UNITED STATES DEPARTMENT OF COMMERCE
National Institute of Standards and Technology
Gaithersburg, Maryland 20899

August 22, 1990

Dr. Charles Interrante
Materials Engineering Section
Division of High-Level Waste Management
Office of Nuclear Materials Safety and Safeguards
U.S. Nuclear Regulatory Commission
Washington, DC 20555

Re: Progress Report for June and July 1990 (FIN-A-4171-0)

Dear Dr. Interrante:

Enclosed is the June and July 1990 progress report for the project "Evaluation and Compilation of DOE Waste Package Test Data" (FIN-A-4171-0). The financial information is reported separately.

Sincerely,

Anna C. Fraker

Anna C. Fraker
Metallurgist
Corrosion Group
Metallurgy Division

Enclosures

Distribution:
WM Docket Control Center (1-original)

A-4171
WM-11
NH14

Progress Report for June - July 1990

Published August 1990

(FIN-A-4171-0)

Performing Organization: National Institute for Standards and Technology (NIST)
Gaithersburg, MD 20899

Sponsor: Nuclear Regulatory Commission (NRC)
Office of Nuclear Materials Safety and Safeguards
Washington, DC 20555

TASK 1 -- REVIEW OF WASTE PACKAGE DATA BASE

Status of Database

	Current Report	Previous Report
Number of citations	1397	1298
Number of completed reviews	100	100

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

1. UCRL-90857, "Parametric Testing of a DWPF Borosilicate Glass", January 1985.

2. UCRL-101615, "Prototype Engineered Barrier System Field Tests Progress Report", July 1989.

3. BNL-52231, UC-810, "^{14}C Release From Failed Spent Fuel Containers", May-September 1989.

4. Bates, J.K., Ebert, W.L., Fischer, D.F., Gerding, T.J.,"The Reaction of Reference Commercial Nuclear Waste Glasses During Gamma Irradiation in a Saturated Tuff Environment", J. Mater. Res., 3(3), May/June 1988.

Status of Recently Listed Reviewable Documents

Reviewable documents are classified as follows: Category 1 documents are currently being reviewed. Categories 2 and 3 are documents that will be entered into the database with citation information and authors abstracts, and the Category 2 documents are flagged "to review when time permits." The number of documents in each category on the Yucca Mountain Project and the Glass Vitrified Waste Form are given below.

YUCCA MOUNTAIN PROJECT

8 Reports currently under review (Category 1).
31 Reports to review when time permits (Category 2).
0 Reports to file with cross reference(s) to other reports (Category 3).
12 Reports identified and not yet categorized.
5 Reports received and not yet categorized.

GLASS -- VITRIFIED WASTE FORM

0 Reports currently under review (Category 1).
4 Reports to review when time permits (Category 2).
0 Reports to file with cross reference(s) to other reports (Category 3).
0 Reports identified and not yet categorized.

Database searches for April and May 1990 include Metadex, NTIS, Engineered Materials Abstract, Compendex Plus and DOE Energy. Examples of the search conducted for each of these databases are in this report (see p. 8).

STATUS OF REVIEWS OF YUCCA MOUNTAIN PROJECT REPORTS

Reports recently identified for review pertaining to the Yucca Mountain Project --

Five reports have been identified for review during this two month period. Two of these reports are on the subject of the performance of copper and its alloys, one is on container performance, and two are on the tuff environment.

Copper

Schweitzer, D.G. and Sastre, C.A.
"Long-Term Isolation of High-Level Radioactive Waste in Salt Repositories Containing Brine"
Nuclear Technology, Vol. 86, p305, September 1989

A thermodynamic analyses of the conditions expected in a salt repository indicates that dissolution of copper will be controlled by the amount of oxygen present, and in an anoxic repository this dissolution is negligible [Schweitzer]. Furthermore, copper reacting with acid brines at constant volume generates hydrogen gas stifling further reaction.

Kundig, K.J.A., Lyman, W.S., and Prager, M.
"Background Studies in Support of a Feasibility Assessment on the Use of Copper-Base Materials for Nuclear Waste Packages in a Repository in Tuff"
UCRL-21082, June 1990.

This is a report of a workshop/Seminar on "Copper-Base Waste Package Container Materials" and a review of five studies, all sponsored by the Copper Development Association, considering the use of copper as a material for fabricating high-level waste packages [Kundig]. The general conclusion is that copper-base materials are viable candidates for this application.

Container Performance

Halsey, W. G.
"Selection Criteria for Container Materials at the Proposed Yucca Mountain High Level Nuclear Waste Repository"
UCRL-102285, November 1989

A draft list of seven topics to be used as the basis for developing a set of selection criteria is described [Halsey]. The selection criteria are divided into two major subjects, 1) Material Performance and 2) Fabricability, Cost, and Other Considerations. The topic with the highest relative weighing factor (30) is the corrosion performance of the canister (chemical performance). The two topics with the lowest relative weighing factors (5) are Cost and Previous Experience With The Material.

