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

October 15, 1987

Mr. Everett A. Wick
Technical Review Branch
Division of High-Level Waste Management
Office of Nuclear Materials Safety and Safeguards
U.S. Nuclear Regulatory Commission
Washington, DC 20555

Re: Monthly Letter Status Report for September 1987 (FIN-A-4171-7)

Dear Mr. Wick:

Enclosed is the September 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.

Recently, you suggested that selected key numerical results that are presented in a paper under review should be included in the NBS review of that paper. We note that this type of information is usually presented in the author(s)' abstract of the paper. For each paper reviewed by NBS, these abstracts will be included in our database. Thus, our lead workers feel that presentation of these numerical results in the NBS review would not enhance the quality of the reviews. Nevertheless, when the abstract does not contain information of this type, the review will highlight key numerical results whenever the data is deemed appropriate for inclusion in the review. Note that reviews contained in this letter report have the abstract included. We will continue this practice in the future.

The lead worker's responsibilities for NBS work related to waste-form degradation have been assigned to Dr. Helen Ondik, who has been active in other aspects of the program. We are confident that this change will enhance our capabilities in this area.

Sincerely,

Charles G. Interrante

Charles G. Interrante
Program Manager
Corrosion Group
Metallurgy Division

Enclosures

Distribution:
NMSS PM (1)
Ofc of the Director
  NMSS (Attn: PMPDAS) (1)
HLWM Div. Director (1)
HLTR Branch Chief (1)
Section Leader
  HLTR/Materials
  Engineering Section (1)
WM Docket Control Center (4)
Office of Research (1)
Office of Administration
  and Resource Mgmt.,
  Div. of Computer and
  Telecommunications
  Services (Attn: Director) (1)

Monthly Letter Report for September 1987

Published October 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 four Draft Reviews not previously submitted. Comments by NRC and its contractors are solicited.

1. UCRL-89988, "Selection of Candidate Canister Materials for High-Level Nuclear Waste Containment in a Tuff Repository," April 1984

2. DP-MS-85-141, "Leaching Fully Radioactive SRP Nuclear Waste Glass in Tuff Groundwater in Stainless Steel Vessels," May 1986

3. PNL-5426, "Corrosion and Environmental-Mechanical Characterization of Iron-Base Nuclear Waste Package Structural Barrier Materials Annual Report -- FY 1984," March 1986

4. "Aluminosilicate Saturation as a Solubility Control in Leaching of Nuclear Waste-Form Materials," Workshop on the Leaching Mechanisms of Nuclear Waste Forms," Summary Report, Gaithersburg, MD, May 19-21, 1982

BWIP -- BASALT WASTE ISOLATION PROJECT

Review has been initiated on three new reports this month. The first document SD-BWI-TI-298, 1985, "Buffering Capacity and Redox Control in Water-Rock Systems," presents an analysis of the water-basalt interactions occurring at elevated temperatures. The authors concluded that the redox conditions in a basalt/water environment are controlled by the products of these reactions and that these products may change with time.

The second report "Pit Propagation of Carbon Steel in Groundwater," by J. A. Beavers and A. J. Markworth, 1987, deals with the corrosion behavior of AISI 1020 carbon steel in simulated Grande Ronde basalt groundwater. In this study, pitting behavior and propagation rates were experimentally measured at 25°C and theoretically modelled. It was found that propagation rates depend critically upon the reactivity of the pit walls. Environments that promote active corrosion of carbon steel would not support pit propagation even in the presence of a differential aeration and pH cell.

The third report SD-BWI-TI-249, 1984, "Preliminary Reliability Analysis of Container Lifetime," presents the results of several computer calculations of the container lifetime (for low-carbon steel containers) in a basalt repository. For these calculations, Monte Carlo simulation of the input parameters was employed and a corrosion model based on uniform corrosion behavior was assumed. The calculations indicated the waste containers will not fail prior to the 1000-year containment period.

BWIP -- Review has been initiated on the following reports:

1. SD-BWI-TI-298, "Buffering Capacity and Redox Control in Water-Rock Systems," October 1985

2. "Pit Propagation of Carbon Steel in Groundwater," by J. A. Beavers and A. J. Markworth, 1987

3. SD-BWI-TI-249, "Preliminary Reliability Analysis of Container Lifetime," September 1984

BWIP -- Review is continuing on the following 13 reports.

1. 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. RHO-BW-LD-48, "Sorption Behavior of Selected Radionuclides on Columbia River Basalts," August 1986

7. NUREG/CR-4309, ORNL-6199, "Valence Effects on Solubility and Sorption: The Solubility of Tc(IV) Oxides," March 1986

8. RHO-BW-SA-554P, "Determination of Dissolved Gases in Basalt Groundwater in the Pasco Basin, Washington," September 1986

9. RHO-BWI-LD-43, "Sorption of Selected Radionuclides on Secondary Minerals Associated with the Columbia River Basalts," Informal Report, April 1981

10. SD-BWI-TA-023, "Feasibility Assessment of Copper-Base Waste Package Container Materials in a Repository in Basalt," September 1986

11. SD-BWI-TI-309, "Determining the Reversibility of Oxidation-Reduction Reactions in Groundwater," December 1985

12. SD-BWI-DP-021, "BWIP Data Package for Waste Characteristics," July 1982

13. RHO-BW-SA-462P, "Gamma and Alpha Radiation Levels in a Basalt High-Level Waste Repository: Potential Impact on Container Corrosion and Packing Properties," September 1985

NNWSI -- NEVADA NUCLEAR WASTE STORAGE INVESTIGATIONS

Review was initiated for a report on oxidation of spent fuel in tuff, HEDL-7540, 1985, "Technical Test Description of Activities to Determine the Potential for Spent Fuel Oxidation in a Tuff Repository." This paper describes a test designed to study the oxidation of spent fuel at high-temperature conditions expected initially in a repository and subsequent lower-temperature conditions that will exist later. The paper describes the rationale for the test procedure described.

NNWSI -- Review was initiated on the following report.

1. HEDL-7540, "Technical Test Description of Activities to Determine the Potential for Spent Fuel Oxidation in a Tuff Repository," June 1985

NNWSI -- Review is continuing on the following 12 reports.

