

UNITED STATES DÉPARTMENT OF COMMERCE National Institute of Standards and Technology (formerly National Bureau of Standards) Gaithersburg, Maryland 20899

July 6, 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 April and May 1990 (FIN-A-4171-0)

Dear Dr. Interrante:

Enclosed is the April and May 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. Iraker

Anna C. Fraker Metallurgist Corrosion Group Metallurgy Division

Enclosures

Distribution: WM Docket Control Center (1-original)

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A4171 / WM-11 NH14 /

### Progress Report for April - May 1990

Published June 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

	<u>Current Report</u>	<u>Previous Report</u>
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. WHC-EP-0065 (formerly HEDL-7637, "Electrochemical Corrosion - Scoping Experiments -- An Evaluation of Results", September 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 month. Two of these reports are on the subject of spent fuel, one on glass dissolution, one on container performance, and one on the performance of Zircaloy.

Spent Fuel

Pescatore, C., "14C Release From Failed Spent Fuel Containers" BNL-52231, UC-810, May-September 1989.

The release of  ${}^{14}C$ , in the form of  ${}^{14}CO_2$ , is a serious concern, since this radionuclide can access the environment rapidly. This report describes a model that assumes limited perforation of the container and occlusion of the perforations by corrosion products which leads to a significant delay to the gaseous release of  ${}^{14}C$  [Pescatore]. Other effects such as the oxidation rate of  ${}^{14}C$ , gaseous diffusion, and radioactive decay also are considered.

Stout, R.B., Shaw, H.F., Einziger, R.E. "Statistical Model For Grain Boundary and Grain Volume Oxidation Kinetics in  $UO_2$  Spent Fuel" UCRL-100859, September 1989.

A statistical model is used to describe the oxidation of  $UO_2$  spent fuel pellets [Stout]. Using thermogravimetric analysis data obtained from the oxidation of small fragments of  $UO_2$ , grain boundary and grain volume oxidation are modeled, and their effect on overall oxidation of spent fuel pellets is evaluated. The report describes the approach used in the development of this model.

### Glass

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.

The rate of dissolution of nuclear waste glass in groundwater has been measured as a function of gamma radiation for a period of up to 278 days [Bates]. The results indicate that in the presence of glass, the pH of the solution increases, leading to an increase in the dissolution of U, Np, and Pu. In the absence of glass, the pH of ground water is unaffected when exposed to gamma radiation.

**Container Performance** 

Frost, R.H., Muth, T.R., and Liby, A.L. "Effects of Manufacturing Variables on Performance of High-Level Waste Low Carbon Steel Containers, Final Report" NUREG/CR-5001, April 1990.

The use of cast steel overpacks is evaluated in terms of casting and welding processing variables [Frost]. The report concludes that centrifugal casting is the most economical and technically favorable approach to manufacture the steel overpacks. The technique requires welding the bottom end during manufacture and welding the top end at the repository. Though the authors believe that corrosion will be the most likely form of failure of the cast steel overpacks, this form of degradation is not evaluated.

### Zircaloy

Santanam, L., Shaw, H., and Chin, B.A. "Modeling of Zircaloy Cladding Degradation Under Repository Conditions" UCRL-100211, July 1989.

The effect of creep and stress corrosion of Zircaloy cladding during repository storage has been studied by stress analysis modeling

[Santanam]. Using this approach, deformation and fracture mapping of the Zircaloy surface is used to predict maximum storage temperatures in the repository. The results indicate that that the maximum allowable temperatures are similar to those expected in the waste containers, and the oxide film on Zircaloy will be under a compressive hoop stress.

Category 1 -- Reports currently being reviewed;

- 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.
- UCRL-21076, "Electrochemical Corrosion Studies on Copper-Based Waste Package Container Materials in Unirradiated 0.1 N NaNO<sub>3</sub> at 95°C", May 1988.
- 4. UCRL-101615, "Prototype Engineered Barrier System Field Tests Progress Report", July 1989.
- 5. BNL-52231, UC-810, "<sup>14</sup>C Release From Failed Spent Fuel Containers", May-September 1989.
- 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.
- 7. NUREG/CR-5001, "Effect of Manufacturing Variables on Performance of High-Level Waste Low Carbon Steel Containers," April 1990.
- 8. UCRL-100211, "Modeling of Zircaloy Cladding Degradation Under Repository Conditions," July 1989.

