June 1986.

DATE REVIEWED: 3/26/87

TYPE OF DATA

Plan for low-temperature oxidation of spent fuel. Oxidation rate to be determined from weight gain as a function of time at specified temperatures.

MATERIALS/COMPONENTS

Selected spent fuel from Turkey Point, air, water vapor, Ni/Cr crucibles.

TEST CONDITIONS

Spent fuel samples consisting of fragments sized from -10/+24 mesh and -24/+60 mesh will be oxidized in dry air, (dew point below -55 0C), and moist air, (dew point of 80°C), at temperatures of 175, 130 and 110°C. Samples of 10 grams will be contained in Ni/Cr crucibles and heated in an aluminum block dry bath. Test duration will be 2 years. Time intervals between periodic weighings of 100 to 400 hours will be adjusted as data become available in order to obtain optimal weight gains. Mesh sizes 10, 24, and 60 correspond to 1.7, 0.71, and 0.25 mm respectively. Dew points of -55 and 80°C correspond to water vapor partial pressures of less than 0.016 and 355 torr respectively.

METHODS OF DATA COLLECTION/ANALYSIS

Rate constants will be determined from weight gain data fit to models. Models assume grain boundary diffusion in early stages of oxidation followed by bulk diffusion. Post test samples will be examined by scanning electron microscopy (SEM) and x-ray diffraction (XRD) to obtain additional phase information.

AMOUNT OF DATA:

Tables:

- 1. Test Matrix.
- 2. Determination of D_g (grain boundary diffusion constant) from Thermogravimetric Analysis (TGA) data.
- 3. Time Ranges when Bulk Diffusion Becomes Controlling Oxidation Mechanism.
- 4. Turkey Point Fuel Available for Testing.
- 4. Thermal Conductivity of Uranium Oxides.

Figures:

- 1. Rate Constant for Spent Fuel Oxidation Determined by TGA Testing, Rate constant $(10^{-9} \text{ to } 10^{-3} \text{ cm}^2/\text{h})$ vs $10^4/\text{T}$ (19 to 26°K^{-1}).
- Weight Gain Projections for 10g Sample Based on Single Mechanism Oxidation. Weight gain (0 to 150 mg) vs time (0 to 20,000 h)
- Weight Gain Projections after Slow Grain Boundary Penetration. Weight gain (0 to 200 mg) vs time (0 to 20,000 h).
- 4. Oxidation of Spent Turkey Point PWR Fuel in Air at 200°C. $[1-(3 \text{ delta } 0/\text{M})^{1/3}]$ (0 to 0.20) vs t^{1/2} (0 to 15 h ^{1/2}).
- 5. Grain Boundary Diffusion of O_2 into Spent Fuel as a Function of Temperature Based on TGA data. Diffusion constant $(10^{-10} \text{ to } 10^{-7} \text{ cm}^2/\text{s}) \text{ vs } 10^4/\text{T K}^{-1}$ (20 to 26).
- Time to Complete the Grain Boundary Diffusion Stage as a Function of Fragment Radius at 175, 130, and 110°C. Radius of fragment (0 to 2 mm) vs t^{1/2} (0 to 140 hrs^{1/2}).
- 7. Cutting Diagram for Rods Used in Testing.
- 8. Relationship of Characterization and Test Rods in Assembly B17.
- Gamma Scans of Companion Rods from Assembly B17. Relative activity (0 to 100) vs distance from bottom fuel pellet (0 to 150 inches).
- 10. SEM Examination of Fuel Fragments from Rod F6.
- 11. Dry Bath Oxidation System.
- 12. Dry bath with Blocks, Thermocouples and Gas Lines.
- 13. Positioning of Crucible and Fuel Sample in the Aluminum Thermal Blocks.
- 14. Placement of Thermocouples in Dry Bath Diagnostic Testing.
- 15. Sample Crucible.

UNCERTAINTIES IN DATA

Not applicable.

DEFICIENCIES/LIMITATIONS_IN_DATABASE

If diffusion along the grain boundaries is slow with respect to the test duration, measurements of weight gain utilizing fuel fragments will not reflect the rate limiting bulk diffusion step but will represent a combination of grain boundary and bulk diffusion.

KEY WORDS

planned work, oxidation of spent fuel, supporting data, diffusion model, weight change, SEM, XRD, laboratory, air, water vapor, spent fuel(PWR reactor), high temperature, ambient pressure,

COMMENTS

Spent fuel is a potential waste form for isolation in a nuclear waste repository. Oxidation of spent fuel is expected to have an effect on both the rate of leaching and the concentration of specific radionuclides in the leachate. This report deals with oxidation of spent fuel in breached Zircaloy cladding. Defected cladding may allow the spent fuel to oxidize to a more leachable higher oxidation state before the fuel comes into contact with groundwater. A plan is described for obtaining oxidation data in dry or moist air at temperatures in the range of 110 to 175°C. From previous measurements, the authors believe that oxidation of spent fuel by air involves an initial oxidation mechanism of grain boundary diffusion followed by a bulk diffusion process into the individual grains. This is a plausible concept but the data presented in support of it is not very convincing because only a single data point falls on the line purported to represent grain boundary diffusion. It is possible that the deviant point thought to represent grain boundary diffusion is due to random error in the measurements. Although the oxidation of spent fuel will not take place within leaktight Zircaloy cladding, the lifetime of cladding under repository conditions is uncertain. The conditions for the oxidation study in dry or moist air are very different. The moist air water pressure would be 355 torr as compared to 0.016 torr for the dry-air measurements. This could lead to different rates of oxidation because at a total pressure of one atmosphere the partial pressure of O_2 would be lowered. Also interaction of radiation from the spent fuel with water vapor might lead to formation of different gaseous species which could change the oxidation mechanism. Knowledge of rate constants for both grain boundary and bulk diffusion could provide an estimate of the overall oxidation rate of the spent fuel if it was possible to determine the time period during which each diffusion process was important.

RELATED HLW REPORTS

Einziger, R. E., and Woodley, R. E., "Low Temperature Spent Fuel Oxidation under Tuff Repository Conditions", HEDL-SA-3271FP (1985). Proceedings of the Symposium on Waste Management, Tucson, Arizona, March 24-28 (1985).

Einziger, R. E., and Woodley, R. E., "Test Plan for Series 2 Thermogravimetric Analysis of Spent Fuel Oxidation," HEDL-7556, February (1986).

Einziger, R. E., and Woodley, R. E., "Evaluation of the Potential for Spent Fuel Oxidation under Tuff Repository Conditions," HEDL-7452, March (1985).

APPLICABILITY OF DATA TO LICENSING:

[Ranking: key data (), supporting data (X)

(a) Relationship to Waste Package Performance Issues Already Identified:

Related to issue 2.3.6.3 in the ISTP of the NWWSI Project. This issue concerns how the presence of defects alter the radionuclide retention capability of the waste form. Oxidation of the spent fuel through pinholes or other fissures is a potential failure mechanism.

- (b) New Licensing Issues
- (c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

(a) Organization Producing Data

Lawrence Livermore National Laboratory, Livermore, CA 94550

(b) Author(s), Reference, Reference Availability

V.M. Oversby, "Spent Fuel as a Waste Form--Data Needs to Allow Long Term Performance Assessment Under Repository Disposal Conditions", UCRL-94659, December 1986.

DATE REVIEWED: 5/4/87; Revised 7/27/87

TYPE OF DATA

(1) Scope of the Report

The report is an analysis and technical review of factors affecting radionuclide release from spent fuel under repository conditions. (See reports listed under <u>Related HLW Reports</u> for the original information and greater detail of the methods and results than are given in this report.) The paper presents an analysis [details of calculations not given in this report] of NRC and EPA regulations and from that analysis a ranking of the radionuclides in spent fuel is derived. Areas are suggested where more work is needed to support performance assessment calculations.

(2) Failure Mode or Phenomenon Studied

Dissolution of spent fuel and assembly components in high-level waste repositories.

MATERIALS/COMPONENTS

Radionuclides for which release rates are discussed include: ²⁴¹Am, ²⁴²Am, ²⁴²Am, ²⁴³Am, ¹⁴C, ²⁴²Cm, ²⁴⁵Cm, ²⁴⁶Cm, ¹³⁵Cs, ¹²⁹I, ⁹⁴Nb, ⁵⁹Ni, ⁶³Ni, ²³⁷Np, ²³⁹Np, ¹⁰⁷Pd, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴²Pu, ²²⁶Ra, ⁷⁹Se, ¹⁵¹Sm, ¹²⁶Sn, ⁸⁹Tc, ²³⁰Th, ²³³U, ²³⁴U, ²³⁵U, ²³⁶U, ²³⁸U, ⁹³Zr.

Besides the spend fuel itself, other radioactive elements found in components are mentioned, specifically the nickel in stainless steels, the zirconium in Zircaloy, and the carbon-14 activation product in the Zircaloy.