1. UCRL-94708, "Carbon-14 in Waste Packages for Spent Fuel in a Tuff Repository," October 1986

2. UCRL-94633, "Experimental Study of the Dissolution Spent Fuel at 85°C in Natural Groundwater," December 1986

3. UCRL-95962, "Hydrogen Speciation in Hydrated Layers on Nuclear Waste Glass," January 1987

4. UCRL-94658, "Integrated Testing of the SRL-165 Glass Waste Form," December 1986

5. UCRL-91258, "Leaching Savannah River Plant Nuclear Waste Glass in a Saturated Tuff Environment," November 1984

6. UCID-20895, "Application EQ3/6 to Modeling of Nuclear Waste Glass Behavior in a Tuff Repository," May 1986

7. UCRL-92891,"LWR Spent Fuel Characteristics Relevant to Performance as a Wasteform in a Potential Tuff Repository," June 1985

8. ANL-84-81, "NNWSI Phase II Materials Interaction Test Procedures and Preliminary Results," January 1985

9. UCRL-94500, "Long-Term Corrosion Behavior of Copper-Base Materials in a Gamma-Irradiated Environment," December 1986

10. HEDL-TME 85-22, "Results from Cycles 1 and 2 of NNWSI Series 2 Spent Fuel Dissolution Tests," May 1987

11. UCRL-94363, "Hydrological Properties of Topopah Spring Tuff - Laboratory Measurements," December 1985

12. UCRL-53761, "Waste Package Performance Assessment: Deterministic System Model Program Scope and Specification," October 1986

SRP -- SALT REPOSITORY PROJECT

SRP -- Review is continuing on the following 10 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

6. BMI/ONWI-490, "Waste Package Materials Testing for a Salt Repository: 1982 Status Report," August 1983

7. PNL-3484, "Investigation of Metallic, Ceramic, and Polymeric Materials for Engineered Barrier Applications in Nuclear-Waste Packages," October 1980

8. 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. ONWI-384, "A Sensitivity Study of Brine Transport Into a Borehole Containing a Commercial High-Level Waste Canister," February 1987

10. PNL-5650, "FY-1984 Annual Report: Spent Fuel and UO_2 Source Term Evaluation Results," February 1986

WASTE FORM DEGRADATION

During September, reviews were initiated on two reports involving leaching studies carried out under simulated repository conditions. The first document SD-BWI-TI-312, Rev. 0, 1986, "Progress Report on the Hydrothermal Interaction of Defense Waste Glasses with Basalt and Groundwater at 140°C," deals with interactions of two glasses (ATM-9 and ATM-11) tested in basalt with GR-3 and GR-4 groundwater. The second report BMI/ONWI-305, 1982, "Reaction and Devitrification of a Prototype Nuclear Waste Storage Glass with Hot Magnesium-Rich Brine," discusses tests on PNL-76-68 glass in brine. These studies used nonstandardized leaching tests and glasses with compositions different from the current "optimized" formulations; nevertheless, these studies are important because of the light they can shed on leaching mechanisms in a realistic repository environment.

Draft reviews of Chapters 1 and 7 of PNL-5157, "Final Report of the Defense High-Level Waste Leaching Mechanisms Program" will be available in October. A review of Chapter 4, "Dissolution of Specific Radionuclides," may also be completed by the end of October. Chapter 2, "Surface Layers in Leached Borosilicate Glass High-Level Defense Nuclear Waste Forms," and Chapter 3, "Environmental Interaction," are expected be completed within a month therafter.

A summary report for the Materials Characterization Center's Workshop on Leaching Mechanisms of Nuclear Waste Forms, May 19-21, 1982, Gaithersburg, Maryland (PNL-4382) will be assigned for review. Selected papers in the report are under review and others will be assigned to appropriate reviewers. This report summarizes the second workshop in a series held as part of the DOE glass leaching mechanisms program. The program was coordinated by J. E. Mendel of the Materials Characterization Center (MCC). Six laboratories and associated lead investigators were the main participants. The workshop consisted of five sessions, and each session contained a number of papers. The session topics included leaching mechanisms, special topics in static and low-flow-rate testing, near-field hydrological, diffusion and convection phenomena, and the status of multicomponent interactions testing. One session was devoted to MCC-14.

WASTE FORM DEGRADATION -- Review has been initiated on the following reports.

1. SD-BWI-TI-312, Rev. 0, "Progress Report on the Hydrothermal Interaction of Defense Waste Glasses with Basalt and Groundwater at 140°C," January 1986

2. BMI/ONWI-305, "Reaction and Devitrification of a Prototype Nuclear Waste Storage Glass with Hot Magnesium-Rich Brine," October 1982

3. PNL-4382, "Materials Characterization Center's Workshop on Leaching Mechanisms of Nuclear Waste Forms," May 19-21, 1982

WASTE FORM DEGRADATION -- Review is continuing on the following 11 reports.

1. "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

2. "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

DATABASE ACTIVITIES

The keyword checklist was modified with changes previously requested by the project leader. A copy of this revised checklist will be appended to "Evaluation and Compilation of DOE Waste Package Test Data," Volume 3. The file structure of the database was also changed to incorporate a new key field. This key field contains a separate set of keywords for Non-metallic waste forms. In addition, data entry programs were modified to incorporate these changes in the file structure. Carla Messina began work on converting Revelation^R files and programs to the new Advanced Revelation^R format. Advanced Revelation^R has menu capabilities regarded to be superior to those developed at NBS. After the conversion is complete, it will facilitate access to the data files using Run-time^R, the program used at both the NRC and the NBS to access the data files.

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.

Conclusions, results, and recommendations for the work reviewed to date are given in each review form under the heading GENERAL COMMENTS OF REVIEWER.

TASK 3 -- Laboratory Testing

The work on each of the four projects reported below is on schedule with the work statements listed in their respective proposals. The work conducted in September 1987 is reported below. Work conducted in previous months was reported earlier.

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

During September, the first working version of the computer program used for the acoustic emission experiment was completed. A few enhancements to the program were recommended by the project leader and work has begun on these embellishments. Techniques and methods for preliminary testing of the program were discussed. The program will be subjected to these preliminary tests in October.