Tuff Environment

Latorre, V.
"Microwave Measurements of Water Vapor Partial Pressure at Temperatures up to 350 C"
UCRL-101866, September 1989

The amount of moisture available in the Tuff atmosphere at repository temperatures is important in the evaluation of corrosion degradation of the nuclear waste container [Latorre]. This report describes a new technique under development that can measure the vapor pressure of water at temperatures up to 350°C.

Lin, W. and Daily, W.D.
"Laboratory Study of Fracture Healing in Topopah Spring Tuff - Implications for Near Field Hydrology"
UCRL-100624, September 1989

The permeability of the tuff, at repository temperatures, is of importance because permeability will determine the rate of moisture and oxygen ingress to the waste containers, both of which are important parameters in the corrosion of the waste container [Lin]. This study shows that raising the temperature of the Tuff to 150°C, decreases its moisture permeability by three orders of magnitude.

Category 1 -- Reports currently being reviewed;

1. WHC-EP-0096 (formerly HEDL-7665), "Initial Report on Stress-Corrosion-Cracking Experiments Using Zircaloy-4 Spent Fuel Cladding C-Rings," September 1988.

2. UCRL-100395, "Waste Package Performance Assessment for the Yucca Mountain Project", February 1989.

3. UCRL-21076, "Electrochemical Corrosion Studies on Copper-Based Waste Package Container Materials in Unirradiated 0.1 N $NaNO_3$ at 95°C", May 1988.

4. NUREG/CR-5001, "Effect of Manufacturing Variables on Performance of High-Level Waste Low Carbon Steel Containers," April 1990.

5. UCRL-100211, "Modeling of Zircaloy Cladding Degradation Under Repository Conditions," July 1989.

6. Schweitzer, D.G. and Sastre, C.A., "Long-Term Isolation of High-Level Radioactive Waste in Salt Repositories Containing Brine" Nuclear Technology, Vol. 86, p305, September 1989.

7. UCRL-102285, "Selection Criteria for Container Materials at the Proposed Yucca Mountain High Level Nuclear Waste Repository", November 1989.

8. UCRL-21082, "Background Studies in Support of a Feasibility Assessment on the Use of Copper-Base Materials for Nuclear Waste Packages in a Repository in Tuff", June 1990.

Category 1 (continued) - Status of Reviews not yet sent to NRC and WERB

Document No.	Assigned to Reviewer	First Draft Completed	Lead Worker	Program Manager
WHC-EP-0096	2/21/89			
UCRL-100395	12/18/89			
UCRL-21076	2/2/90		7/30/90	
NUREG/CR-5001	5/31/90	7/31/90		
UCRL-100211	5/31/90			
Schweitzer, 1989	6/27/90			7/27/90
UCRL-102285	7/23/90			
UCRL-21082	7/31/90			

Category 2 -- Review as time permits (new entries for this reference data file)

None this quarter.

Category 3 -- File and cross reference

1. UCRL-101866, "Microwave Measurements of Water Vapor Partial Pressure at Temperatures Up to 350 C.

2. UCRL-100624, "Laboratory Study of Fracture Healing in Topopah Spring Tuff - Implications for Near Field Hydrology", September 1989.

OTHER REPORTS INCLUDES VITRIFIED WASTE FORM --

Category 1 -- Reports currently being reviewed

None this quarter.

Category 2 -- Review as time permits

None this quarter.

Category 3 -- File and cross reference

None this quarter.

TASK 3 -- LABORATORY TESTING

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

June - July, 1990

The abstract of the paper entitled "The Effect of Oxygen Transport and Resistivity of the Environment on the Rate and Form of Corrosion of Steel in Simulated Soils" was submitted and has been accepted by the Materials Research Society. The paper is directed to the Symposium on the Scientific Basis for Nuclear Waste Management meeting this November in Boston, MA.

Title of Study: The Underground Corrosion of Stainless Steels - A Twenty Year Study.
Principal Investigator: Edward Escalante

Contact with personnel in charge of site B, located in Baltimore County, was established, and a visit to the site confirmed that the test area is intact. At this point, four of the six underground test sites have been visited and confirmed as undisturbed and accessible for specimen retrieval. The two sites that have not been visited are site A, in the state of Washington, and site G, in the Southern end of Maryland. Cost estimates are being obtained on rental of equipment needed for excavation and retrieval of specimens.

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

June - July, 1990

The purpose of this study is to provide information and data on the corrosion behavior of Zircaloy that can be used to determine the long-term durability of nuclear fuel cladding made of this material. Zircaloy obtains its corrosion resistance by the formation of a protective oxide surface film. All of the work in this task relates to the formation or breakdown of this film. This experimental work involves electrochemical measurements made primarily using potentiostatic polarization techniques to study the corrosion behavior of bulk Zircaloy-2 and -4 as well as specimens of cladding tubes made from these two materials. Measurements are made on both the inner and outer walls of the cladding, and most measurements have been made at 95°C. Measurements have been made in simulated J-13 water, a water that may be typical of that which could be present in the Yucca Mountain, Nevada site.