TEST CONDITIONS

(1) State of the Material being Tested

In the tests for which results are reviewed (see Reports HEDL-TME 85-22 and UCID-20926, Figures 1, 2, 3), the spent fuel used was PWR fuel of average burnup from the H.B. Robinson reactor. Bare fuel as well as fuel with the cladding hulls were included in the tests. Some Turkey Point bare fuel was also tested.

(2) Specimen Preparation

No information is given.

(3) Environment of the Material being Tested

The Nevada Nuclear Waste Storage Investigations (NNWSI) Series 2 dissolution test conditions were used (details describing these tests are not given in the report). The solution used for all cycles of the testing was J-13 well water, a dilute sodium bicarbonate groundwater.

METHODS OF DATA COLLECTION/ANALYSIS

The author discusses and compares the NRC rule (10 CFR 60) and Environmental Protection Agency standards (40 CFR 191) for HLW disposal in geologic repositories. The EPA standards set limits on the cumulative release of radionuclides to the accessible environment for 10,000 years. Release limits are set for individual radionuclides and for total activity released. For individual radionuclides, there must be a probability of less than 0.1 for releases to exceed one part in 100,000 of the inventory at 1000 years after repository closure and a probability of less than 0.001 for the releases to exceed 10 times those values. The NRC has rules to implement the EPA standards. Siting Guidelines (10 CFR 960) also require long range performance assessment of proposed repositories and a comparison of performance prior to specific site selection. The comparison is based on two calculations of system performance for 100,000 years. The first calculation uses a specified high value for release of radionuclides from the engineered barrier system, and the second uses a realistic sitespecific value. These calculations [not given in the report] require performance predictions for ten times longer than the NRC rule. The NRC rule requires that the release rate of any radionuclide from the engineered barrier system following the 300-1000 year containment period be less than one part in 100,000 of the inventory of that radionuclide present after 1000 years after closure. The most stringent control that must be demonstrated is one part in 10^5 of a radionuclide inventory at 1000 years after closure or one part in 10^8 of the total inventory present 1000 years after emplacement, whichever is greater. For most radionuclides the total inventory value is applicable.

The analysis of release rates indicate that americium and plutonium require the most reduction in release, that is, the greatest release rate control of the radionuclides. Suggested methods to accomplish this are lower waste form dissolution rates, demonstration of reduction in transportable species resulting from precipitation reactions, retardation of transport due to ion exchange or other sorption processes, or long groundwater travel times.

The results of NNWSI Series 2 dissolution tests of the effect of leaching solutions on the concentrations of radionuclides in stored fuel are quoted. Tests were run in cycles of approximately six months in the same solution. At the end of each cycle the solution and fuel samples were removed from the quartz tests vessel and transferred to a new vessel with fresh solution. Solution samples were taken periodically during a cycle. At the end of a cycle, the vessel was rinsed and acid stripped to recover any precipitated solution.

AMOUNT OF DATA

There are four tables, five figures, and seventeen references cited.

Table I - "Release rate control required by 10 CFR 60," lists ten radionuclides whose release must be controlled to 1 part in 100,000 of their own inventory at 1000 years after repository closure and, twentythree radionuclides for which the calculated release rate applies and the factors [for which the calculations are not given in this report] by which their release rates may exceed 1 part in 100,000 of their own inventory at 1000 years after repository closure.

Table II - "Release rate control based on chemical element," lists the control of release rate for 18 chemical elements based on the most stringent control required for any isotope of that element, in parts in 100,000 of the 1000 year post-closure inventory of that element.

Table III - "Comparison of NRC and EPA allowed releases assuming that no nuclide contributes more than 0.035 EPA units to the sum of release ratios. Comparison is made at the edge of the engineered barrier system," lists the ratios of NRC/EPA allowed releases for 17 elements.

Table IV - "Summary of release data for NNWSI Series 2 H.B. Robinson bare fuel samples, Cycles 1 and 2, conducted at ambient hot cell temperature," lists Total Release in parts in 10^5 , and Percent in Solution for nine elements.

Figure 1 - "Concentration of uranium in solution for the four cycles of the NNWSI Series 2 dissolution tests. Test run at ambient hot cell temperature in quartz vessel using bare fuel with split cladding hulls present in the test." Data are plotted for 0 to 5 ppm uranium for tests of up to 240 days duration for four different cycles of tests.

Figure 2 - "Activity in solution for cesium-137, NNWSI Series 2 tests, cycles 2, 3, and 4. Release for cycle 1 was about 40 times higher than for cycle 2." Data for tests of up to 240 days duration ranging from 0 to 3 microcuries/ml are plotted.

Figure 3 - "Activity of technetium in solution, NNWSI Series 2 tests." Data for four different cycles for tests of up to 240 days duration are plotted for pCi/ml values ranging from 0 to 500.

Figure 4 - "Correlation of fission gas release and rapid release fraction of cesium from CANDU fuel." Measured and calculated values for stable xenon release ranging from 0.01 to 10.0 percent are plotted versus ¹³⁷Cs release values ranging from 0.01 to 10.0 percent.

Figure 5 - "Uranium concentrations in unfiltered solutions for NNWSI Series 3 tests and for the H.B. Robinson Series 2 bare fuel sample. See text for symbol explanation." Concentration in micrograms per milliliter for seven solutions is plotted against test durations up to 240 days.

UNCERTAINTIES IN DATA

The review discusses several factors for which available data are inadequate to characterize radionuclide release from waste-form components. These factors are described in three categories:

- (1) Effects of sample preparation,
- (2) Sample storage conditions,
- (3) Experiment design variables.

See the next section for details of the deficiencies in the database.

DEFICIENCIES/LIMITATIONS IN DATABASE

The author identifies six areas where new data are most needed to judge repository license application:

- (1) the effect of reactor type and burnup on dissolution properties of spent fuel; fuel dissolution probably will not depend heavily on these parameters, but population variability must be addressed in licensing arguments.
- (2) dissolution studies on stainless steel clad fuel; part of the existing inventory of spent fuel, and fuel in use, is clad in stainless steel and no dissolution data exist.
- (3) dissolution studies of oxidized spent fuel; the effect of oxidation state on dissolution rate and solubility is needed to assess the effects in the repository environment and the effects of air storage of test specimens.
- (4) dissolution studies using assembly components; there are no data for the rate of radionuclide release from such components in aqueous solutions.
- (5) inventory and release characteristics of carbon-14; data are needed on both release in air and release into aqueous solutions of carbon-14.
- (6) thermodynamic properties of solids that might limit radionuclide solubility; silicate minerals may limit the solubility of uranium and affect the dissolution properties of spent fuel. Data for uranium silicates are sparse to nonexistent as well as data for compounds thought to limit solubility of actinides. Modelling of long term behavior of geochemical systems depends on good thermodynamic data for the relevant phases.

KEY WORDS

data analysis; literature review; leaching solution analysis; simulated field; air; J-13 water; ambient temperature; stainless steel; zirconium base; spent fuel (PWR reactor); spent fuel (BWR reactor); ²⁴¹Am, ²⁴²Am, ²⁴²Am, ²⁴³Am, ¹⁴C, ²⁴²Cm, ²⁴⁵Cm, ²⁴⁶Cm, ¹³⁵Cs, ¹²⁹I, ⁹⁴Nb, ⁵⁹Ni, ⁶³Ni, ²³⁷Np, ²³⁹Np, ¹⁰⁷Pd, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴²Pu, ²²⁶Ra, ⁷⁹Se, ¹⁵¹Sm, ¹²⁶Sn, ⁹⁹Tc, ²³⁰Th, ²³³U, ²³⁴U, ²³⁵U, ²³⁶U, ²³⁸U, ⁹³Zr.

COMMENTS

A comprehensive review of spent fuel radionuclide release factors and data are presented with identification of needs for further study. No new data are included. See the section "Deficiencies/Limitations in Database" for the data needed to provide an adequate database for characterizing radionuclide release from waste-form components.

RELATED HLW REPORTS

UCRL-94222 HEDL-TME 85-22 UCID-20926 UCRL-90855 UCRL-94708 PNL-5109 UCRL-94633 HEDL-TME 84-30 See also 40 CFR Part 191, Federal Register vol. 50, No. 182 (1985); 10 CFR Part 60, U.S. Government Printing Office (1983); 10 CFR Part 960, Federal Register vol. 49, No. 236 (1984).

APPLICABILITY OF DATA TO LICENSING

[Ranking: key data (), supporting data (x)]

(a) Relationship to Waste Package Performance Issues Already Identified

2.3 When, how, and at what rate will radionuclides be released from the waste form?

(b) New Licensing Issues

(c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

. .

(a) Organization Producing Data:

Swanson Engineering Associates Corporation, McMurray, PA

(b) Author(s), References, Reference Availability:

R. H. Mallett, "Buckling Design Criteria for Waste Package Disposal Containers in Mined Salt Repositories", BMI/ONWI-597, December 1986

DATE REVIEWED: 6/16/1987

TYPE OF DATA

- Scope: This document is a survey of the literature on buckling analysis and experiments; no new theoretical development or buckling experiments were conducted. The purpose of this work was to provide guidance in establishing a waste-container design criteria to ensure protection against buckling of the container
- 2. Failure Mode: Elastic, elastic-plastic and plastic buckling

MATERIALS/COMPONENTS

Waste disposal container of low carbon steel

No experiments were performed; however, the survey focuses on low-carbon steels and the properties of these alloys.