Other developments this month were in the areas of set-up of chemical and electrical systems.

In order to eliminate errors caused by thermal variations and other factors, the polarity of the current passed through the specimen will be oscillated. To do this, an electrical circuit was designed and built around a series of two-pole, one-throw relays. These relays are interfaced to and actuated by the minicomputer, which controls the experiment. The relays are reset at selected preset, user-defined intervals. A small junction box was also designed and built to permit easy "plug-in-type" wiring of the specimen into the circuit. Contained in this junction box is a high-accuracy resistor, which will be used to accurately determine the current furnished to the test specimens. This current value is used to calculate the crack length. Both the current junction box and the circuit for changing the polarity were extensively tested both manually and by computer operation. This was done to assure that deviations in the output current occurred beyond the forth significant figure. The settling time required by the current source was in the millisecond range, and therefore it was determined to be appropriately short for these measurements.

To comply with NBS's safety standards, a chemical system for scrubbing H_2S from the effluent gas has been revamped, and two large capacity dryer/scrubber towers have been installed. These scrubbers should permit long operating times before replacement of their contents (soda ash) is needed. The various items of equipment designed for use in the experiment were set up in the fume-hood location in which these tests will first be conducted.

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

Measurements of the coefficient of diffusion for oxygen in a 3.5 percent NaCl solution at room temperature have been carried out. This measurement is important because its results can be compared to values (in the literature) that were obtained using other techniques. Our results indicate a range of coefficient-of-diffusion values of from 1.8 to 3.0 x 10^{-5} cm^2/s. These results are in excellent agreement with values reported in the literature.

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

Cyclic polarization tests were conducted with A27 low-carbon steel exposed to 95°C Grande Ronde No. 4 water containing a mixture of 75 percent crushed Cohassett Flow basalt and 25 percent Wyoming bentonite. The basalt rock and bentonite were to fill one half of the workable volume of the testing flask, but the bentonite expanded upon becoming wet and the mixture filled approximately two thirds of the flask.

Two effects of adding the basalt and bentonite to the testing environment were observed: (1) An expansion of the test media, and (2) a lowering of the pH from 9.75 to 8.04 at 45°C and to 7.66 at 64°C. The solution and mixture were in place at temperature for approximately one hour. Environmental effects could change more with a longer time.

The effects of this change in the environment on the corrosion behavior did not appear to be great, based on a single test. The initial specimen electrode potential in the water/basalt/bentonite mixture was -0.769 volts vs. a saturated calomel electrode, and the potential dropped to -0.788 volts after a period of 15 minutes. Except for the shift in potential, the polarization curves appear to be similar to those measured in Grande Ronde No. 4 water without the mixture, and pitting does not seem to be indicated. Further analysis will be made of these data, additional tests will be conducted, and the specimens will be studied microscopically for any evidence of pitting.

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

Approximately one half of the brief literature survey on Zircaloy corrosion has been completed. The work on the review is in progress.

The cyclic polarization curves on Zircaloy-4 in J-13 water at 95°C have been measured and are being studied. Additional tests are planned for Zircaloy-4 and Zircaloy-2 including material in tubular form for cladding and welded material.

TASK 4 -- General Technical Assistance

Our reply to your request of August 26, 1987, for general technical assistance, rendered under Task 4 of FIN-A-4171-7 and concerning the ORNL document titled "Responses to Comments on Draft Repository Environmental Parameters and Models/Methodologies Relevant to Assessing the Performance of HLW Packages in Basalt, Tuff and Salt," NUREG/CR-4134lR1, was transmitted to E. Wick on September 2, 1987.

Dr. Charles Interrante and Mr. Edward Escalante attended a preliminary meeting on August 11, 1987, in Rockville, Maryland, on NNWSI/NRC/Nevada Waste Package Appendix VII visit at Lawrence Livermore National Laboratory, Livermore, California.

Dr. Charles Interrante and Mr. Edward Escalante met with K. Chang and others at the Wilsite Building (NRC), Silver, Spring, Maryland on September 8, 1987, for a briefing on the Consultational Draft SCP for Yucca Mountain.

Dr. Charles Interrante and Mr. Edward Escalante visited the reading room in the Forrestal Building on September 15 and September 23, 1987 to read selected parts of the Consultational Draft SCP for Yucca Mountain.

Draft Copy

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

DATA SOURCE

(a) Organization Producing Data

Lawrence Livermore National Laboratory, Livermore, CA

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

McCright, R. D., Weiss, H., Juhas, M. C. and Logan, R. W., "Selection of Candidate Canister Materials For High-Level Nuclear Waste Containment in a Tuff Repository, UCRL-89988, April 1984

DATE REVIEWED: 9/30/87

TYPE OF DATA

1) Experimental
2) General corrosion, localized corrosion, stress corrosion.

MATERIALS/COMPONENTS

AISI Type 304L, Type 316L, Type 317L, Type 321, Type 409, Type 416, Alloy 825, 2.25Cr - 1Mo alloy steel, 9Cr - 1Mo alloy steel, C1020 carbon steel, C1025 carbon steel, A36 carbon steel, A366 carbon steel.

TEST CONDITIONS

1) a. As received - all materials
   b. Sensitized - 304L
   c. Annealed - 304L
2) Rectangular 1 x 2 in coupons
3) a. Tuff Conditioned J-13 Water at 50, 70, 80, 90, 100, and 150°C.
   b. J-13 Water and Tuff conditioned J-13 Water at 105°C with 3×10^5 rd/h, and at 150°C with 6×10^5 rd/h.
   c. Steam above J-13 water.

METHOD OF DATA COLLECTION/ANALYSIS

1) Electrochemical polarization tests include Tafel slope extrapolation, linear polarization resistance measurements for determining corrosion rates, cyclic anodic polarization scanning for indications related to pitting and crevice corrosion.
2) Slow strain rate and four-point bent beam techniques for stress corrosion testing. Gravimetric weight loss measurements for determining corrosion rates.