The current phase of this study is to investigate the effects of the fluoride, chloride and iodide ions on the passive film and on the electrochemical behavior. There is concern regarding the effects of these ions on the zirconium oxide film. Initially, this work will be conducted at 23°C for the purpose of acquiring reproducible data to determine effects of the fluoride ion on zirconium oxide. These data can also to be used in analyzing measurements taken at higher temperatures where experimental procedures are more difficult.

During this reporting period, tests have been completed in 0.1 M solutions of each of the following salts; NaF, NaCl and NaI. Some of the tests were of Zircaloy cladding tubes and other tests were of extruded Zircaloy material. Extruded Zircaloy was tested in all solutions. One additional set of tests will be made to show effects of being exposed in the solution for a few weeks time. Then the data will be summarized and submitted in the August-September report. Indications from the brief analytical work so far indicate that effects of the NaF solution on the electrochemical behavior of Zircaloy are more detrimental than effects of solutions of the NaCl or NaI.

SDI008, UD 9007, SER. DA016

File(s) searched:

File 8:COMPENDEX PLUS _ 70-90/JUL Copr. Engineering Info
Inc. 1990)

Sets selected:

Set	Items	Description
1	1	WASTE(W)PACKAGE?
2	5	CANISTER?
3	173	CORROSION
4	25	LEACHING
5	418	GLASS
6	3	VITRIFICATION
7	606	S3-S6/OR
8	1	HIGH(W)LEVEL(W)WASTE?
9	11	RADIOACTIVE(W)WASTE?
10	4	NUCLEAR(W)WASTE?
11	1	(S1 OR S2) AND S7 AND (S8 OR S9 OR S10)
12	0	ANNA FRAKER RM. B-106 BLDG. 223 X6009
13	0	JILL RUSPI

Prints requested ('*' indicates user print cancellation) :

Date	Time	Description
28jun	07:28EST	PR 11/5/1-20 (items 1-1)

Total items to be printed: 1

SDI293, UD 9007, SER. DD023

File(s) searched:

File 293:ENGINEERED MATERIALS ABS_86-90/JUL
(COPR. 1990 ASM INTERNATIONAL)

Sets selected:

Set	Items	Description
1	0	HIGH()LEVEL()WASTE? ? OR RADIOACTIVE()WASTE? OR NUCLEAR()WASTE?
2	276	STEEL? ? OR ZIRCALOY? ? OR TITANIUM? ? OR COPPER
3	0	S1*S2
4	0	ANNA FRAKER, 223, B-254, X6009

Prints requested ('*' indicates user print cancellation) :

Date	Time	Description
28jun	05:35EST	PR 3/5/1-25 (no items to PRINT)

Total items to be printed: 0

SDI006, UD 9014, SER. DD016

File(s) searched:

File 6:NTIS - 64-90/ISSUE14
(COPR. 1990 NTIS)

Sets selected:

Set	Items	Description
1	1	WASTE(W)PACKAGE?
2	4	CANISTER?
3	51	CORROSION
4	5	LEACHING
5	68	GLASS
6	2	VITRIFICATION
7	118	S3-S6/OR
8	8	HIGH(W)LEVEL(W)WASTE?
9	104	RADIOACTIVE(W)WASTE?
10	17	NUCLEAR(W)WASTE?
11	3	(S1 OR S2) AND S7 AND (S8 OR S9 OR S10)
12	0	ANNA FRAKER RM. B-106 BLDG. 223 X6009
13	0	JILL RUSPI

10

Prints requested ('*' indicates user print cancellation) :

Date	Time	Description
21jun	23:56EST	PR 11/5/1-25 (items 1-3)

Total items to be printed: 3

SDI103, UD 9011, SER. DD017

File(s) searched:

File 103:DOE ENERGY _ 83-90/JUN(ISS11)

Sets selected:

Set	Items	Description
1	1	WASTE(W)PACKAGE?
2	12	CANISTER?
3	151	CORROSION (1974 DEC)
4	45	LEACHING (1974 DEC)
5	122	GLASS (1974 DEC)
6	8	VITRIFICATION (1974 DEC)
7	308	S3-S6/OR
8	10	HIGH(W)LEVEL(W)WASTE?
9	215	RADIOACTIVE(W)WASTE?
10	31	NUCLEAR(W)WASTE?
11	1	(S1 OR S2) AND S7 AND (S8 OR S9 OR S10)
12	0	ANNA FRAKER RM. B-106 BLDG. 223 X6009
13	0	JILL RUSPI

Prints requested ('*' indicates user print cancellation) :

Date	Time	Description
20jun	19:48EST	PR 11/5/1-25 (items 1-1)

Total items to be printed: 1

SDI008, UD 9006, SER. DA016

File(s) searched:

File 8:COMPENDEX PLUS _ 70-90/JUN Copr. Engineering Info Inc. 1990)

Sets selected:

Set	Items	Description
1	1	WASTE(W)PACKAGE?
2	2	CANISTER?
3	183	CORROSION
4	34	LEACHING
5	254	GLASS
6	7	VITRIFICATION
7	461	S3-S6/OR
8	2	HIGH(W)LEVEL(W)WASTE?
9	20	RADIOACTIVE(W)WASTE?
10	15	NUCLEAR(W)WASTE?
11	1	(S1 OR S2) AND S7 AND (S8 OR S9 OR S10)
12	0	ANNA FRAKER RM. B-106 BLDG. 223 X6009
13	0	JILL RUSPI

12

Prints requested ('*' indicates user print cancellation) :

Date	Time	Description
02jun	06:55EST	PR 11/5/1-20 (items 1-1)

Total items to be printed: 1

SDI032, UD 9007, SER. DD022

File(s) searched:

File 32:METADEX_66-90/JUL
(COPR. 1990 ASM INTERNATIONAL)

Sets selected:

Set	Items	Description
1	4	HIGH()LEVEL()WASTE? ? OR RADIOACTIVE()WASTE? OR NUCLEAR()WASTE?
2	1653	STEEL? ? OR ZIRCALOY? ? OR TITANIUM? ? OR COPPER
3	2	1*2
4	0	ANNA FRAKER, 223, B-254, X6009

Prints requested ('*' indicates user print cancellation) :

Date	Time	Description
16jun	03:08EST	PR 3/5/1-25 (items 1-2)

Total items to be printed: 2

13

SDI103, UD 9010, SER. DD017

File(s) searched:

File 103:DOE ENERGY _ 83-90/JUN(ISS10)

Sets selected:

Set	Items	Description
1	7	WASTE(W)PACKAGE?
2	12	CANISTER?
3	212	CORROSION (1974 DEC)
4	47	LEACHING (1974 DEC)
5	147	GLASS (1974 DEC)
6	17	VITRIFICATION (1974 DEC)
7	398	S3-S6/OR
8	13	HIGH(W)LEVEL(W)WASTE?
9	205	RADIOACTIVE(W)WASTE?
10	24	NUCLEAR(W)WASTE?
11	2	(S1 OR S2) AND S7 AND (S8 OR S9 OR S10)
12	0	ANNA FRAKER RM. B-106 BLDG. 223 X6009
13	0	JILL RUSPI

14

Prints requested ('*' indicates user print cancellation) :

Date	Time	Description
02jun	18:52EST	PR 11/5/1-25 (items 1-2)

Total items to be printed: 2

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

DATA SOURCE

(a) Organization Producing Data

Lawrence Livermore National Laboratory for the
U. S. Department of Energy
Contract No. W-7405-Eng-48.

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

Bazan, F. and Rego, J.
"Parametric Testing of a DWPF Borosilicate Glass"
UCRL-90857, January 1985.

DATE REVIEWED: 2/20/90

PURPOSE

"To characterize the chemical stability of a DWPF borosilicate glass sample".

KEY WORDS

Experimental data, spectroscopy, simulated field, air, J-13 Water, distilled water, tuff, high temperature, static (no flow), dissolution.

CONTENTS

The report has 8 pages, with 4 tables and two figures.

AMOUNT OF DATA

Table 1 - Composition of glass
Table 2 - Dissolution of DWPF glass in Deionized water
Table 3 - Dissolution of crushed DWPF glass
Table 4 - Leach of Li

Figure 1 is a comparison of leachates for monolithic vs crushed tuff, and Figure 2 is the leachate of Li vs time x surface area volume.

TEST CONDITIONS

"The parameters investigated were leachant composition, ratios of waste form surface area to water volume (SA/V), effects of the presence of crushed tuff in some tests and crushed tuff and stainless steel in others, and leaching times ranging from 1 to 182 days. All tests were conducted at 90°C and were of a static nature".

UNCERTAINTIES IN DATA

Standard deviations are tabulated for glass composition, and a few listed for the leach rates in Table IV.

DEFICIENCIES/LIMITATIONS IN DATABASE

None given.

CONCLUSIONS OF AUTHOR

"First, it appears that glass behavior is very much leachant-type dependent, since the leach rates in deionized water are about one order of magnitude higher than in J-13 water. Secondly, it is not clear that the additions of tuff in Test II and Tuff and stainless steel in Test IV have any effect on the Li leach rates; these appear to be about the same as the Test II with the exception of the day-3 samples."

COMMENTS OF REVIEWER

The results of this type of dissolution experiment are necessary for the modeling programs under development. However, it is difficult to interpret data where such a variety of parameters are changing. Because of this difficulty, the authors can only make broad, general conclusions, leaving important questions unanswered. For example, based on the results, it is unclear whether dissolution of the elements is congruent. What is the effect of metal ions in the leachate solution on the dissolution of glass. The results of these short term tests suggest that dissolved metal ions have no effect on the leach rate of the glass, but in the long exposure burial times in the repository, the effect of the metal ions may become significant. The authors state that this report contains a very small portion of the data collected, suggesting that further analyses of the results will yield a better understanding of waste glass leaching.