TEST CONDITIONS

This document is a survey of the literature on buckling analysis and experiments; no new tests or experiments were performed for this document.

METHODS OF DATA COLLECTION/ANALYSIS

This document is a survey of the literature on buckling analysis and experiments. The results of buckling experiments are compared to the predictions of different analytical models. No new experiments or analyses are presented.

AMOUNT OF DATA

12 Tables:

3-1. Summary of Waste Package Design Features (Container and Canister length, diameter and thickness also borehole information)

- 3-2. Summary of Waste Package Performance Parameters (temperatures, heat loads, corrosion penetrations and corrosion allowances)
- 3-3. Comparison of Low Carbon Steel Specifications (Chemistry and physical properties of container material, AISI 1018 and ASTM 216 steels)
- 3-4. ASME Boiler and Pressure Vessel Code Design Stress Intensity Values (Nominal composition of steel, yield strength, ultimate tensile strength and critical stress intensity for metal at different temperatures)
- 4-1. Reference Disposal Container Length Parameters
- 4-2. Reference Disposal Container Thickness Parameters
- 4-3. Reference Disposal Container Thick-Wall Stress
- 4-4. Reference Disposal Container Ovality Effect
- 4-5. Reference Disposal Container Buckling
- 4-6. Minimum (R/t) for Elastic Buckling and Maximum (Et/E) for Elastic-Plastic Buckling
- 4-7. Recent Cylinder Buckling Results
- 4-8. Small Cylinder Buckling Results
- 22 Figures:
- 3-1. Conceptual Waste Disposal Container Design (diagram)
- 3-2. ASME Code Buckling Chart for Factor A (Length to Outside Diameter ratio) (Factor A)
- 3-3. ASME Code Buckling Chart for Factor B (10-5 to 100 Factor A) (2,500 to 25,000 Factor B)
- 3-4. Creep Behavior for Salt (Time, 0-1.6x10-7 seconds) (axial strain, 0-10%)
- 3-5. Defense High-Level Waste Package Performance (0-150°C temperature, 15-20 MPa pressure and 0-1.0 cm. corrosion penetration) (0.1-1000 years after emplacement)
- 3-6. Commercial High-Level Waste Package Performance (0-150°C temperature, 15-20 MPa pressure and 0-1.0 cm. corrosion penetration) (0.1-1000 years after emplacement)

- 3-7. Consolidated Spent Fuel (12 Pressurized-Water Reactor Assemblies) Waste Package Performance (0-150°C temperature, 15-20 MPa pressure and 0-1.0 cm. corrosion penetration) (0.1-1000 years after emplacement)
- 4-1. Typical Stress Versus Strain Curve (0-0.10 in/in strain) (0-50 ksi stress)
- 4-2. Typical Tangent Modulus Curve (15-50 ksi stress) (0-5x10-6 MPa Tangent Modulus)
- 4-3. Compilation of Cylinder Buckling Results (1-1000 in/in Radius to thickness ratio) (0-1.4 collapse pressure to theoretical collapse pressure ratio)
- 4-4. Steel Capsules After Pressure Testing

...

- 4-5. Correlation of Buckling Analysis and Test (0-15 radius to thickness ratio) (0-20 ksi collapse pressure)
- 4-6. Collapse Mechanism Diagrams (a) (0-0.1 logarithmic strain) (0-600 MPa true stress), (b) (-0.06-0 circumferential strain)-(0-0.5 pressure/yield strength ratio), (c) (-1.0-1.0 load parameter) (0-0.8 pressure to yield strength ratio) and (d) (-1.0-1.0 load parameter) (0-0.6 pressure to yield strength ratio)
- 4-7. Bifurcation Pressure Versus Thickness to Mean Radius Ratio (0-0.5 wall thickness to radius ratio) (0-2.0 bifurcation pressure to yield strength ratio)
- 4-8. Curves of Pressure Versus Displacement on the External Surface (0-0.02 displacement/radius ratio) (0-1.0 pressure to critical pressure ratio)
- 4-9. Atomic Energy of Canada Limited Prototype Waste Package Container Collapse Shape (diagram)
- 4-10. Atomic Energy of Canada Limited Prototype Waste Package Container Collapse Shape (photograph)
- 4-11. Atomic Energy of Canada Limited Prototype Waste Package Container Behavior (+400 to -2000 5m/m strain) (0-18.96 MPa pressure)
- 4-12. Stress Versus Strain Curve for Atomic Energy of Canada Limited Test Specimen (0-20000 5m/m strain) (0-413.7 MPa stress)
- 4-13. Tangent Modulus Curve for Atomic Energy of Canada Limited Test Specimen (50-250 MPa) (0-0.150x106 MPa tangent modulus)
- 5-1. BS5500 Buckling Design Curves (0-12 K) (0-0.7 k)

7-1. Selected ASME Code Provisions for Plastic Analysis (strain or displacement, no units) (load, no units)

UNCERTAINTIES IN DATA

The buckling analyses neglect the effects of transient pressure, seismic loading, shear loading and corrosion other than uniform. Also, interactions such as creep, radiation-induced creep, aging of the metal (thermal and radiation induced), radiation-induced corrosion and stress corrosion are neglected. The properties of the material are assumed uniform and inhomogeneities such as welds, weld heat affected zones, casting defects (inclusion and voids) are neglected.

DEFICIENCIES/LIMITATIONS IN DATABASE

The waste disposal container can be treated in a buckling analysis as a long cylinder. Neglect of the support provided by the end closure is conservative and will not significantly alter the results of the analysis. The waste disposal container must be analyzed as a thick wall cylinder. The use of average stresses in the basic buckling equations is an approximation which must be verified by testing. The collapse pressure of a thick walled cylinder is strongly dependent on the work hardening characteristics of the material. Imperfections in the container will increase the ovality of the container reducing the collapse pressure. However, for practical imperfections the reduction in the collapse pressure should not be significant. The cylinder collapse test results reviewed indicate that buckling of a thick wall cylinder does not result in loss of wall continuity. That is, buckling of a waste disposal container onto the waste form may not result in leaking. However, the tensile stresses resulting from buckling may lead to failure by other mechanisms. As a result, after the retrieval period, buckling of the container does not by itself constitute failure. The basic equations for buckling overestimate the critical buckling pressure for thin wall cylinders and overestimate the critical pressure for thick wall cylinders. As a result, these equations are conservative estimates of the critical pressure. The experimental results reviewed in this report indicated that finite-element analyses frequently result in predictions for the critical collapse pressure, which are no better than the predictions of the simple relationships. However, a finite element analysis can predict the critical pressure within 3 percent. The simple buckling equations underestimate the critical collapse pressure and the use of a load factor (defined as the ratio of the critical collapse pressure to the maximum expected service pressure) of 1.5 or 3, as is used in external pressure buckling design standards, should be a conservative design criteria. However, it is assumed that the geometry, material and applied loading are specified with conservatism appropriate to their uncertainty.

KEYWORDS

literature review, design, salt, steel, collapse load tests, waste form (CHLW, DHLW and spent fuel) modulus of elasticity, tangent modulus, yield

strength, tensile strength, stress, strain, ovality, buckling, elastic buckling, plastic buckling, and elastic-plastic buckling

COMMENTS

This is not a critical review.

RELATED HLW REPORTS

American Society of Mechanical Engineers, 1983. ASME Boiler and Pressure Vessel Code, Nuclear Power Plant Components, ASME, New York. Bathe, K. J., 1976. ADINA - A Finite Element Program for Automatic Dynamic Incremental Nonlinear Analysis, Report 82448-1, Acoustics and Vibration Laboratory, Mechanical Engineering Department, Massachusetts Institute of Technology, Cambridge MA. Crosthwaithe, J. L., J. N. Barrie and K. Nuttall, 1982. Design, Fabrication and Testing of a Prototype Stressed-Shell Fuel Isolation Container, Report AECL-6823, Atomic Energy of Canada Limited, Pinawa, Manitoba, Canada DeSalvo, G. J., and H. Becker, 1957. Handbook of Structural Stability, Part III - Buckling of Curved Plates and Shells, Report NACA-TN-3783, National Advisory Committee for Aeronautics, Washington DC. Hibbitt, Karlsson, 1981. ABAQUS Computer Program Manuals, Volumes 1-4, Sorenson, Inc., Providence, RI. Huang, N. C. and P. D. Pattillo, 1982. "Collapse of Oil Well Casing", J. Pressure Vessel Technology, 104m, pp. 36-41. Lockheed Missiles and Space Co., Inc. 1974. BOSOR5, A Computer Program for Buckling of Elestic-Plastic Complex Shells of Revolution Including Large Deflection and Creep, Report LMSC-D407166, Lockheed Missiles and Space Company, Inc., Sunnyvale, CA. MARC Analysis Corporation and control Data Corporation, 1971. MARC-CDC Nonlinear Finite Element Analysis Program, MARC Analysis Corporation and Control Data Corporation, Minneapolis, MN. Timoshenko, S. P. and Gere, J. M., 1961. Theory of Elastic Stability, Second Edition, McGraw Hill, New York.