AMOUNT OF DATA

Eight tables:

Table 1, chemical composition of welded tuff (%);
Table 2, chemical composition of J-13 water (mg/l);
Table 3, dimensions and power output of waste packages (cm, kw);
Table 4, chemical composition of J-13 water expected at 90 and 100°C (mg/l);
Table 5, chemical composition of reference alloy and four alternative canister materials (wt.%);
Table 6, results of coupon corrosion tests for three carbon steels, two alloy steels, and five stainless steels in conditioned J-13 water at 100°C and steam at 100°C (μm/y and visual description of surface);
Table 7, results of coupon corrosion tests in two gamma radiation levels (3 and 6 x 10^5 rd/h) for C1025 carbon steel, 9Cr-1Mo alloy steel, annealed 304L stainless steel, and sensitized 304L stainless steel (μm/y);
Table 8, results of slow strain rate tests of annealed 304L and sensitized 304L in J-13 water, steam, and air (strain rate, elongation, reduction in area, yield stress, ultimate tensile stress, and results).

Eight figures:

Figure 1, stratigraphic section on Nevada tuff (surface to 750 m below surface);
Figure 2, temperature(0-300°C)-time(0-300 y) of canister surface for different packages;
Figure 3 (initial exposure) and 4(after 500 h), electrochemical determination of corrosion currents(0-1600 nA/cm2) and corrosion rates(μm/y) for different alloys in J-13 water at 100°C;
Figures 5 - 8, corrosion and protection potentials(-600 to +600 mV vs SCE) for 304L, 316L stainless steels, 321 and 825 alloy steels in tuff conditioned J -13 water at different temperatures(40-100°C).

UNCERTAINTIES IN DATA

Not discussed.

DEFICIENCIES/LIMITATIONS IN DATABASE

The authors acknowledge that these are preliminary results based on short-term exposure times (max. 2 months).

Not discussed.

KEYWORDS

experimental data, electrochemical, simulated field, Yucca Mountain, J-13 water, air, tuff composition, gamma radiation field, ambient temperature, high temperature, ambient pressure, nickel base, stainless steel, steel, 304L stainless steel, 316L stainless steel, 321 stainless steel, 1020 carbon steel, 1025 carbon steel, high-nickel alloy 825, annealed, sensitized, slow strain rate, four-point bent beam, J-13 steam, tuff, corrosion (crevice), corrosion (general), corrosion (stress cracking) SCC, sensitization

GENERAL COMMENTS OF REVIEWER

This report describes initial results of corrosion studies on alloys being considered as possible canister or canister liner materials. The longest exposure time in any given environment is 2 months. The slow strain rate results on annealed 304L and sensitized 304L stainless steel in J-13 water did not reveal any susceptibility to stress corrosion failure. Future tests in water vapor may reveal a more aggressive environment.

The 1000-hour coupon tests revealed no evidence of localized attack for the stainless steels and the 9Cr-1Mo alloy steel, but there was considerable attack on all the carbon steels and the 2.25Cr-1Mo alloy.

In a gamma radiation field, the 304L stainless steel, and the 9Cr-1Mo alloy steel had higher rates of corrosion than similar specimens not irradiated.

These preliminary data indicate that the test procedures are reasonable, but longer exposures and more data are needed before any conclusions can be drawn.

APPLICABILITY OF DATA TO LICENSING

Ranking: key data ( ), Supporting (X)

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

Related to NNWSI ISTP issues 2.2.4, the potential corrosion failure modes for the waste package container, and 2.2.4.2, the effects of radiation on the corrosion failure modes and associated corrosion rates for the waste package container.

(b) New Licensing Issues

(c) General Comments

AUTHOR'S ABSTRACT

A repository located at Yucca Mountain at the Nevada Test Site is a potential site for permanent geological disposal of high level nuclear waste. The repository can be located in a horizon in welded tuff, a volcanic rock, which is above the static water level at this site. The environmental conditions in this unsaturated zone are expected to be air

and water vapor dominated for much of the containment period. Type 304L stainless steel is the reference material for fabricating canisters to contain the solid high-level wastes. Alternative stainless alloys are considered because of possible susceptibility of 304L to localized and stress forms of corrosion. For the reprocessed glass wastes, the canisters serve as the recipient for pouring the glass with the result that a sensitized microstructure may develop because of the times at elevated temperatures. Corrosion testing of the reference and alternative materials have begun in a tuff-conditioned water and steam environments.

Draft Copy

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

DATA SOURCE

(a) Organization Producing Data

E. I. du Pont de Nemours & Company, Savannah River Laboratory, Aiken, South Carolina 29808

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

Bibler, N. E., Leaching Fully Radioactive SRP Nuclear Waste Glass in Tuff Groundwater in Stainless Steel Vessels," DP-MS-85-141, May 1986

DATE REVIEWED: 6/19/87; Revised 9/30/87

TYPE OF DATA

Experimental data, borosilicate glass leaching

MATERIALS/COMPONENTS

Borosilicate glass with Savannah River Plant (SRP) radioactive waste
" " " simulated SRP radioactive waste
Type 316 stainless steel container (Parr reaction vessel)

TEST CONDITIONS

Aqueous environment - J-13 groundwater with borosilicate glass, 90°C, surface area to volume ratio = $100 m^{-1}$

METHODS OF DATA COLLECTION/ANALYSIS

Plasma induced spectroscopy
Ion chromatography
Calibrated radionuclide counting techniques

AMOUNT OF DATA

Four tables

(1) Composition of SRP radioactive glass
(2) Normalized mass losses (g/m^2) for ^{90}Sr and ^{238}Pu for 14 to 134 day exposures in 316 stainless steel containers.
(3,4) Final pH values and concentrations of anions (F^-, Cl^-, NO_2^-, NO_3^-, $SO_4^=$) for leach tests in 316 stainless steel and Teflon^R containers

Three graphs

(1) Normalized mass losses for ^{137}Cs (0 to 5 g/m^2 for 0 to 150 days) based on tests in Teflon^R containers
(2) Comparison of leaching rates for B, Li, and ^{137}Cs in 316 (0-20 g/m^2) stainless steel and Teflon^R (0 to 5 g/m^2) containers for exposures of up to 134 days
(3) Comparison of leaching (0 to 6 g/m^2) rates for B and Li from glass with radioactive and nonradioactive waste , J-13 water, 90°C, approximately 200 day exposure

Twelve references cited

UNCERTAINTIES IN DATA

Data scatter reflects nonuniform surface finish achieved during remote glass disc polishing and the presence of insoluble particulates in the nonacidified leachates leading to nonrepresentative sampling.