Finally, some procedure must be enacted that will verify incorporation of all the nuclear waste sludge into the glass. Heterogeneous distribution of waste or localized pockets of undissolved sludge in the glass will seriously affect the expected leaching of radionuclides.

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

(a) Relationship to Waste Package Performance Issues Already Identified

2.3.2 What is the solubility of the waste form under the range of potential repository conditions?

(b) New Licensing Issues

(c) General Comments

AUTHOR'S ABSTRACT

A series of tests have been performed to characterize the chemical stability of a DWPF borosilicate glass sample as part of the Waste Package Task of the NNWSI Project. This material was prepared at the Savannah River Laboratory for the purpose of testing the 165-frit matrix doped with a simulated non-radioactive waste. All tests were conducted at 90°C using deionized water and J-13 water (a tuffaceous formation groundwater). In the deionized water tests, both monoliths and crushed glass were tested at various ratios of surface area of the sample to volume of water in order to compare leach rates for different sample geometries or leaching times. Effects on the leach rates due to the presence of crushed tuff and stainless steel material were also investigated in the tests with J-13 water.

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

DATA SOURCE

(a) Author(s), Reference, Reference Availability

Ramirez, A. L., Beatty, J., Buscheck, T., Carlson, R., Daily, W., LaTorre, R., Lee, D., Lin, W., Mao, N., Titao, J., Towse, D., Ueng, T., Watwood, D., and Wilder, D.
"Prototype Engineered Barrier System Field Tests Progress Report"
UCRL-101615, July 1989.

(b) Organization Producing Data

Lawrence Livermore National Laboratory, Livermore, CA.

DATE REVIEWED: 7/5/90

PURPOSE

This paper describes the results of the first prototype test to "study the hydrothermal perturbation of welded tuff near a horizontally oriented heater. The tests measured several parameters as a function of location and time to examine the effects of heating and cooling during a thermal pulse that lasted approximately 28 weeks".

KEYWORDS

Experimental data, field, ambient temperature, temperature distribution, moisture distribution, welded tuff, Rainier Mesa.

CONTENTS

7 pages of text, including 12 figures, and 10 references.

AMOUNT OF DATA

A limited amount of data on moisture content and temperature distribution around the borehole measured during the first 30 days after heat was applied to the borehole.

TEST CONDITIONS

An underground facility consisting of drifts in welded tuff under Rainier Mesa, NV, with properties similar to those expected for the Topopah Springs welded tuff unit in Yucca Mountain.

UNCERTAINTIES IN DATA

It is speculated that the rock fractures increase vapor permeability, affecting the distribution of moisture and heat.

DEFICIENCIES/LIMITATIONS IN DATABASE

None given.

CONCLUSIONS OF AUTHOR

"The test confirmed elements of the conceptual model of predicted environmental conditions. Test results confirm that a dry zone develops around the heater borehole, and the degree of drying increases with proximity to the heater. A "halo" of increased saturation develops adjacent to the dry region and migrates away from the heater as rock temperatures increase. Some of the fractures intercepting the heater borehole increase the penetration of hot-dry conditions into the rock mass. A build-up of pore gas pressure develops in rock regions where vigorous evaporation is occurring. The air permeability of the fracture system exhibits a strong heterogeneity.

"The test also yielded some surprises in terms of environmental conditions. The temperature above the heater container is approximately 30°C (54°F) higher than below the container. This condition might be a consequence of hotter air accumulating at the top of the container; it might also be related to the higher moisture content present below the heater borehole. The amount of steam predicted by scoping calculations to invade the heater borehole is much less than that expected. The reason(s) for this discrepancy is not known at present; it might be a consequence of an inadequate system used to collect and condense the steam or a result of the calculation's assumption that the heater was infinitely long."

COMMENTS OF REVIEWER

The study described is a very important part of the Yucca Mountain Project in that it is a field evaluation of the effect of borehole heating on moisture distribution. Controlled experiments in the laboratory are extremely important in developing detailed information on phenomenon observed in the field, but the field studies must be performed to provide direction for laboratory studies and, ultimately, verification of laboratory results.

These preliminary indicate that heating of the borehole drives moisture away, forming a "halo" of increased moisture ahead of the heated region. Presumably, future measurements will show dissipation of this "halo" of moisture. Fractures in the vicinity of the borehole cause heterogeneity of moisture and temperature. A major uncertainty in this study is the effect of rock fractures on moisture diffusion.

RELATED HLW REPORTS

1. Zimmerman, R. M. and Finley, R. E., "Summary of Geomechanical Measurements Taken in and Around the G-Tunnel Underground Facility, NTS," SAND86-1015, Sandia National Laboratories, Albuquerque, NM, 143p (1986).

2. Daily, W., Lin, W., and Buscheck, T., "Hydrology of Topopah Spring Tuff - Laboratory Measurements," J. Geophys. Res. 92(B8), pp. 7854-7864 (1987).