<u>APPLICABILITY OF DATA TO LICENSING</u> [Ranking: Key Data (), Supporting Data (x)]

- (a) Relationship to Waste Package Performance Issues Already Identified The information in this report is pertains to ISTP issue 2.2.3 "What are the possible mechanical failure modes for the waste package container".
- (b) New Licensing Issues
- (c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE:

(a) Organization Producing Data:

Brookhaven National Laboratory, Upton, New York 11973

(b) Author(s), Reference, Reference Availability:

Levy, Paul W., "Radiation Damage Studies on Natural Rock Salt from Various Geological Localities of Interest to the Radioactive Waste Disposal Program," Nuclear Technology, <u>60</u> 231-243, (1983).

DATE REVIEWED: 4/27/87

TYPE OF DATA

Radiation damage of rock salt by 1.5 MeV electron beam which is identical to damage produced by gamma radiation. Emphasis is on sodium colloid formation in the rock salt.

MATERIALS/COMPONENTS

Fourteen natural rock salt samples were irradiated by a 1.5 MeV electron beam.

TEST CONDITIONS

Samples are irradiated at constant temperature and extent of radiation damage is determined by optical measurements.

METHODS OF DATA COLLECTION/ANALYSIS

Extent of radiation damage is determined by optical absorption methods. Measurements are made as a function of temperature, dose, and strain in the crystals.

AMOUNT OF DATA:

Figures:

- 1. Appearance of rock salt as function of distance from spent fuel element in the Radioactive Waste Disposal Demonstration Project, Lyons Kansas.
- 2. Schematic of test radiation equipment.
- 3. Typical optical absorption and luminesence data.
- Colloid concentration versus irradiation time curves as a function of temperature. Absorption Coefficient (0 to 50 cm⁻¹) vs Irradiation Temperature (100 to 300°C) vs Irradiation Time (0 to 10000 s).
- 5. Colloid formation in 14 natural rock salt samples irradiated at 150°C at a dose rate of 1.2x10⁸ rd/h. Colloid Band Area (.1 to 50 arbitrary) vs Time (1 to 10⁴ s³.

- 6. Colloid growth curves for natural rock salt from WIPP site recorded using strained crystals. Absorption at 2.14 eV, (0 to 50 cm⁻¹ vs irradiation time (0 to 9x10³s) for 11%, 3.9% and undeformed crystals at 150°C, 120 Mrd/h)
- 7. Colloid formation in natural rock salt at an irradiation temperature of 150°C and dose rates of 30, 60 and 120 Mrd/h Absorption Coefficient (0 to 40 cm⁻¹) vs dose, (0 to 200 Mrad).
- 8. A black and white version of a colored photomicrograph of irradiated rock salt showing regions of F centers and colloidal sodium metal.

Tables:

- Constants for 14 specimens for the equation, mole fraction NaCl converted to colloidal sodium = Ctⁿ where C and n are constants and t is the time in seconds.
- 2. Impurities in colloid-rich and colloid-free areas in irradiated natural rock salt.
- 3. Percent sodium metal colloid expected in rock salt at canister interfaces for 10^{10} and $2x10^{10}$ rd, (about 50 to 400 y dose rate at $2x10^4$ rd/h).

UNCERTAINTIES IN DATA

Not dealt with.

DEFICIENCIES/LIMITATIONS IN DATABASE

The author states that because of the differences in dose rates, irradiation times, and rapidly applied stresses in laboratory experiments in contrast to anticipated dose rates, irradiation times and stresses under repository conditions, an accurate estimate of the total amount of radiation induced colloid and its spatial distribution requires appreciably more data than is presently available. To extrapolate from laboratory to repository conditions requires a better understanding of the radiationdamage formation kinetics in rock salt.

KEY WORDS

brine, chlorine, colloidal sodium in SRP, experimental data, gamma radiation field, high temperature, laboratory, radiation damage, radiation damage of rock salt

<u>COMMENTS</u>

This report presents experimental work on radiation induced sodium metal colloid formation in rock salt. The technique involves bombardment of rock salt with a high energy (1.5 MeV) electron beam which is equivalent to the effect produced by gamma ray recoil electrons. This work is of importance to salt repositories because of the drastic change in pH which could take place in the repository environment if chlorine is lost from the rock salt and at some later time, large amounts of sodium colloid react with brine. The experimental data show that the induction period for colloid formation is shortened with strained samples, increases on a unit dose basis as the dose rate decreases, and appears to be affected by the salt impurity level, being suppressed in regions of crystals containing about 1% calcium and sulfur. The colloid formation rate is low or negligible below irradiation temperatures of 100 to 115, increases to a broad maximum at 150 to 175 then decreases to a negligible level at 275 to 300°C. Using the C(dose)ⁿ relationship to estimate the colloid formed in actual repositories indicates that in 50 to 400 years, a dose of 10¹⁰ rad will convert .1 and 10%, and $2x10^{10}$ rad will convert between 1 and 50% of the salt to colloidal sodium. In the laboratory cleaved or broken irradiated samples emit the odor of chlorine. Obviously, the fate of chlorine in the irradiated rock salt is very important. The authors plan to attempt to detect the escape of chlorine from irradiated samples using a mass spectrometer detector. Whether chlorine escapes from the rock salt or can limit the sodium colloid buildup by some type of recombination reaction is an important problem for the salt repository.

RELATED HLW REPORTS

Levy, P. W. and Kierstad, J. A., "Very Rough Preliminary Estimate of the Colloidal Sodium Induced in Rock Salt by Radioactive Waste Canister Radiation"., Mat. Res. Soc. Symp. Proc., Elsevier Science Publ. Co., Vol 26 (1984) Pg 727-734.

<u>APPLICABILITY OF DATA TO LICENSING</u>: [Ranking: key data (), supporting data (X)

- (a) Relationship to Waste Package Performance Issues Already Identified
- (b) New Licensing Issues
- (c) General Comments

None of the ISTP issues previously identified appear to specifically relate to the interaction of brine with colloidal sodium in rock salt. Such an interaction could lead to a large change in pH which would drastically change the local waste canister environment.

NBS Review of Technical Reports on the High Level Package for Nuclear Waste Storage

DATA SOURCE

(a) Organization Producing Data:

Office of Nuclear Waste Isolation, Battelle Memorial Institute, 505 King Ave., Columbus, OH 43201-2693

(b) Author(s), References, Reference Availability:

D. E. Clark, "ERG Review of the SRP Salt Irradiation Effects Program", BMI/ONWI-626, November, 1986.

DATE_REVIEWED: 6/19/87

TYPE OF DATA

- 1. Scope: This document is a report of the August 1985 meeting of the engineering review group (ERG) where the salt repository project (SRP) salt irradiation effects program was reviewed; no new tests or experiments were performed for this document and no data is presented in this document. This review is the seventh in the series of regular ERG reviews conducted for ONWI. For this review, the ERG reviewed the work on irradiation effects at Pacific Northwest Laboratory and Brookhaven National Laboratory which was supported by the SRP and completed at the time of the review (see "Pertinent Documents").
- 2. Failure Mode: For a salt repository, the concern is that radiation induced changes in the chemical and physical properties of the nearfield environment will impact waste package performance with respect to any of the potential failure modes.

MATERIALS/COMPONENTS

The experiments reviewed by the ERG were conducted on single crystals of pure NaCl for the purpose of modeling the behavior of the bedded salt environment.

TEST CONDITIONS

This document is a report of the 1985 ERG meeting where the SRP salt irradiation effects program was reviewed; no new tests or experiments were performed for this document. Specific test conditions vary for each experiment and are not covered in this document (see HLW RELATED REPORTS).

METHODS OF DATA COLLECTION/ANALYSIS

This document is a report of the 1985 ERG meeting where the SRP salt irradiation effects program was reviewed; the methods of data collection and analysis are not covered in this document (see HLW RELATED REPORTS).

AMOUNT OF DATA

No new tests or experiments were performed for this document and no data is presented (see HLW RELATED REPORTS).

UNCERTAINTIES IN DATA

The ERG identified uncertainties in the salt irradiation effects database and made recommendations on action which should remove these uncertainties. ONWI's response to each of these items is given in the section of this review <u>DEFICIENCIES/LIMITATIONS IN DATABASE</u>. The uncertainties in the database identified by the ERG are:

- 1. Experiments on pure single crystals are not relevant to the bedded salt environment and the extent of the damage to the salt structure itself is of little interest.
- 2. Repository conditions and designs need to be finalized so that testing in representative conditions can begin.
- 3. Long-term and low-dose-rate tests need to begin now so that the lowdose-rate hypothesis of Levy et al., 1984, can be tested.
- 4. Investigations need to be integrated.
- 5. The significance of hydrogen production by irradiation of the brine, oxidation of radiation induced colloidal sodium and oxidation of the steel overpack should be evaluated.
- 6. Radiolysis will promote oxidizing conditions and corrosion of the steel overpack will promote reducing conditions. The relative significance of these two need to be evaluated.
- 7. The effects of various reactions on the pH of the environment need to be assessed using the results of ongoing projects.
- 8. Factors contributing to waste package degradation appear minor based on current design assumptions and knowledge. However, these factors need to be quantified to the extent possible and unanticipated factors identified.
- 9. To determine the chemical environment at the waste package as a function of time, the SRP needs to (1) place bounds on the colloidal sodium and chlorine production rates, (2) determine the mobility of Cl_2 and H_2 , (3) identify sources of volatile gases in natural salt, (4) compute H_2 fugacities and (5) integrate radiation, corrosion and brine migration models.