DEFICIENCIES/LIMITATIONS IN DATABASE

None stated by author

KEYWORDS

data analysis, experimental data, supporting data, spectroscopy, ion chromatography, laboratory, J-13 water, high temperature, neutral solution (ph = 7), static (no flow), stainless steel, 316 stainless steel, Teflon, defense high level waste (DHLW), 239Pu, 137Cs, 90Sr, radiation effects

GENERAL COMMENTS OF REVIEWER

This paper describes leaching rate test procedures used to evaluate the performance of SRP high level nuclear waste borosilicate glass in a tuff environment. Test results are used to substantiate important conclusions regarding interactions between borosilicate glass and the containment vessel proposed for glass waste forms. Type 316 stainless steel containers do not introduce radiolysis effects which the authors previously reported for Teflon. These effects are attributed to leaching of significant amounts of F^-, Cl^-, NO_2^- and NO_3^- from the Teflon. The paper also states important conclusions regarding a lack of previously observed radiolysis effects on tuffaceous groundwater (J-13 water at pH 7.4), even in the presence of dissolved air. Similarity of leach rates for radioactive SRP glass and nonradioactive glass with simulated waste are also noted. A major limitation of the test procedures is the use of the static MCC-1 leach test simulating a water saturated environment. Considerable data scatter was introduced by nonuniform surface finishes on the glass samples which were produced by the remote surface polishing techniques used.

APPLICABILITY OF DATA TO LICENSING

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

(a) Relationship to Waste Package Performance Issues Already Identified

(b) New Licensing Issues

Related to issue 2.3, when, how, and at what rate will radionuclides be released from the waste form?

(c) General Comments

AUTHOR'S ABSTRACT

SRP glass containing actual radioactive waste was leached in static tests at 90°C in a tuffaceous groundwater (J-13 water at pH ~7.4) at a SA/V ratio of 100^{m1} in 316 stainless steel vessels. Tests were performed for time periods up to 134 days. Normalized mass losses were calculated for Cs-137, Sr-90, and Pu-238. The Cs-137 in the leachate appeared to reach a steady value of ~3 g/m^2, corresponding to a steady-state concentration of only 1.0 ppb for total cesium. The mass losses based on Sr-90 and Pu-239 appearing in solution were low (<0.3 and <0.01, respectively) because of their low solubilities. However, significant amounts of these radionuclides had deposited on the steel vessel while the amount of deposited Cs-137 was negligible. During the leach tests, the pH changed <0.4 units and the only significant effect of radiolysis was reduction of NO_2^-. When compared to earlier tests, the results confirm that leach rates in the earlier tests with radioactive glass in Teflon^R (product of Du Pont) vessels were high due to radiolysis of the Teflon^R. The results also indicate that radioactive and nonradioactive glasses of comparable composition and surface finish leach essentially identically.

Draft Copy

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, WA 99325

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

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

DATE REVIEWED: 7/29/87; Revised 9/30/87

TYPE OF DATA

Experimental data.
Uniform corrosion, non-uniform corrosion (pitting), stress corrosion cracking (slow strain rate and corrosion-fatigue tests).
Statistical analyses

MATERIALS/COMPONENTS

Waste package overpack
ASTM A216 grade WCA cast steel
- as cast (majority of tests)
- normalized (930°C/1 hr/air cool)
- homogenized (930°C/24 hr/air cool)

ASTM A27 grade 60-30 cast steel
- as cast
- normalized (927°C/5 hr/air cool)

AISI 1025 wrought steel
- hot rolled

2.5Cr - 1Mo cast steel
- as cast

ASTM A536-77 ductile cast iron, grade 60-40-18
- as cast

High purity iron (99.87% Fe)

Note: ASTM A216 test samples obtained from single 352 lb. casting with minimum dimension of 4.7 in. Casting/plate sizes for other materials not given.

TEST CONDITIONS

Three synthetic Permian Basin aqueous brine environments, PBB1, PBB2, and PBB3:

(1) intrusion brines (saturated brine) simulating (a) dissolution of salt horizon core (PBB1) and (b) supernatant fluid above precipitated solids after heating brine to 150°C (PBB2).

(2) high Mg inclusion brine (PBB3).

Tests conducted:

(1) Intrusion brine tests conducted in flowing autoclave tests at 150oC with PBB2 brine under both "anoxic" (~50 ppb O_2) and "oxic" (~1.5 ppm O_2) conditions.

(2) Intrusion brine tests in irradiated flowing autoclaves with PBB2 brine at 150°C. ^{60}Co gamma-radiation intensities of 1×10^5 and 2×10^3 rad/h. Specimens with artificial pits included.

(3) Moist salt exposures in sealed cans at 150°C. Dried PBB1 or reagent grade NaCl with liquid added as PBB1, PBB3 or saturated NaCl brine.

(4) Slow strain rate tests (SSR) in flowing PBB2 brine and in air at 150°C with strain rates of 2×10^{-7}/s and 1×10^{-4}/s. Inlet oxygen levels of 0.1, 1 and 2-3 ppm. Some tests run with pre-exposed specimens.

(5) Corrosion fatigue tests with notched compact tension specimens exposed in air, deionized water and PBB2 brine at 150°C. Tests conducted at frequencies of 0.1, 1 and 10 Hz.

METHODS OF DATA COLLECTION/ANALYSIS

Corrosion rates based on weight loss versus time. Measurements were taken after descaling with formaldehyde-inhibited HCl. Analysis of corrosion products by X-ray diffraction and chemical analysis. Reduction in area and elongation measurements of specimens in SSR tests, energy absorption calculations, metallographs and scanning electron micrographs of fracture surfaces. Crack-growth rates recorded as function of stress intensity in corrosion fatigue tests.

AMOUNT OF DATA

Eleven tables in text plus 19 tables in Appendix. Thirty-three figures.