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			
UCRL-101615	2/12/90			
BNL-52231, UC-810	2/12/90	5/31/90		
Bates, 1988	2/12/90			
NUREG/CR-5001	5/31/90			
UCRL-100211	5/31/90			

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

 Stout, R.B., Shaw, H.F., Einziger, R.E., "Statistical Model For Grain Boundary and Grain Volume Oxidation Kinetics in UO<sub>2</sub> Spent Fuel", UCRL-100859, September 1989.

Category 3 -- File and cross reference

None this quarter.

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

Date: April through May 1990

An abstract, entitled, "The Effect of Oxygen Transport and Resistivity of the Environment on the Rate and Form of Corrosion of Steel in Simulated Soils", is being prepared for submission to the Symposium on the Scientific Basis for Nuclear Waste Management sponsored by the Materials Research Society. The fall meeting will meet in Boston, MA in November of this year.

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

Because of the interest in retrieval of the 20 year stainless steel specimens now located at six underground test sites, information on accessibility of the sites and costs of retrieval are being obtained. Contact with personnel at these sites, all on government owned land, reveals that five of the six sites are intact, and the sixth site, in the Baltimore area, is still being investigated. Records on the history of the stainless steel underground program have been examined, showing that at least one set of specimens is still at each of the six sites. A set of specimens consists of 200, 300, and 400 series stainless steels, several special combinations of high-chromium, high-nickel, or high-molybdenum alloys, and three composite alloys. These materials were exposed in several geometries (sheet, strip, Ubend) and heat treatments, (sensitized, unsensitized, half hard, full hard, welded).

The results of the first eight years indicated that, in the two most corrosive sites, practically all materials suffered some degree of deterioration, but in the site most similar to the tuff environment, Site A, several materials survived the first eight years unaffected by corrosion attack. Site A is of particular interest, because its soil is of volcanic origin, and is made up of an uncompacted volcanic ash in an arid section of Washington. The location of the site is approximately 110 miles Southeast of Seattle with an average precipitation of less than 10 inches per year. The remaining sites are in the Eastern portion of the U.S., and are exposed to higher levels of moisture. Preliminary retrieval plans are being developed.

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

April through May 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 longterm 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.

Zircaloy-2, Zircaloy-4 cladding specimens (outside wall of the tubing exposed) for extended time tests were immersed in 0.1 M NaF solution at a temperature of 23°C and a pH of 8 on April 4, 12 and 20, 1990. Cyclic

polarization measurements from the Zircaloy-4 bulk wrought material, Zircaloy-4 tubing and Zircaloy-2 tubing all show a hysteresis loop in the cyclic polarization curve. Also, the protection potential is at a value negative to the corrosion potential indicating that localized corrosion will occur. The general corrosion rate is negligible.

The data for the Zircaloy materials exposed in the 0.1M NaF solution are different from data obtained on these materials exposed in J-13 water. Two striking differences are the increase in the amount of hysteresis in the cyclic polarization curves and the value of the protection potential being negative to the original corrosion potential. The general corrosion rate still appears to be negligible, but this may not prove to be the case after additional testing and data analysis. Specimans exposed in the NaF solution will remain there for an extended time and additional testing. Future work involves setting up and testing the materials in 0.1M NaCl and 0.1M NaI solutions.

### TASK 4 - General Technical Assistance

The request of May 10, 1990 for general technical assistance was on the report entitled, "Report to Congress on the Potential Use of Lead in the Waste Packages for a Geological Repository at Yucca Mountain, Nevada". This was reviewed by Mr. E. Escalante and sent to the NRC on May 18, 1990. This review also was prepared for inclusion in the data base.