- 10. Calculations of the chlorine release at low dose rates need to be performed to indicate whether or not the pH will change significantly and alter the corrosion behavior.
- 11. Chlorine release rate calculations need to be integrated with brine migration studies.
- 12. Calculations should be made based on available data to determine the potential impact of colloid formation on brine chemistry.
- 13. Modeling of coupled subsystems will asses the importance of individual processes.
- 14. Calculations need to be carried out to indicate whether or not the dose-rate dependence of Levy et al. is of major engineering significance.
- 15. The effect of higher pH around the waste package needs to be assessed.
- 16. The effect of chlorine release on corrosion needs to be assessed.
- 17. A detailed calculation of the effects of stored energy are needed.
- 18. The amount of colloidal sodium present does not appear significant.
- 19. To properly assess the effects of irradiation, brine chemistry and corrosion, a suite of realistic scenarios must be invoked and evaluated.
- 20. Experiments on the mechanical properties of irradiated salt are needed.
- 21. Compare Levy's F-center formation rate data to those measured by others and available in the literature.
- 22. Conduct experiments which compare colloid concentration data to TEM examinations.
- 23. The effect of corrosion and hydrogen embrittlement on the waste package cannot be ascertained at this time and an ERG meeting should be called with experts on steel corrosion and embrittlement present.

DEFICIENCIES/LIMITATIONS IN DATABASE

. .

ONWI responded to each point raised by the ERG and identified the action being taken to eliminate the deficiencies in the salt irradiation effects database which are responsible for these uncertainties. ONWI's response to each point can be summarized as:

- 1. The testing has been limited to single crystals because of experimental constraints. However, experiments on aggregates have begun which measure the chlorine release. These experiments will be correlated with the measurements of colloidal sodium. In the future, small-angle neutron scattering may be employed to determine colloidal sodium.
- 2. ONWI agrees with the need to finalize site selection and repository design.
- 3. While there is potential value in carrying out long-term testing, there is not a pressing need to start them in the near future.
- 4. ONWI agrees and integration will be taken into account in future plans.
- 5. ONWI agrees and these factors will be considered in future package analyses.
- 6. Future testing should resolve this matter.
- 7. ONWI agrees. The work of Pederson addresses this subject in part (see pertinent documents section).
- 8. The SRP waste package testing program is directed towards providing a quantification of these and other factors.
- 9. Current testing and future modeling efforts will address these issues.
- 10. ONWI agrees. Currently, data are not available; however, results should be available in FY 86.
- 11. ONWI agrees that, to the extent possible, these calculations should be integrated.
- 12. ONWI does not anticipate any significant problems of this nature.
- 13. ONWI agrees and has structured its program accordingly.
- 14. These calculations have been performed and demonstrate that without a detailed knowledge of the temperature and transport, reliable estimate cannot be obtained.
- 15. While changing the pH may alter the reaction rate, the total extent of corrosion is limited by stoichemetry and the quantity of brine reaching the container.
- 16. This will be included in more detailed analyses of the expected waste package environment.
- 17. The effects of stored energy do not appear significant.

- 18. ONWI agrees that this is not a significant amount and ONWI feels that this is still a conservatively high estimate. More precise calculations will be carried out in the detailed performance assessment.
- 19. ONWI agrees.

. .

- 20. Some experiments on the mechanical properties of salt are planned.
- 21. So far as known to ONWI, there are no other F-center formation rate measurements to compare to Levy's.
- 22. TEM provides only qualitative information and cannot be used to obtain concentrations.
- 23. ONWI agrees with this importance of this meeting and has scheduled a meeting for the fourth quarter of FY 86.

KEYWORDS

review, gamma radiation, chlorine, hydrogen, radiolysis, colloid, salt, corrosion (uniform or general), hydrogen embrittlement, radiation effects

COMMENTS

The report reviewed here (BMI/ONWI-626) is a report covering a meeting of the ERG where the SRP salt irradiation effects program was reviewed. This report does not present detailed descriptions of the experimental procedures nor is any data formally presented. As a result, these factors cannot be critically reviewed here. In their review of the SRP salt irradiation effects program, the ERG reach two very important conclusions about the overall program. First, they point out that radiation induced changes in the structure and properties of the crystalline salt are not likely to significantly influence container performance for the expected repository conditions. The ERG point out that experiments can be conducted to test this conclusion. Second, the ERG concluded that while the experiments were well designed, the program was structured to address fundamental scientific questions and ignored the applied questions of interest to the program. That is, the program is designed to address fundamental questions about radiation damage in ionic solids while issues concerning container integrity and potential failure modes are ignored.

RELATED HLW REPORTS

Bergsma, J., R. B. Helmholdt, and R. J. Heijboer, 1985. "Radiation Dose Deposition and Colloid Formation in a Rock Salt Waste Repository," Nuclear Technology, Vol. 71, pp. 597-607.

DOE, see U.S. Department of Energy. Hobbs, L. W., 1973.

"Transmission Electron Microscopy of Defects in Alkali Halide Crystals," Journal de Physique (Paris), Colloque C9-227. Hobbs, L. W., 1975. "Transmission Electron Microscopy of Extended Defects in Alkali Halide Crystals," Surface and Defect Properties of Solids, Vol. 4, The Chemical Society, London, UK. p. 152. Jockwer, N., 1984.

. .

,

"Laboratory Investigations on Radiolysis Effects on Rock Salt With Regard to the Disposal of High-Level Radioactive Wastes," in G. L. McVay, ed., Scientific Basis for Nuclear Waste Management VII, Proceedings of a Materials Research Society Symposium, Vol. 26, Elsevier Science Publishing Company, New York, NY, pp. 17-23. Levy, P. W., 1983.

"Radiation Damage Studies on Natural Rock Salt from Various Geological Localities of Interest to the Radioactive Waste Disposal Program. " Nuclear Technology, Vol. 60, pp. 231-243. Levy, P. W., and J. A. Kierstead, 1984.

"Very Rough Preliminary Estimate of the Colloidal Sodium Induced in Rock Salt by Radioactive Waste Canister Radiation," in G. L. Mcvay, ed., Scientific Basis For Nuclear Waste Management VII, Proceedings of a Materials Research Society Symposium, Vol. 26, Elsevier Science Publishing Company, New York, NY, pp. 727-734.

Levy, P. W., J. M. Loman, and J. A. Kierstead, 1984. "Radiation Induced F-Center and Colloid Formation in Synthetic NaCl and Natural Rock Salt: Applications to Radioactive Waste Repositories," Nuclear Instruments and Methods in Physics Research B1, pp. 549-556. Levy, P. W., J. M. Loman, K. J. Swyler, and R. W. Klaffky, 1981.

"Radiation Damage Studies on Synthetic NaCl Crystals and Natural Rock Salt for Radioactive Waste Disposal Applications." On P. L. Hofman, ed., The Technology of High-Level Nuclear Waste Disposal, DOE/TIC-4621, Vol. 1, Technical Information Center, U.S. Department of Energy, Oak Ridge, TN. Panno, S. V., and P. Soo, 1984.

"Potential Effects of Gamma Irradation on the Chemistry and Alkalinity of Brine in High-Level Nuclear Waste Repositories in Rock Salt," Nuclear Technology, Vol. 67, pp. 268-281. Pederson, L. R., 1985.

"Chemical Implications of Heat and Radiation Damage to Rock Salt," in C. M. Jantzen, J. A. Stone, and R. C. Ewing, eds., Scientific Basis for Nuclear Waste Management VII, Proceedings of a Materials Research Society Symposium, Vol. 44, Materials Research Society, Pittsburgh, PA, pp. 701-708. U.S. Department of Energy, 1985a.

Mission Plan for the Civilian Radioactive Waste Management Program, DOE/RW-0005, Vols. I and II, Office of Civilian Radioactive Waste Management, Washington, DC. U.S. Department of Energy, 1985b.

Salt Repository Project Technical Progress Report for the Quarter 1 January-31 March, 1985, DOE/CH/10140-03(85-2), Office of Civilian Radioactive Waste Management, Washington, DC. Westinghouse Electric Corporation, 1983. Engineered Waste Package Conceptual Design: Defense High-Level Waste (Form 1), Commercial High-Level Waste (Form 1), and Spent Fuel (Form 2) Disposal in Salt, ONWI-438, prepared for Office of Nuclear Waste Isolation, Battelle Memorial Institute, Columbus, OH. Westinghouse Electric Corporation, 1986.