Table 1 - Compositions of cast and wrought ferrous materials
ASTM A216 Grade WCA, ASTM A27 Grade 60-30, AISI 1025 wrought steel, 2.5Cr - 1Mo cast steel, ASTM A536-77 Grade 60-40-18, high purity iron. Weight % of C, Mn, Si, P, S, Mo, Cr, Ni, Fe

Table 2 - compositions of synthetic brines (ion, concentration - mg/l)

	PBB1	PBB2	PBB3
Na^+	123,000	123,000	23,200
Ca^{2+}	1,560	1,110	14,700
Mg^{2+}	134	122	53,200
K^+	39	39	10,500
Sr^{2+}	35	35	--
Zn^{2+}	8	8	8
Cl^-	191,000	191,000	210,000
SO_4	3,200	1,910	160
HCO_3	30	23	--
Br^-	32	24	2,400
F^-	1	1	--

Table 3 - List of materials included in general corrosion/intrusion brine corrosion studies - anoxic and oxic test conditions, A216, A27 (as cast and normalized), 1025 steel, ductile iron, 2.5Cr - 1Mo cast steel, high purity iron. Exposure times of 7 to 21 months, 5 to 13 duplicate specimens tested under each condition.

Table 4 - Comparisons of estimated corrosion rates for general corrosion in anoxic simulated inclusion brine PBB2 at 150°C. Corrosion rates range from 4.8 to 14.6 μm/y with standard errors of 0.71 to 1.11.

Table 5 - Similar to Table 4, data for oxic brine. Corrosion rates range from 10.1 to 25.2 with standard error of 0.68 to 1.06.

Table 6 - Corrosion rates of reference A216 Grade WCA steel in moist salt environments - 150°C, 1 month, 20% H_2O. Corrosion rates range from 4.3 to 4.8 μm/y in NaCl/NaCl brine, 10 to 11 in PBB1/PBB1 brine and 580 to 910 μm/y in PBB1/PBB3 brine.

Table 7 - List of materials included in irradiation-corrosion studies. A216, A27 (as cast and normalized), 1025 steel, ductile iron, 2.5Cr - 1Mo steel, high-purity iron, ^{60}Co gamma radiation intensities of 1 x 10^5 and 2 x 10^3 rd/h with maximum exposure times of 5 to 18 months. Seven to fourteen reduplicate specimens included in each test.

Table 8 - Comparison of corrosion rates (8.4 to 48.4 μm/y) in 150°C, PBB2, 2 x 10^3 rd/h test vs. exposure time (1300 to 9000 hours) and top, center and bottom locations in autoclave.

Table 9 - Same as Table 8, 1 x 10^5 rd/h. Corrosion rates range from 37 to 185 μm/y.

Table 10 - Statistical analysis of reults of slow strain rate tests. Standard errors for reduction of areas range from 26 to 70 in air and 15 to 72 in PBB2 brine. Ranges for elongation range from 18 to 31% in air and 13 to 32% in PBB2 brine.

Table 11 - Results of slow strain rate tests on A216 at 30°C and 90°C with strain rate of 2×10^{-7}/s and irradiation intensity of 3×10^5 rd/h

Approximately 19 additional tables included in Appendices - tabulate detailed data for all tests:

Appendix A - general corrosion tests
Appendix B - moist salt studies
Appendix C - irradiation-corrosion studies
Appendix D - slow strain rate and corrosion-fatigue tests
Appendix E - compilation of statistical data

Figure 1 - Schematic of flowing autoclave test

Figure 2 - Schematic of moist salt test configuration

Figure 3 - Schematic of irradiation-corrosion test facility

Figure 4 - Schematic of slow strain rate test system (unirradiated)

Figure 5 - Schematic of slow strain rate test system (irradiated)

Figure 6 - Schematic of corrosion fatigue test facility

Figure 7 - Metal penetration rate vs. time with 95% confidence limits - A2126 steel, 150°C, PBB2 brine, unirradiated. Penetration rates range from 10 to 40 μm/y with exposures of up to 6000 hours.

Figure 8 - Corrosion rates of A216, 1025 steel and ductile cast iron in dried synthetic PBB1 salt/PBB3 brine as a function of water content. One month at 150°C. Corrosion rates range from 0.1 to 0.75 μm/y for water contents of 18 to 32%.

Figure 9 - Same as Figure 8, three month exposure. Similar range of corrosion rates.

Figure 10 - Corrosion rate vs. Mg content of brine. Moist salt test with A216 cast steel. Corrosion rates range from 4 to 700 μm/y with Mg contents ranging from 0 to 3%.

Figure 11 - As cast A216 specimen surface microstructure after moist salt test (PBB1/PBB3)

Figure 12 - Effect of 1×10^5 rd/h radiation on corrosion of ferrous materials in PBB2 intrusion brine at 150°C. Maximum corrosion rate is 200 μm/y, maximum exposure time is 21 months.

Figure 13 - Comparison of ferrous materials is intrusion brines irradiated at 1×10^5 and 2×10^3 rd/h. Units same as figure 12.

Figure 14 - Mean penetration rates for A216 in irradiated PBB2 brine at 150°C. Corrosion rates of 10 to 25 μm/y for 2×10^3 rd/h and 100 to 150 μm/y for 1×10^5 rd/h. Maximum exposure time is 6000 hr.

Figure 15 - Photo of A27 specimen after PBB2 brine exposure with 1×10^5 rd/h. Illustrates spalling of corrosion product.

Figure 16 - Scanning electron micrograph of surface film on ASTM A27 specimen exposed six months in PBB2 with 2×10^3 rd/h

Figure 17 - Schematic of pitting corrosion specimen (drilled holes to simulate pits)

Figure 18 - Photograph of corrosion sample with artificial pits - exposed 4 months in PBB2 with 1×10^5 rd/h

Figure 19 - SSR data for 1025 steel in 150°C PBB2 brine and air - reduction in area (10 to 70 %) vs. strain rate (10^{-7} to 10^{-4}).

Figure 20 - SSR data for A27 cast steel - similar to figure 19

Figure 21 - SSR data for A27 cast steel in PBB2 brine at 150°C - energy absorbed (2 to 8 kg-m) with strain rates of 10^{-7} to 10^{-4}.