Another request was to review the report on "Techniques for Determining Probabilities of Events and Processes Affecting the Performance of Geological Repositories: Volume 2". This was reviewed by Dr. K. Coakley and sent to the NRC on May 14, 1990.

# SDI006, UD 9009, SER. DD016

File(s) searched:

File 6:NTIS - 64-90/ISSUE09 (COPR. 1990 NTIS)

Sets selected:

Set	Items	Description
1	2	WASTE(W)PACKAGE?
2	3	CANISTER?
3	32	CORROSION
4	11	LEACHING
5	72	GLASS
6	6	VITRIFICATION
7	113	S3-S6/OR
8	1	HIGH(W)LEVEL(W)WASTE?
9	69	RADIOACTIVE(W)WASTE?
10	5	NUCLEAR(W)WASTE?
11	0	(S1 OR S2) AND S7 AND (S8 OR S9 OR S10)
12	0	ANNA FRAKER RM. 8-106 BLDG. 223 X6009
13	0	JILL RUSPI

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

Date Time Description OGapr OG:28EST PR 11/5/1-25 (no items to PRINT)

Total items to be printed: 0



# SDI032, UD 9005, SER. DD022

File(s) searched:

File 32:METADEX 66-90/MAY (COPR. 1990 ASM INTERNATIONAL)

Sets selected:

Set	Items	Description
1	9	HIGH()LEVEL()WASTE? ? OR RADIOACTIVE()WASTE?
		OR NUCLEAR()WASTE?
2	1709	STEEL? ? OR ZIRCALOY? ? OR TITANIUM? ? OR Copper
3	5	1*2
4.	0	ANNA FRAKER, 223, B-254, X6009

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

Date Time Description 07apr 04:04EST PR 3/5/1-25 (items 1-5)

Total items to be printed: 5

# SD1006, UD 9010, SER. DD016

File(s) searched:

File 6:NTIS - 64-90/ISSUE10 (COPR. 1990 NTIS)

Sets selected:

Set	Items	Description
1	8	WASTE(W)PACKAGE?
2	6	CANISTER?
3	65	CORROSION
4	27	LEACHING
5	134	GLASS
6	7	VITRIFICATION
7	210	53-56/OR
8	4	HIGH(W)LEVEL(W)WASTE?
9	179	RADIOACTIVE(W)WASTE?
10	22	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 19apr 15:18EST PR 11/5/1-25 (1tems 1-1)

Total items to be printed: 1

PAGE: 10

# SDI103, UD 9007, SER. DD017

File(s) searched:

File 103:DOE ENERGY - 83-90/MAY(ISSO7)

#### Sets selected:

Set	Items	Description
1	6	WASTE(W)PACKAGE?
2	3	CANISTER?
3	180	CORROSION (1974 DEC)
4	33	LEACHING (1974 DEC)
5	94	GLASS (1974 DEC)
6	6	VITRIFICATION (1974 DEC)
7	307	\$3-\$6/0R
8	6	HIGH(W)LEVEL(W)WASTE?
9	199	RADIOACTIVE(W)WASTE?
10	27	NUCLEAR (W)WASTE?
11	4	(S1 OR S2) AND S7 AND (S8 OR S9 OR S10)
12	O	ANNA FRAKER RM. B-106 BLDG. 223 X6009
13	0	JILL RUSPI

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

Date Time Description 21apr 06:39EST PR 11/5/1-25 (1tems 1-4)

Total items to be printed: 4



INFORMATION SERVICES, INC.

# SDI103, UD 9008, SER. DD017

File(s) searched:

File 103:DOE ENERGY - 83-90/MAY(ISSO8)

#### Sets selected:

Set	Items	Description
1	8	WASTE(W)PACKAGE?
2	3	CANISTER?
3	68	CORROSION (1974 DEC)
4	46	LEACHING (1974 DEC)
5	61	GLASS (1974 DEC)
6	6	VITRIFICATION (1974 DEC)
7	165	S3-S6/OR
8	13	HIGH(W)LEVEL(W)WASTE?
9	131	RADIDACTIVE(W)WASTE?
10	25	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

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

Date TimeDescription04may02:45ESTPR11/5/1-25(items 1-3)

Total items to be printed: 3



INFORMATION SERVICES, INC.