Waste Package Reference Conceptual Designs for a Repository in Salt, BMI/ONWI-517, prepared for Office of Nuclear Waste Isolation, Battelle Memorial Institute, Columbus, OH.

<u>APPLICABILITY TO LICENSING</u> [Ranking: Key Data (), Supporting Data (x)]

(a) Relationship to Waste Package Performance Issues Already Identified:

This report is related to issue 2.1.3.1 involving how the chemical characteristics of the brine reaching the waste package container will be affected by radiolysis.

- (b) New Licensing Issues
- (c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

(a) Organization Producing Data

Pacific Northwest Laboratory, Richland, Washington 99352

(b) Author(s), Reference, Reference Availability

Westerman, R. E., Haberman, J. H., Pitman, S. G., and J. S. Perrin, "Corrosion of Iron-Base Waste Package Container Materials in Salt Environments," PNL-SA-14029, March 1986

DATE REVIEWED: 6/22/87; Revised 7/31/87

TYPE OF DATA

Experimental, general corrosion

MATERIALS/COMPONENTS

Cast ASTM A-216 mild steel, grade WCA (candidate waste package material)

TEST_CONDITIONS

Steel tested as-cast, homogenized (long-term austenization -- $930^{\circ}C/24$ h/AC)) and normalized (short-term austenization -- $930^{\circ}C/1$ h/AC).

Casting size: 160 kg with minimum dimension of 120 mm (Ref. 1)

Specimen size: 15mm x 15mm x 1.5mm (as machined)

Aqueous environments: Bulk of data from tests reported conducted in brine/water (moist salt). Two synthetic Permian Basin brines used the following solutions: (1) dissolution of salt horizon cores & (2) high Mg⁺⁺ "inclusion brine". Moist salt prepared from dried synthetic salt horizon brine with 20 weight percent synthetic inclusion brine added to provide solid salt/brine mixture. Single test reported for unrefreshed autoclave exposure in synthetic salt horizon brine. Specimens exposed in sealed cans at 150°C. Maximum test period was 12 months.

DATA_COLLECTION/ANALYSIS

Corrosion rates based on weight-loss measurment after descaling with formaldehyde-inhibited HCl. Analysis of corrosion products by X-Ray diffraction and chemical analysis.

AMOUNT OF DATA

Data presented are summarized from the tables and figures, as follows:

Two tables:

٠.

1.	Composition	of a	singled	casting Stee	1 Tested (element,	weight %)	
	C 0.25	M	In 0.71	Si 0.	45 C:	r 0.41		
	Ní 0.23	C	u 0.14	S 0.0	18 F	e balano	ce	

2. Compositions of Synthetic Brines (ion, concentration mg/l))

<u>ion</u>	<u>synthetic salt horizon</u>	synthetic inclusion brine
N . +	103 000	<u>62</u> 000
Na	123,000	23,000
Ca ²⁺	1,600	15,000
Mg ²⁺	130	53,000
КŦ	40	10,000
C1-	191,000	210,000
SO4	3,200	160
HCO ₃	30	-
Br ⁻	32	2,400

Four figures:

- (3) composition of corrosion products from 12 month moist-salt test - Cl, Fe, H₂O, Na, Mg - O to 40 weight % on dry weight basis
- (4) microstructures of as-cast and normalized steel.

UNCERTAINTIES IN DATA

Author describes test conditions as "severe and conservative" with unlimited quantities of reactants.

DEFICIENCIES/LIMITATIONS IN DATABASE

None stated by author.

KEYWORDS

experimental data, supporting data, corrosion, x-ray diffraction, weight change, simulated field, brine, brine (high ionic content), brine (low

ionic content), Cl, Mg, high temperature, static (no flow), cast mild steel, A216 Grade WCA, cast, austenitized, homogenized, normalized, corrosion (general), corrosion (pitting)

COMMENTS

Paper quantifies several important factors affecting corrosion of cast steel in brine environments including:

(1) significantly higher corrosion rates observed in brines containing 53,000 mg/l Mg as compared with low Mg (130 mg/l) brines,

(2) austenizing heat treatment (normalization or homogenization) can reduce corrosion resistance of cast steel, a factor which must be considered when assessing microstructural changes due to weld closures or specified casting heat treatments to optimize mechanical properties.

(3) presence of solid brine phase is not necessary for high corrosion rates to be occur.

(4) corrosion rates decrease rapidly over the initial 12 months of exposure reflecting corrosion product buildup

(5) pitting rates in moist salt are similar to average penetration rates based on weight losses

Authors assume no effect of irradiation on corrosion rate citing previous report (Ref.1). Authors do not address potential effects of H_2 pressure buildup in sealed containers in reducing cathodic reactions. Changing corrosion rate with exposure time and environmental fluctuation is a key factor needing further study to assure valid extrapolations to longer term exposures and anticipated environmental variations. More detailed studies of corrosion product formation, structure and retention would be particularly useful. The experiments described in this report assume canisters will be subject to constant environment. Potential effects of alternate wetting and drying or waterline exposure should be considered.

RELATED HLW REPORTS

(1) Westerman, R.E., Haberman, J.H., Pitman, S.G., and Pulsipher, B.A., "Corrosion and Environmental-Mechanical Characterization of Iron-Base Nuclear Waste Package Structural Barrier Materials Annual Report FY 1984," PNL-5426, March 1986.

APPLICABILITY OF DATA TO LICENSING

[Ranking: key data (), supporting data (x)]

(a) Relationship to Waste Package Performance Issues Already Identified

ISTP Issue 2.2.4.1 What are the rates of corrosion as a function of time for the various corrosion modes of the waste package container.

ISTP Issue 2.4 How and at what rates will radionucides migrate through failed waste package.

- (b) New Licensing Issues
- (c) General Comments

••

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

(a) Organization Producing Data

Vitreous State Laboratory, The Catholic University of America, Washington, D. C.

(b) Author(s), Reference, Reference Availability

X. Feng, R. Adiga, A. Barkatt, A. Barkatt, W. Freeborn, P. Macedo, R. Mohr, C. Montrose, R. Mowad, E. Saad, and W. Sousanpour, "Effects of Composition On the Leach Behavior of West Valley HLW Glasses," September 1986

DATE REVIEWED: 4/21/87

TYPE OF DATA

Experimental data on glass leaching. Measured viscosities at 1100°C.

MATERIALS/COMPONENTS

West Valley Demonstration Project reference glass WV-205 and 18 other compositional variants were studied. Six of the glasses contained radiocactive uranium (0.5-1.0 weight percent UO_2) and thorium (3.2-3.6 weight percent ThO_2); the remainder contained Al and Zr surrogates.

TEST CONDITIONS

The 4-gram glass samples were ground into -100/+200 mesh powders. A static powder leach test (modified MCC-3 test) was used. The leaching environment was 40 mls of deionized water at T = 90°C.

METHODS OF DATA COLLECTION/ANALYSIS

Leaching Experiments:

Leachate concentrations were analyzed at 7 and 28 days. Dissolved boron was used to indicate the extent of glass dissolution. Boron concentration in leachant was plotted vs. reduced composition variable (referenced to WV-205 glass). No procedures were specified for determining viscosity.

AMOUNT OF DATA

Data presented are summarized in two tables and one figure, as follows:

Two tables:

- 1. Compositions of WV-205 and Derivative Glasses.
- 2. Powder Leach Test Results and Viscosities for WV-205 and Derivative Glasses.

One figure:

 Concentration of boron in leachant for modified MCC-3 leach tests plotted as a function of the reduced composition variable. (Ordinate: boron concentration, mg/l. Abscissa: reduced composition variable, wt%.)

UNCERTAINTIES IN DATA

None given.

DEFICIENCIES/LIMITATIONS IN DATABASE

In the abstract, the authors state that the conclusion that glass durability can be expressed as a function of a single reduced composition variable is valid for the narrow glass composition range studied and as long as the pH of the leachate remains nearly constant.

KEYWORDS

data analysis, experimental data, laboratory, deionized, high temperature, static (no flow), 90°C (leaching), borosilicate glass, U, Th, viscosity, leaching

COMMENTS

Corrosion resistance and processability are two of the primary criteria for nuclear waste glasses. Modifying the composition of borosilicate waste glasses to improve one of these properties generally has an adverse effect on the other. The present work attempts to show that certain compositional changes increase durability more than they degrade processability, i.e., increase viscosity. Specifically, aluminum is cited as being somewhat more effective in increasing durability than in increasing viscosity.

The data do in fact indicate that several high-aluminum glasses are among the most corrosion resistant, but viscosity data on several of these glasses are absent. As a result, it is doubtful that the conclusion is justified from the data presented. In fact, inspection of Table 2 indicates that the Defense Waste Reference Glass has a better combination of corrosion resistance and viscosity than any of the West Valley glasses, which are the focus of this study. The work does, however, demonstrate that, for the compositional variants in this study, the durability can be related to a single reduced composition variable.