APPLICABILITY OF DATA TO LICENSING

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

(a) Relationship to Waste Package Performance Issues Already Identified

2.1.2 What will be the physical characteristics (e.g., temperature, pressure, phase, and flow rates) of the groundwater reaching the waste package container as a function of time?

(b) New Licensing Issues

(c) General Comments

AUTHOR'S ABSTRACT

This paper presents selected preliminary results obtained during the first 54 days of the Prototype Engineered Barrier System Field Tests (PEBSFT) that are being performed in G-Tunnel within the Nevada Test Site. The test described is a precursor to the Engineered Barrier Systems Field Tests (EBSFT). The EBSFT will consist of in situ tests of the geohydrologic and geochemical environment in the near field (within a few meters) of heaters emplaced in welded tuff to simulate the thermal effects of waste packages. The PEBSFTs are being conducted to evaluate the applicability of measurement techniques, numerical models, and procedures for future investigations that will be conducted in the Exploratory Shaft Facilities of the Yucca Mountain Project (YMP). The paper discusses the evolution of hydrothermal behavior during the prototype test, including rock temperatures, changes in rock moisture content, air permeability of fractures, gas pressures, and rock mass gas-phase humidity.

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

DATA SOURCE

(a) Organization Producing Data

Brookhaven National Laboratory, Upton, Long Island, New York 11973

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

Pescatore, C.
"^{14}C Release from Failed Spent Fuel Containers," Progress Report
BNL-52231, UC-810, May-September, 1989

DATE REVIEWED: 6/13/90

PURPOSE

"A modeling approach is outlined and sample calculations are provided that show the effect of "release due to a limited perforation area" of containers, "to decreasing temperature, and to partial occlusion of the perforated area by corrosion products" on the release of $^{14}CO_2$.

KEYWORDS

Theory, gas diffusion, air, carbon dioxide, ^{14}C, ambient temperature, perforated containers with a partial occlusion by corrosion products, diffusion.

CONTENT

This report consists of an abstract, an introduction, 4 tables, and the following content:

CONTENT	NUMBER OF PAGES
Modeling Formalism	4
Numerical Results	1
Conclusions	1
References (5)	1

TEST CONDITIONS

Materials: Spent-fuel
Specimen Preparation: N/A
Environments: Gaseous environments at temperatures of 250, 190, 150 and 80°C

UNCERTAINTIES IN DATA

The thickness of container wall is ~ 1cm.

"The diffusion coefficient of $^{14}CO_2$ in air was taken as the same as that of CO_2 in nitrogen."

"The gas volume within the container was taken to be $1m^3$."

"We assume, conservatively, that as soon as the perforation penetrates the container all the oxygen needed to oxidize the ^{14}C is instantly available and instantly replenished."

"Assume that 70,000 MTHM of spent fuel have to be disposed of, all in the form of PWR 17x17 assemblies with a burnup of 33,000 MWd/MTHM."

Assume "there are six spent fuel assemblies per container."

DEFICIENCIES/LIMITATIONS IN DATABASE

"Further work is needed to incorporate 'breathing' of the container caused by barometric pressure variations in the outside environment and the effect of occasional pressure surges due to failing fuel rods."

"The relevant expressions for ^{14}C release should be convoluted with the container breach rate distribution functions that are being developed within the waste management community."

"The results of the ^{14}C release analyses from failed containers could be also verified via gas diffusion experiments."

"The dependence" of "the diffusion mass flux away from the container" "on the hole perforation area is negligible."

CONCLUSIONS OF AUTHORS

"Our preliminary analysis supports the assumption that partly failed containers may offer a significant delay to gaseous ^{14}C release both during the substantially complete containment and the controlled release periods."

So far, our analysis can model (a) the finite amount of ^{14}C present in the spent fuel container, (b) container wall thickness, (c) perforated area equivalent diameter and growth rate, (d) degree of occlusion of the perforation due to corrosion products, (e) thermal oxidation rate of the ^{14}C into $^{14}CO_2$, (f) gaseous diffusion of the $^{14}CO_2$ in the (partly occluded) hole(s), and (g) radioactive decay. All rate processes can be implemented with their temperature dependence. Further work is needed to incorporate "breathing" of the container caused by barometric pressure variations in the outside environment and the effect of occasional pressure surges due to failing fuel rods. Once these analyses are completed, the relevant expressions for ^{14}C release should be convoluted with the container breach rate distribution functions that are being developed within the waste management community. One such analysis will identify the allowable container perforation area in order to meet the applicable performance criteria.

The results of the ^{14}C release analyses from failed containers could be also verified via gas diffusion experiments, which are amply within reach of available technology.

The approach proposed here for gaseous radionuclide release can also be proposed for the release of radionuclides potentially leached by groundwater infiltrating a breached container. The likely presence of crud in container penetrations may allow only very small releases regardless of radionuclide solubilities. This would be an important effect during the NRC-regulated (10 CFR 60) controlled release period."