# SDI006, UD 9011, SER. DD016

File(s) searched:

File 6:NTIS - 64-90/ISSUE11 (COPR. 1990 NTIS)

### Sets selected:

Set	Items	Description
1	0	WASTE(W)PACKAGE?
2	9	CANISTER?
3	44	CORROSION
4	15	LEACHING
5	105	GLASS
6	8	VITRIFICATION
7	156	\$3-\$6/OR
8	5	HIGH(W)LEVEL(W)WASTE?
9	143	RADIOACTIVE(W)WASTE?
10	28	NUCLEAR(W)WASTE?
11	4	(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

13

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

Date Time Description O5may 07:16EST PR 11/5/1-25 (items 1-4)

Total items to be printed: 4

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# SDI008, UD 9005, SER. DA016

File(s) searched:

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

### Sets selected:

Set	Items	Description
1	0	WASTE(W)PACKAGE?
2	7	CANISTER?
3	280	CORROSION
4	17	LEACHING
5	370	GLASS
6	7	VITRIFICATION
7	657	S3-S6/OR
8	3	HIGH(W)LEVEL(W)WASTE?
9	41	RADIOACTIVE(W)WASTE?
10	2	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

14

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

Date TimeDescription04may08:01ESTPR11/5/1-20(1tems 1-1)

Total items to be printed: 1



# SDI103, UD 9009, SER. DD017

File(s) searched:

File 103:DDE ENERGY - 83-90/MAY(ISSO9)

#### Sets selected:

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15

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Set	Items	Description
1	4	WASTE(W)PACKAGE?
2	10	CANISTER?
3	129	CORROSION (1974 DEC)
4	46	LEACHING (1974 DEC)
5	75	GLASS (1974 DEC)
6	7	VITRIFICATION (1974 DEC)
7	232	\$3-\$6/OR
8	17	HIGH(W)LEVEL(W)WASTE?
9	172	RADIOACTIVE(W)WASTE?
10	40	NUCLEAR (W)WASTE?
11	3	(S1 OR \$2) AND \$7 AND (S8 OR \$9 OR \$10)
12	0	ANNA FRAKER RM. B-106 BLDG. 223 X6009
13	Ō	JILL RUSPI

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

Date Time Description 11may 04:22EST PR 11/5/1-25 (items 1-3)

Total items to be printed: 3



# SD1006, UD 9012, SER. DD016

File(s) searched:

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File 6:NTIS - 64-90/ISSUE12 (COPR. 1990 NTIS)

Sets selected:

Set	Items	Description
1	4	WASTE(W)PACKAGE?
2	2	CANISTER?
3	54	CORROSION
4	17	LEACHING
5	101	GLASS
6	3	VITRIFICATION
7	165	53-56/0R c
8	10	HIGH(W)LEVEL(W)WASTE?
9	193	RADIOACTIVE(W)WASTE?
10	25	NUCLEAR(W)WASTE?
11	3	(S1 OR \$2) AND \$7 AND (S8 OR \$9 OR \$10)
12	.0	ANNA FRAKER RM. B-106 BLDG. 223 X6009
13	0	JILL RUSPI

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

DateTimeDescription17may12:26ESTPR11/5/1-25(items 1-3)

Total items to be printed: 3



# SDI103, UD 9008, SER. DD017

File(s) searched:

File 103:DOE ENERGY - 83-90/MAY(ISSO9)

### Sets selected:

Set	Items	Description
1	8	WASTE(W)PACKAGE?
2	3	CANISTER?
3	68	CORROSION (1974 DEC)
4	46	LEACHING (1974 DEC)
5	61	GLASS (1974 DEC)
6	6	VITRIFICATION (1974 DEC)
7	165	\$3-\$6/0R
8	13	HIGH(W)LEVEL(W)WASTE?
9	131	RADIOACTIVE(W)WASTE?
10	25	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

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

Date TimeDescription22may16:18ESTPR11/5/1-25(items 1-3)