RELATED HLW REPORTS

APPLICABILITY OF DATA TO LICENSING [Ranking: key data (), supporting (x)]

(a) Relationship to Waste Package Performance Issues Already Identified

This document address issues 2.3.2.1.1, which waste form dissolution mechanism or mechanisms are most likely?, 2.3.2.1.2, what are the rates of dissolution associated with the potential waste form dissolution mechanism?, 2.3.2.2, what non-radioactive dissolution products are likely to be produced from the waste form?

(b) New Licensing Issues

(c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

(a) Organization Producing Data

West Valley Demonstration Project, West Valley, New York

(b) Author(s), Reference, Reference Availability

L. R. Eisenstatt, "Description of the West Valley Demonstration Project Reference High-Level Waste Form and Canister," Revision 0, WVDP-056, July 28, 1986

<u>DATE REVIEWED: 4/20/87</u>

TYPE OF DATA

Three main data groups are given: 1) Composition, radioactivity, leaching behavior and physical properties of the WVDP High Level Waste form, each primarily on experimental measurements; 2) WVDP HLW canister dimensions, material, fabrication, labeling, and handling, according to present plans; 3) Expected characteristics of canistered waste, based on calculations.

MATERIALS/COMPONENTS

West Valley 205 glass waste forms that were melted in a full-scale Slurry Fed-Ceramic Melter (SFCM) or laboratory scale melter. Surface finish was either 200 grit as-cut or 600 grit as-polished.

TEST_CONDITIONS

Leaching tests were carried out using MCC-1 tests (90°C deionized water for 28 days, 400 mm² glass monoliths with S/V = 10/m) and pulsed flow tests (90°C deionized water, 25 percent exchange per week for 12 weeks).

METHODS OF DATA COLLECTION/ANALYSIS

Methods for chemical and radioactivity analyses were not specified. Viscosities were determined by beam bending viscometer (annealing range) and rotating spindle viscometer (high temperature). Heat capacities were determined on differential scanning calorimeter at 20°C per minute.

Mathematical methods were used to calculate expected of canistered waste characteristics.

AMOUNT OF DATA

Twenty-three tables:

- 1. A. PUREX Insoluble Solids Chemical Composition.
 - B. PUREX Solids Fission Products.
- 2. PUREX Supernatant Chemical Composition.
- 3. THOREX Waste Chemical Composition.
- 4. IE-96 Nominal Composition.
- 5. Glass Formers Added to West Valley HLW to Melt WV-205.
- 6. Reference 1987 Radionuclide Content (Curies) of West Valley Waste.
- 7. Composition of WV-205.
- 8. Reference Radionuclide Content of a Canister of WVDP HLW.
- 9. Composition of a Nonradioactive Analogue WV-205.
- 10. WV-205/SFCM MCC-1 Test Results, 200 Grit Cut Finish Specimens.
- 11. WV-205/SFCM MCC-1 Test Results, 600 Grit Polish Specimens.
- 12. WV-205/SM MCC-1 Test Results, 200 Grit Cut Finish Specimens.
- 13. WV-205/SM MCC-1 Test Results, 600 Grit Polish Specimens.
- 14. WV-205/SFCM Pulsed Flow Test Results.
- 15. WV-205/SM Pulsed Flow Test Results.
- 16. WV-205 Glass Physical Property Data: Summary.
- 17. Annealing Range Viscosity Data: WV-205.
- 18. High Temperature Viscosity Data.
- 19. Glass Transition Temperatures.
- 20. Heat Capacity (C_n) of WV-205.
- 21. Chemical Composition Requirements for Type 304 Stainless Steel.
- 22. Temperature Distribution in the Canister.
- 23. Fissionable Material Content of a Canister of WVDP HLW.

Eight figures:

- 1. West Valley High Level Waste Processing Flow Sheet.
- 2. Flow Test Results. (Leach rate of DWRG plotted vs. residence time in years.)
- 3. Viscosity vs. Temperature for WV-205.
- 4. Linear Thermal Expansion vs. Temperature for WV-205.
- 5. Heat Capacity vs. Temperature for WV-205.
- 6. CTS WVNS Canister. (Design drawing.)
- 7. West Valley High Level Waste Canister Labeling.
- 8. WVNS Canister Grapple.

UNCERTAINTIES IN DATA

Activities of fission products may vary from Table VI values as follows: U and Pu about 5 percent, Th about 20 percent, and other actinides about 50 percent.

Table VII contains composition ranges for major WV-205 components, based on uncertainties in waste sample analysis and expected waste stream variations.

Ranges of canister radioactivities are given for each radionuclide corresponding to 80-90 percent canister filling. Uncertainty limits are presented with several glass physical properties: Glass transition temperature: 451.3-494.8°C Linear expansion coefficient: ±5 percent Temperature for 100 poise glass viscosity: ±5 percent Dilatometric softening point of WV-205: ±5°C Upper and lower annealing points: ±5°C

DEFICIENCIES/LIMITATIONS IN DATABASE

Further glass durability testing needed. Repository groundwater will be added to the test matrix.

KEYWORDS

experimental data, planned work, radioactivity measurements, chemical analysis, differential scanning calorimetry, laboratory, deionized, high temperature, static (no flow), dynamic (flow rate given), stainless steel, 304 stainless steel, cold worked, commercial high level waste (CHLW), borosilicate glass, density, heat capacity, thermal expansion, viscosity, leaching (spent fuel)

GENERAL COMMENTS

The purpose of this report is to provide information on the West Valley High Level Waste product which is a) useful in dealing with the waste, and (b) required for Waste Acceptance Preliminary Specifications (WAPS). The report does contain a considerable amount of tabulated information on waste form characteristics based on experimental measurements. It is also a good source of summary information on waste canister plans and calculated expected properties of canistered waste. Glass properties discussed include density, viscosity, thermal expansion, and heat capacity. Leaching studies are given without much detail and include data reported elsewhere. As a result, this report is not a primary source of leaching information.

RELATED_HLW_REPORTS

- "Waste Acceptance Preliminary Specifications for the West Valley Demonstration Project High-Level Waste Form, Draft for Concurrence," OGR/B-9 (draft), U. S. Department of Energy, Washington, D.C., April 1986.
- R. G. Baxter, "Description of Defense Waste Processing Facility Reference Waste Form and Canister," DP-1066, Rev. 1, E. I. DuPont de Nemours and Co., Savannah River Plant, Aiken, SC, August 1983.

<u>APPLICABILITY OF DATA TO LICENSING</u> [Ranking: key data (), supporting data (x)]

(a) Relationship to Waste Package Performance Issues Already Identified

This document address issues 2.3.1., what the physical, chemical, and mechanical properties of the waste form?, 2.3.2.2, what non-radioactive dissolution products are likely to be produced from the waste form?

- (b) New Licensing Issues
- (c) General Comments

••

NBS Review of Technical Reports on the High-Level Waste Package for Nuclear Waste Storage

DATA SOURCE

(a) Organization Producing Data

E. I. duPont de Nemours and Co., Savannah River Laboratory, Aiken, South Carolina 29808.

(b) Author(s), Reference, Reference Availability

G. C. Wicks, N. E. Bibler, C. M. Jantzen, and M. J. Plodinec, "Repository Simulation Tests," American Ceramic Society Annual Meeting, 1984

DATE REVIEWED: 6/12/87

TYPE OF DATA

Description of repository simulation leaching test methods. Experimental data on glass leaching.

MATERIALS/COMPONENTS

SRP simulated waste glasses (exact type and composition not specified), both non-radioactive and radioactive.

TEST CONDITIONS

The repository simulation test is a static MCC-1 based test, modified so that the primary leaching vessel is made of the host rock (Tuff was used in this experimental study). The leachant was G-4 groundwater at 90°C. Surface are a to volume (SA/V) ratio was 1.0 cm^{-1} . The cup also contained a 304L stainless steel sample cage.

METHODS OF DATA COLLECTION/ANALYSIS

Tests were run for periods of 1, 3, and 6 months. Leachates were analyzed after exposure to determine both pH and species extracted from the waste glass and package components. Method of leachate analysis was not specified.

AMOUNT OF DATA

Three tables:

- 1. Repository Simulation Studies (list of three repository sites).
- 2. Repository Simulation Test Conditions (see Test Conditions above).
- 3. Tuff Tests Phase 1 (pH changes vs. time).

Seven figures:

۰. ۲

- 1. Analyses and Responsibilities (division of responsibilities between SRL and repository designer).
- 2. Experimental Apparatus.
- 3. Rock Cups and Waste Glass Samples (photograph).
- 4. Experimental Unit (photograph).
- 5. NL (normalized elemental mass loss) of Lithium in J-13 Tuff GW and System (graphed vs. time, 0-6 months).
- 6. NL (normalized elemental mass loss) of Boron in J-13 Tuff GW and System (graphed vs. time, 0-6 months).
- 7. NL (Li) for Radioactive vs. Non-Radioactive Tests (J-13 Tuff GW and System) (graphed vs. time, 0-90 days).

UNCERTAINTIES IN DATA

Even with rock samples from adjacent positions within a single core, the leaching behavior may vary significantly. Thus, the results of a few experiments may not give a true indication of the effects of the host rock on glass leaching performance.