COMMENTS OF REVIEWER

This paper deals with release of ^{14}C from spent fuel waste and discusses meeting the Nuclear Regulatory Commission (NRC) requirement for substantially complete containment that permits an annual release of 10^{-6} of the total ^{14}C inventory. Roughly 38% of the ^{14}C resides in the UO_2 pellets and 62% is in the rest of the cladding assembly. The half life of ^{14}C is 5730 years. Calculations show that for 25,285 containers with six spent fuel assemblies each, not even one container per year could be allowed to fail. A model is presented to show that a small perforation relative to the total surface of the container would result in ^{14}C release that is slow enough to allow more than one container per year to fail and to meet the NRC requirement in 10 CFR 60. The authors state that prompt oxidation of the ^{14}C to $^{14}CO_2$ is expected. A model is developed to show that the release will be affected by diffusion, time and relative size of the perforation. Effects of temperature also are incorporated into the model. Experimental data are needed to support the model. The authors suggest that one method of verification would be with gas diffusion experiments. ^{14}C can be produced by the transmutation of nitrogen, and this would have to be added to the amounts from other sources.

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

(a) Relationship to Waste Package Performance Issues Already Identified

2.3.6.3 How will defected cladding after radionuclide release?

(b) New Licensing Issues

(c) General Comments on Licensing

ABSTRACT

Partially failed containers may provide a meaningful barrier to the release of gaseous $^{14}CO_2$. A modeling approach is outlined and sample calculations are provided that show the effect on release due to a limited perforation area, to decreasing temperature, and to partial occlusion of the perforated area by corrosion products.

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

DATA SOURCE

(a) Author(s), Reference, Reference Availability

Bates, J. K., Ebert, W. L., Fischer, D. F., and Gerding, T. J.
"The Reaction of Reference Commercial Nuclear Waste Glass During Gamma Irradiation in a Saturated Tuff Environment"
J. Mater. Res., 3(3), 576, May/June 1988.

(b) Organization Producing Data

Chemical Technology Division, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439-4837.

DATE REVIEWED: 7/13/90

PURPOSE

To determine the influence of gamma radiation in the presence of water and air on the dissolution of nuclear glass waste forms.

KEY WORDS

Data analysis, experimental data, weight change, laboratory, air, J-13 water, tuff composition, tuff, cobalt 60, acidic solution (pH <7), basic (alkaline) solution (pH >7), high temperature. PNL 76-68, ^{237}Np, ^{239}Pu, ^{99}Tc.

CONTENTS

21 pages, 12 figures, 8 tables; Sections: I.Introduction, 1.5 pgs. II. Experimental, A. Experimental matrix, B. Experimental components, C. Sample description, 2 pgs., III. Analyses, 1/4 pg., IV. Results, Solution analyses-blank, B. Solution analyses-ATM glasses, 1. pH, 2. Cations, 3. Anions, 4. Actinides,C. Component analyses, 1. Weight loss, 2. SEM/EDS, 3. SIMS, 9 pgs., V. Discussion A. pH, B. Actinide release, C. Reacted glass, D. Parameter effects and comparison with other results, 1 1/2 pgs, VI. Conclusions, 1/2 pg., 30 references

AMOUNT OF DATA

Tables; 1. Composition of EJ-13 water, ppm; 2. Composition of ATM glasses used in testing, wt%; 3. Nitrate and nitrite measurements, ppm; 4. pH and normalized elemental release for 12 glass components as function of radiation level and time; 5. Concentrations of B, Si, and U in 2R experiments; 6. Np and Pu release from ATM-8 glasses; 7. Measured layer thicknesses for ATM-1c glass; 8. X-ray diffraction powder pattern of the reacted layer formed on ATM 1-c glass; Figures; 2-4 pH (6-9) vs time in days for different radiation levels; 5-6, Normalized release of Pu, Np, and U; 7-12, Photomicrographs, EDS spectra,and line profiles, and SIMS profiles of reacted or unreacted glass surfaces or cracks.

TEST CONDITIONS

Two glasses based on PNL 76-68 were studied. ATM-1c glass was doped with uranium. ATM-8 glass was doped with uranium plus ^{237}Np, ^{239}Pu and ^{99}Tc. The following matrixes were run in duplicate using either of these glasses.

Matrix 1 data obtained at $2E^5$ R/h (2R experiments) were:(1) two glass disks in EJ-13 water, $S/V = 0.3cm^{-1}$; (2) two glass disks in EJ-13 water, $S/V = 0.3cm^{-1}$ with 0.2 g(<100 mesh) crushed tuff; (3) crushed glass (+40 -80 mesh) in EJ-13 water, $SA/V = 0.7\ cm^{-1}$. Each matrix was run for 7, 14, 28, and 56 days.

Matrix 2 data obtained at $1.E^3$ R/h (1R experiments) were a duplicate of (1) above, and (2) two glass disks, $S/V = 0.3cm^{-1}$,in EJ-13 water with a 2.54 cm diam tuff wafer. Each of these matrixes were run for 28, 56, 91, 182, and 278 days.