Total items to be printed: 3



# SDI293, UD 9006, SER. DD023

File(s) searched:

File 293:ENGINEERED MATERIALS ABS 86-90/JUN (COPR. 1990 ASM INTERNATIONAL)

### Sets selected:

Set	Items	Description
1	3	HIGH()LEVEL()WASTE? ? OR RADIDACTIVE()WASTE? OR NUCLEAR()WASTE?
2	326	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 22may OO:22EST PR 3/5/1-25 (no items to PRINT)

Total items to be printed: 0

18



# SDI103, UD 9009, SER. DD017

File(s) searched:

File 103:DOE ENERGY - 83-90/MAY(ISSO9)

Sets selected:

Set	Items	Description
1	4	WASTE(W)PACKAGE?
2	10	CANISTER?
3	129	CORROSION (1974 DEC)
4	46	LEACHING (1974 DEC)
5	75	GLASS (1974 DEC)
6	7	VITRIFICATION (1974 DEC)
7	232	53-56/0R
8	16	HIGH(W)LEVEL(W)WASTE?
9	172	RADIOACTIVE(W)WASTE?
10	40	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

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

Date Time Description 22may 16:45EST PR 11/5/1-25 (items 1-3)

Total items to be printed: 3



## SDI032, UD 9006, SER. DD022

### File(s) searched:

F11e 32:METADEX 66-90/JUN (COPR. 1990 ASM INTERNATIONAL)

### Sets selected:

Set	Items	Description
1	4	HIGH()LEVEL()WASTE? ? OR RADIOACTIVE()WASTE? OR NUCLEAR()WASTE?
2	2010	STEEL? ? OR ZIRCALOY? ? OR TITANIUM? ? OR Copper
3	3	1*2
4	0	ANNA FRAKER, 223, B-254, X6009

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

Date Time Description 24may 09:06EST PR 3/5/1-25 (items 1-3)

Total items to be printed: 3



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

### DATA\_SOURCE

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

H. D. Smith "Electrochemical Corrosion Scoping Experiments -- An Evaluation of the Results" WC-EP-0065, September, 1988.

(b) Organization Producing Data

Westinghouse Hanford Laboratory, P. O. Box 1970, Richland, Washington 99352

<u>DATE REVIEWED</u>: 4/30/90

### PURPOSE

"The purpose of this report is to present the results of the electrochemical corrosion scoping experiment, which investigated the corrosion behavior of Zircaloy spent fuel cladding in hot (90°C), tuff-equilibrated Well J-13 water."

### **KEYWORDS**

Experimental data, supporting data, general corrosion, scoping test, corrosion, microscopy, surface film, visual examination, electron diffraction, laboratory, simulated field, Yucca Mountain, J-13 water, tuff composition, tuff, basic (alkaline) solution (pH>7), high temperature, cladding stainless steel, Zircaloy, zirconium base, spent fuel, <sup>14</sup>C, corrosion (galvanic), corrosion (local).

### **CONTENTS**

This 45 page report contains 16 figures, 5 tables and cites 12 references. Some information an measured data on  $^{14}$ C release also are included. The content is categorized as follows:

Topic	No. of Pages
Summary	2
Introduction	2
Experimental Design and Procedure	11
Post Experiment Evaluation of the Spent Fuel Cladding	21
Discussion and Conclusions	8
References	1

### TEST CONDITIONS

The test cladding sections were 4.5 to 4.75 in. long and were plugged with unirradiated Zircaloy-4 plugs held in place with EPON (Trademark of the Shell Oil Co.) Resin 828 that was cured at 150°C for 2 hours. Cladding was gathered into bundles of seven (six around one) test pieces and held in place in jars with a 2 in. by 6 in. 304 stainless steel strip with a 301 stainless steel hose clamp and a 410 stainless steel screw. Prewashed crushed tuff was placed around the bundle and J-13 well water was added to a level about half way up the side of the bundle. The jars were placed in a heated (100 to 101°C) oil bath to maintain a specimen temperature of 90°C, and the test duration was from 2 to 12 months. The value of the pH ranged row 8.59 to 8.76, and the solution conductivity ranged from 724 to 754 uS. Any lost water was replaced with deionized water.