Figure 22 - SSR data for A27 cast steel in 150°C PBB2 sparged with Ar, Ar-20% O_2 and O_2 - energy absorbed (3 to 5 kg-m) versus solution oxygen content (relative values).

Figure 23 - SSR data for A27 cast steel in 150°C PBB2 - effect of pre-exposure on energy absorbtion (1 to 8 kg-m) at 10^{-4} strain rate.

Figures 24-26 - Scanning electron micrographs of A27 cast steel SSR test specimen fracture surfaces - illustrate different fracture modes, pitting and cracking associated with porosity defects.

Figure 27 - SSR data for A216 cast steel in air at 150°C - reduction in area and elongation (10 to 40%) versus strain rate (10^{-7} to 10^{-4}).

Figure 28 - Similar to figure 27 - data for PBB2 brine

Figure 29 - 1025 steel corrosion fatigue data for deionized water, air and PBB2 brine. Crack growth rate (10^{-8} to 10^{-4} m/cycle) vs. stress intensity (20 to 60 MPa$\sqrt{m}$)

Figure 30 - Similar to figure 29 - data for A216 cast steel in air and deionized water

Figure 31 - Similar to figure 29 - data for A216 is deionized water comparing frequencies of 0.1 and a Hz

Figure 32 - Similar to figure 29 - data for A216 in PBB2 brine and air

Figures 33a and 33b - Low magnification photographs of fatigue cracks in A216 specimens, before and after cleaning.

UNCERTAINTIES IN DATA

Author includes extensive statistical analysis of data reliability.

DEFICIENCIES/LIMITATIONS IN DATABASE

Author notes (1) controversy over use and existance of "threshold" stress intensity for corrosion failure predictions, (2) difficulties in extrapolation of short-term corrosion rates to significantly longer exposure periods, (3) complications due to corrosion product buildup in corrosion fatigue specimen notch of and (4) unexplained effects of sample location in irradiated autoclave tests.

KEYWORDS

data analysis, experimental data, corrosion, microscopy, visual examination, weight change, simulated field, laboratory, brine, brine (high ionic content), brine (low ionic content), ^{60}Co, high temperature, static (no flow), dynamic (flow rate given), steel, carbon steel, 1025 carbon steel, A216 grade WCA, A27 grade 60-30, 2.5Cr-1Mo, A536 grade 60-40-18, high-purity iron, cast, homogenized, normalized, slow strain rate, modified compact, precracked, chloride, corrosion (general), corrosion (pitting), fatigue (corrosion), cracking (environmentally assisted)

GENERAL COMMENTS OF REVIEWER

This is a detailed review of broad based corrosion studies to assess suitability of ferrous materials for use as waste package overpack. The program addresses general corrosion in simulated inclusion and intrusion brines, irradiation effects and environmental cracking based on slow strain rate and corrosion fatigue tests. The report acknowledges the potential importance of bacterial corrosion and hydrogen embrittlement in overall material degradation model but does not reference related data.

The report weakens arguments for use of corrosion fatigue studies to assess environmental cracking susceptibility. This is done by an acknowledgment of the controvery over the concept of a threshold stress intensity in a dynamic corrosion environment and of the problems with corrosion product buildup in the specimen notch. The buildup prevents crack closure during the low-stress portion of a fatigue cycle (this increases K_{min} and decreases ΔK).

The large majority of the tests on the reference cast steel (A216 grade WCA) were conducted with as-cast samples from a single casting. Statistical comparisons of corrosion data from multiple castings should be considered. The report discounts potential mechanical or corrosion related effects that could result from specific heat treatments. This is an area that needs better understanding to deliniate overpack fabrication restrictions.

Perhaps the most significant observation is the corrosion acceleration effects in the simulated PPB3 inclusion brine. These effects are attributed to the higher Mg^{2+} content. The significance of these effects and the corrosion mechanisms involved need detailed study.

RELATED HLW REPORTS

Westerman, R. E., Haberman, J. H., Pitman, S. G., and Perrin, J. S., "Corrosion of Iron-Base Waste Package Container Materials in Salt Environments," Nuclear Power Conference, Philadelphia, PA, PNL-SA-14029, July 20, 1986

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

(a) Relationship to Waste Package Performance Issues Already Identified

Related to ISTP issues 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? and 2.4, how and at what rates will radionucides migrate through failed waste package?

(b) New Licensing Issues

(c) General Comments

AUTHOR'S ABSTRACT

The disposal of high-level nuclear waste in deep underground repositories may require the development of waste packages that will keep the radioisotopes contained for time periods up to 1000 years. The primary geologic media currently being considered in the United States for repository siting are salt, basalt, tuff, and granite. A number of iron-base materials are being considered for the structural barrier members of waste packages. Their uniform and nonuniform (pitting and intergranular) corrosion behavior and their resistance to stress-corrosion cracking in aqueous environments relevant to salt media are under study at Pacific

Northwest Laboratory (PNL). The purpose of the work is to provide data for a materials degradation model that can ultimately be used to predict the effective lifetime of a waste package overpack in the actual repository environment. This report summarizes the results of the studies conducted at PNL during the FY 1983-FY 1984 time period in support of the Salt Repository Project of the Department of Energy.

The corrosion behavior of the candidate materials was investigated in simulated intrusion brine (essentially NaCl) in flowing autoclave tests at 150°C, and in combinations of intrusion/inclusion (high-Mg) brine environments in moist salt tests, also at 150°C. Studies utilizing a ^{60}Co irradiation facility were performed to determine the corrosion resistance of the candidate materials to products of brine radiolysis at dose rates of 2×10^3 and 1×10^5 rad/h and a temperature of 150°C. These irradiation-corrosion tests were "overtests," as the irradiation intensities employed were 10 to 1000 times as high as those expected at the surface of a thick-walled waste package.

Slow-strain-rate (SSR) tests and corrosion fatigue tests conducted in intrusion brine environments at 150°C and, in the case of some SSR tests, with a superimposed radiation field of 3×10^5 rad/h, were used to determine the resistance of the candidate alloys to environmentally enhanced crack propagation.