Matrix 3 data was obtained at 0 R/h (0R experiments). These included a duplicate of (1) above, a duplicate of (2) above using the 2.54 cm tuff wafer and a second duplicate of (1) using a silicone gasket material. These were run for 7 14 28 and 56 days.

Blanks were run for matrices 1 and 3 using only EJ-13 water, and for matrix 2 using EJ-13 water and tuff.

Samples were sealed in vessels containing both a compression fitting and a silicone gasket seal. Blank experiments were run using teflon gaskets in addition to the one sample using a silicone seal. Teflon gaskets were not used under radiation conditions because of contamination of leachates by fluoride ions. Silicone gaskets have the potential of introducing Si or organic carbon into the leachate during the experiment or post test handling procedures.

Other components included the vessel and glass specimen support stand which was 304 stainless steel, 16 ml of EJ-13 water and 4 cm^3 of air.
EJ-13 water is J-13 water which has been pre-equilibrated with tuff at 90°C.

UNCERTAINTIES IN DATA

It is stated that " In general, the precision of the solution analytical measurements was better than 10%, while radioactivity counting procedures (Np and Pu), which in some instances were affected by count rates that were near background levels, produced data of 20% precision."

DEFICIENCIES/LIMITATIONS IN DATABASE

Experimental difficulties were encountered (mainly in the 2.E5 R/h experiments) due to (1) dissolved gas in the leachate, (2) the extrusion and hardening of the silicone rubber gaskets, and (3) the dispersion of the crushed tuff during the test setup.

CONCLUSIONS OF AUTHOR

In the presence of radiation, the leachate pH is controlled by a balance between the rate of formation of nitrogenous acids, the extent of the glass reaction, the buffering capacity of the leachate and the amount of precipitation of secondary phases.

"These experiments demonstrate that the final leachate pH, the behavior of actinide elements in solution, and the nature of the reacted glass surface and associated reaction layer vary as a function of exposure rate, but that for the duration of comparable experiments through 56 days, the release of soluble glass components (B, Mo, Na) is relatively insensitive to experimental conditions."

"While the extent of glass reaction appears to be unaffected by the exposure rate, the amount of U, Pu, and Np released from the glass and the behavior of these elements in solution is a strong function of exposure rate. The present results indicate that as the exposure rate increases and the effective solution pH decreases, the normalized release of each actinide element increases, the fraction of Np and Pu that passes through a 50 Angstrom filter increases, the fraction of Np and Pu sorbed to metal, tuff, or glass decreases, and the depletion of U in the reacted glass layer increases."

COMMENTS OF REVIEWER

The decrease in the pH of leachates in the presence of radiation as well as the behavior of Np, Pu, and U is a cause for concern. It is questionable whether the ratio of the amount of air to water in the present experiments is a reasonable representation for the expected conditions in the repository. Since the amount of nitrogenous species formed is dependent on the amount of air, ground water in the repository might become more acidic than indicated by these experiments in which the ratio of air to leachate is 4/16. Scoping experiments with air to water ratios of 8/8 and 16/4 should be carried out.

RELATED HLW REPORTS

Bates, John K., Fisher, Donald F., and Gerding, Thomas J., " The Reaction of Glass During Gamma Radiation in a Saturated Tuff Environment, Part 1: SRL 165 Glass, ANL 85-62, February (1986).
Abrajano, T., Bates, J., Ebert, W., and Gerding, T., "The Effect of Gamma Radiation on Groundwater Chemistry and Glass Leaching as Related to the NWWSI Repository Site". UCRL-15825, Preprint SANL-510-001.

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

(a) Relationship to Waste Package Performance Issues Already Identified

2.3.2.1 What are the possible dissolution mechanisms of the waste form under the range of potential repository conditions?

(b) New Licensing Issues

(c) General Comments

AUTHOR'S ABSTRACT

The effect of gamma irradiation on groundwater and the reaction between groundwater and glass have been investigated at radiation exposure rates of 2 x 10^5, 1 x 10^3, and 0 R/h. These experiments, which bound the conditions may occur in a high-level nuclear waste repository located in tuff, have been performed using the actinide-containing glasses ATM-1c and ATM-8, and have been performed for time periods up to 278 days. The experimental results indicate that when only the repository groundwater is present, the pH of the system remains near-neutral, regardless of the radiation field, due to the buffering capacity of the solution. When glass is added to the system, the subsequent reaction is governed by the solution chemistry, which results from a complex interaction between radiolysis products, glass reaction products, and groundwater components. While no long-term reaction trends have been extracted from the current data, it is noted that there are no outstanding differences in the reaction of the glasses as measured by the release of the soluble components B, Mo, Na, as a function of radiation exposure rate. However, there is a marked difference in the amount of U, Np, and Pu released from the glasses as a function of radiation exposure rate. This difference can be correlated with the pH values of the leachate, with more basic solutions resulting in lower actinide release.