DEFICIENCIES/LIMITATIONS IN DATABASE

The number of tests performed thus far under simulated repository conditions is limited, and interpreting the many possible interactions in the system is complex. Determining the concentrations of various species in the leachate is not sufficient to describe the glass behavior if those species can also originate from the host rock or other system components. Accordingly, internal calibrations for the species leached from the host rock are obtained from a separate volume of groundwater outside the host rock cup (i.e., in contact with the cup but not with the glass specimen). Data thus far are limited and more tests are needed to determine applicability of the method. Ultimately, a complete mass balance may necessary.

KEYWORDS

Experimental data, supporting data, simulated field, Yucca Mountain, tuff groundwater, tuff, static, DHLW, leaching

COMMENTS

This paper describes repository simulation tests designed "to assess the performance of SRP waste glass under the most realistic repository conditions that can be obtained in the laboratory." The factors that went into development of the test method are described. Particularly useful is a discussion of the limitations in repository simulation and the ways that they are addressed by the present method. Some limited data are presented from experiments in which the test is used to characterize leaching in tuff host rock.

In the experimental work, the effects of the host rock and canister material were incorporated into the leaching experiment. This procedure represents an advance in laboratory testing of glass leaching over the standard MCC-1 test. The use of actual groundwater as the leachant contributes to the realism of the simulation, although testing in pure water to approximate "worst case" conditions still has merit. Incorporation of groundwater flow into the test, which the authors suggest is possible, would make the test an even better simulation of repository conditions.

The limited experimental data illustrate the test method but have limited applicability. Because the composition of the waste glasses was not specified, the data are useful only for establishing internal consistency and demonstrating that the radioactive and non-radioactive glasses behaved similarly. The preliminary results presented suggest that the buffering action of tuff will stabilize groundwater pH at lower values and thereby retard glass leaching. This critical aspect of work on the tuff and other repositories is being investigated and reported in other papers scheduled for review. Such behavior should be substantiated and quantified with the final candidate waste form, once its composition has been decided.

RELATED HLW REPORTS

<u>APPLICABILITY OF DATA TO LICENSING</u> [Ranking: key data (), supporting data (x)]

(a) Relationship to Waste Package Performance Issues Already Identified

This review address the issue 2.3.2, what is the solubility of the waste form under the range of potential repository conditions? (Tuff primarly)

- (b) New Licensing Issues
- (c) General Comments

NBS Review of Technical Reports on the High Level Waste Package for Nuclear Waste Storage

DATA SOURCE

۷.

(a) Organization Producing Data

E. I. duPont de Nemours and Company, Savannah River Laboratory, Aiken, South Carolina 29808

(b) Author(s), Reference, Reference Availability

N. E. Bibler, M. J. Plodinec, G. G. Wicks, and C. M. Jantzen, "Glass Performance in a Geologic Setting," Summer National Meeting of the American Institute of Chemical Engineers, 1986

DATE REVIEWED: 6/24/87

TYPE OF DATA

The paper is a literature review of experimental leaching tests and leaching models. The tests reviewed are laboratory and in-situ simulated repository tests.

MATERIALS/COMPONENTS

Four glass compositions were generally studied in the reviewed work:

- 1. SRL-165
- 2. Defense Waste Reference Glass
- 3. SRL-131
- 4. Radioactive glass containing waste from SRP Tank 8.

TEST CONDITIONS

Detailed descriptions of test conditions were not given. In general, tests were conducted under simulated repository conditions, both in the laboratory and in -situ.

METHODS OF DATA COLLECTION/ANALYSIS

Not applicable - review paper.

AMOUNT OF DATA

Data presented are summarized in four tables and seven figures:

Four tables:

- 1. Principal Composition of Typical Glasses Used to Measure Performance of DWPF Glass.
- 2. Summary of Expected Repository Conditions.

- 3. Compositions of Groundwater in Potential Repositories.
- 4. Calculated Release from DWPF Glass in a Saturated Tuff Repository Using the Mass Transfer Model.

Seven figures:

- 1. Time Dependent Release of B from DWPF Glass (SRL-131) at 90°C in Deionized Water and Three Synthetic Groundwaters. (0 to 750 days, normalized mass loss range 0 to 140 g/m^2)
- 2. Effect of pH (1 to 11) on Silica Dissolution from Simulated DWPF Glass (SRL-131) at 90°C.
- 3. Dissolution of Cs-137 (0 to 4 g/m^2) from Radioactive DWPF Glass at 90°C in Tuff Groundwater in the Presence and Absence of Tuff Rock. (Leach time 0 to 160 days)
- Time Dependent Release of Li and B from DWPF Glass (SRL-165 Glass) at 90°C in Reduced and Oxic Simulated Basalt and Granite Groundwaters. (0 to 30 day leach periods)
- 5. Comparison of the Durability of DWPF Glass (SRL-165 Glass and SRL-13 1 Glass) with Basalt Rock and also Other Glasses Based on the Thermodynamic Hydration Model. (Mass loss based on Si, ranging from 0.1 to 1000 gram/m^2 , plotted vs. free energy of hydration, 10 to -20 kcal/mole)
- Release of Pu-239 from DWPF Glass (DWRG Glass) in Brine Solution at 90°C. (ppb Pu-239 plotted vs time, 0 to 60 days)
- 7. Coupling of Glass Performance and Repository Performance Based on the Model of Cheung. Peak Individual Dose Rates Calculated using Release Ra tes for DWPF Glasses Measured under Laboratory or In-situ Conditions. (Limiting individual peak dose rate as fraction of background, 10^{-6} to 1, plotted vs glass release rate 10^{-8} to $10^{-3}/y$)

UNCERTAINTIES IN DATA

Not applicable - review paper.

DEFICIENCIES/LIMITATIONS IN DATABASE

- 1. Site-specific properties of the actual repositories need to be determined.
- 2. Testing is needed using site-specific groundwaters and rock.
- 3. The role of colloids in repositories containing iron as the waste package overpack is not understood.
- 4. The effect of radiation on leaching in reducing groundwaters needs to be studied further.
- 5. Leaching of additional radionuclides in groundwater needs to be studied.

KEYWORDS

Literature review, leaching, simulated field, DHLW

COMMENTS

This paper reviews published studies on Defense Waste Processing Facility (DWPF) glass degradation in simulations of the proposed repository environments. Such studies overcome some of the limitations of similar experiments in which repository conditions are ignored. This is important because the performance of the glass, as the authors point out, can be directly affected by the test method. This question of test methodology and the ability to predict and simulate reposit ory conditions remains a central issue in evaluating studies of glass waste form leaching. A considerable amount of the discussion in this paper concerns test methods.

The specific effects of simulated tuff, salt, basalt, and granite repository environments on waste glass durability comprise the major part of the review. The paper contains a short overview of the effects of solution composition, pH, and pressure. In addition to reviewing experimental data, the paper discusses two models that couple glass leaching with repository conditions. The first model calculates radionuclide migration rates using known equations for mass transfer by convection and diffusion, assuming that groundwater flow is so slow that radionuclide concentrations at the glass surface are controlled by solubility. The second model is similar to the first, except that it calculates individual peak dose rates that would be obtained at the edge of the repository by drinking the groundwater. Both models are shown to predict low radionuclide release rates in repository environments.

The studies reviewed represent a growing body of work which supports the authors' hypothesis that Defense Waste Processing Facility glass will be more durable in the repository than in predictive laboratory tests. It must be recognized, however, that this hypothesis includes the implicit assumption that the repository simulation tests do indeed simulate the (as-yet poorly characterized) repository sites.

While the review is not exhaustive or extremely detailed, it is a well written, clear, useful summary of available data. Furthermore, it presents current thinking on some of the relationships between the repository environment and glass durability, and identifies several areas which must be studied further.

It is interesting to note that the paper points out that the 131-TDS glass, discussed in the paper, is no longer used for testing glass durability. There has been a shift to higher silica glasses to maximize overall glass properties. A considerable amount of the existing data on waste form leaching, and resulting hypotheses on leaching mechanisms, are based on this glass. As a result, it will be necessary to verify that what has been learned is applicable to the new compositions.

RELATED HLW REPORTS

 "Repository Simulation Tests", G. C. Wicks, N. E. Bibler, C. M. Jantzen, and M. J. Plodinec, American Ceramic Society Annual Meeting, 1984.

<u>APPLICABILITY OF DATA TO LICENSING</u> [Ranking: key data (), supporting data (x)]

(a) Relationship to Waste Package Performance Issues Already Identified

This review address issues 2.3, when, how, and at what rate will radionuclides be released from the waste form?, 2.3.2, what is the solubility of the waste form under the range of potential repository conditions?, 2.3.2.1.1, which waste form dissolution mechanism or mechanisms are most likely?, 2.3.2.2, what non-radioactive dissolution products are likely to be produced from the waste form?, 2.3.2.3, what are the solubilities of the radionuclides released from the waste form?, 2.3.2, what colloids or other suspended particles will be produced from the waste form?

- (b) New Licensing Issues
- (c) General Comments