With the exception of the high general corrosion rates found in the tests using moist salt containing high-Mg brines, the ferrous materials exhibited a degree of corrosion resistance that indicates a potentially satisfactory application to waste package structural barrier members in a salt repository environment.

Draft Copy

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

DATA SOURCE

(a) Organization Producing Data

The Catholic University of America, Washington, D.C.

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

Barkatt, A., Macedo, P. B., Sousanpour, W., Boroomand, M. A., Szoke, P. and Rogers, V., "Aluminosilicate Saturation as a Solubility Control in Leaching of Nuclear Waste-Form Materials." Workshop on the Leaching Mechanisms of Nuclear Waste Forms, Summary Report, May 19-21, 1982,

DATE REVIEWED: 9/16/87

TYPE OF DATA

Experimental study of glass leaching.
Predictive model for solubility and leach rates.

MATERIALS/COMPONENTS

Five waste-form materials and various basalt, granite and kaolinite minerals.

TEST CONDITIONS

Glasses were in solid form for most tests; some static tests used crushed powders. Except for the powders, specimen preparation methods were not specified. Test environment was water at 70°C under slow flow conditions.

METHODS OF DATA COLLECTION/ANALYSIS

Chemical analysis of leachants, method unspecified.

AMOUNT OF DATA

Four tables:
1. Results of Dynamic Leach Tests on Various Waste Form Materials in the Slow Flow Region, 70°C. (Leachant concentration vs. residence time for five waste form glasses.)
2. Silica and Alumina Content of Various Nuclear Waste Solids and Rock Specimens, Weight Percent.
3. Composition of Rock Leachants in Laboratory Test and Nature.
4. Al:Si Mole Ratios in Solid and Leachate Compositions.

One figure:
1. AlxSi and $(AlxSi)^2$ Concentration Products in Saturated Leachates of Nuclear Waste-Form Materials and of Rock Materials at 70°C and in Basaltic Ground Water as a Function of pH.

UNCERTAINTIES IN DATA

None given by author.

DEFICIENCIES/LIMITATIONS IN DATABASE

Conclusions of the present paper require further experimental confirmation. Comprehensive analyses of leachants corresponding to very long contact times of waste forms with water and of aged groundwater from geologic formations are needed. The influence of low-solubility species on leachate saturation levels should be studied further.

KEYWORDS

experimental data, laboratory, PH, flow rate, significant dissolve species concentration, Al-Si, sorption, leaching

GENERAL COMMENTS OF REVIEWER

The fact that the solubility of silicon in aqueous media is lower when alumina is present in the system has been reported in the literature, and lower solubility of aluminum in the presence of silica has likewise been reported. [In this paper the authors specify the concentration product (C_{Al} x C_{Si}) by using either the symbols AlxSi or the symbol/term Al-Si concentration product.]

The primary objective of the paper is to demonstrate that the solubilities of various waste glasses and minerals are controlled by the product of the concentrations of aluminum and silicon in the leachate. The authors observe equimolar concentrations of these two elements in the leachate and they hypothesize that the solubilities of aluminum and silicon are controlled by readsorption of a combined (and equimolar) aluminosilicate (Al-Si) species. At a slow flow rate characteristic of geologic repositories, this product is governed by temperature and pH of the leachate. Thus, accurate predictions of long-term leach rate can be made for a repository providing the limits are known for pH and temperature. However, in slow flow conditions, which commonly lead to saturation for all of the main glass constituents, the minor glass constituents leach much more slowly. This decreased rate of leaching is due to the presence of the re-adsorbed Al and Si, which block the exposure (to the leachate) of the minor constituents .

The need for additional data, as stated by the authors, is well taken.

The technique that was used in this work to measure the surface area of the powder samples is very interesting and could be used in the MCC-3 procedure. This technique was carried out to compare the concentrations of leach products obtained with powder and with regularly-shaped solid blocks of the same glass and in the same conditions.

RELATED HLW REPORTS

Aa. Barkatt, A. Barkatt, P.E. Pehrsson, P. Szoke, P.B. Macedo, "Static and Dynamic Tests for the Chemical Durability of Nuclear Waste Glass", Nucl. Chem. Waste Management, Vol. 2, pp. 151-164, 1981.

APPLICABILITY OF DATA TO LICENCING

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

(a) Relationship to Waste Package Performance Issues Already Identified

Related to 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 forms under the range of repository conditions? 2.3.2.1, what are the possible dissolution mechanism 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? 2.3.2.1.1, what are the rates of dissolution associated with the potential form dissolution mechanism?

(b) New Licensing Issues

(c) General Comments

AUTHOR'S ABSTRACT

In the slow flow region, material loss rates of nuclear waste form materials are determined by the solubilities and the flow rates. Present studies show that the solubilities of various, widely different waste forms, as well as of minerals, can be correlated with the AlxSi concentration products. This indicates that in these cases the solubilities are controlled by a combined equimolar aluminosilicate species. This observations serves as a basis for a predictive model for the long-term stability of waste forms under slow flow conditions. This model also provides explanations of other experimental finings, such as the increases in solubility upon departure from a neutral pH in the low as well as in the high PH region, the small magnitude of the temperature dependence, and the observation that the release rate of Cs is low relative to that of Na in the slow flow region. The relative concentrations of Si and of Al, respectively, in the leachates are related to the composition of the leach solids and are shown to depend on the immersion time in different ways in the cases of high-silica solids and of high-alumina solids, respectively. In both cases, however, the dependence of the Si:Al ratio on contract time furnishes another strong indication for the formation of a solid surface with comparable contents of Al and of

Si, respectively, upon prolonged immersion in water. The results are shown to form the basis for an accurate long-term prediction of material loss rates.

PDR-1
LPDR WM-10(2)
WM-11(2)
WM-16(2)

WM DOCKET CONTROL
CENTER

'87 OCT 19 P2:46

WM RES
WM Record File
A4171
NBS

WM Project 10, 11, 16
Docket No.
x PDR
x LPDR (B, N, S)

Distribution:
x Wick
x Donn - Tickets

(Return to WM, 623-SS)

4219