### UNCERTAINTIES IN DATA

Organic anions interfere with the fluoride analyses of the J-13 water. Difficulties associated with the limited room to insert the conductivity probe are believed to account for the amount of scatter in the data and the obscureness of small trends that might have been present. Too small an amount of liquid may have caused one inaccurate measurement of  $1^{4}C$ .

#### DEFICIENCIES LIMITATIONS IN DATABASE

None were given.

### CONCLUSIONS OF AUTHOR

- "Metallography and standard SEM techniques did not identify any kind of corrosion (homogeneous) or localized) on Zircaloy-4 spent fuel cladding in contact with tuft equilibrated Well J-13 water at 90°C for up to 1 yr. Prepolishing part of the cladding surface would greatly enhance the probability of identifying localized corrosion by microscopic methods after a test."
- 2. "If the model for aqueous oxidative corrosion of zirconium and its alloys summarized by Rothman1 can be extrapolated to 90°C and 170°C, then metallographic techniques and SEM are not sensitive enough to observe homogeneous corrosion on spent fuel cladding. However, Auger/ion milling techniques should be able to measure oxidative corrosion films that would develop on polished surfaces."
- 3. "The TEM, SEM and electron diffraction were used to characterize ultrathin sections of cladding from the 12-mo experiment and archived cladding material from the same fuel rod, but were unable to detect any differences between them believed to be due to corrosion. It is recommended, however, that these techniques be applied to material from the 170°C autoclave tests when they become available."
- 4. "Carbon-14 is expected to be a valuable indicator of cladding interaction with the environment once its distribution and state in the cladding is well understood and described."

### COMMENTS OF REVIEWER

Zircaloy spent fuel cladding from the Turkey Point spent fuel assembly B-17 (removed from service in 1975) was used. Portions of the cladding from various levels in the core were used to include material with thick and thin oxide films and a corresponding range of hydrides, "crud" scale of different kinds.

After the completion of each test, the bundles of spent fuel cladding were inspected visually, allowed to dry, and documented from several angles photographically before disassembling. After disassembly, additional visual examination and photographic documentation were made. Two cladding sections from each experiment, one with a thin oxide and one with a thick oxide, were selected for analysis using metallography, scanning electron microscopy (SEM), transmission electron microscopy (TEM) and electron diffraction. Chemical analyses were conducted to monitor water at different times during the test, and also to determine organic and inorganic carbon as well as  $1^4$ C.

The tests reported in this paper were carefully planned and conducted, and the data obtained are useful. These data are on actual fuel cladding sections that already had oxide films and crud layers present, and the tests conducted from 2 to 12 months at 90°C involved configurations with a water/air line, creviced areas and contact with the steel. The data were obtained by visual observation, light and electron microscopy, and results indicate no localized corrosion and less than 0.1 um of general corrosion on either the Zircaloy cladding or the stainless steel. Metallographic studies were conducted to compare cladding specimens exposed in the tests with cladding specimens not exposed, and no change in the oxide film or surface was observed. This study referenced the report of Rothman<sup>1</sup>, and based on extrapolation of the corrosion is negligible even for long time periods such as 10,000 years. There were assumptions made in the review by Rothman, and these need to be checked.

The <sup>14</sup>C measurements indicated a release of 180/mL (disintegrations per minute per milliliter of solution), and this appeared to be close to a steady state concentration for J-13 water. The solutions lost <sup>14</sup>C when out of contact with the cladding bundles, and this loss may have been to the atmosphere. The <sup>14</sup>C measurement was the only observable reaction of the cladding with the tuff-equilibrated J-13 water. It was stated that it is important to determine the distribution and chemical state of <sup>14</sup>C in the cladding.

The work reported here can be complemented with some measurements of electrode potentials, corrosion rates and associated environmental effects. The conclusions presented in this report contain good suggestions for further work.

## RELATED HLW REPORTS

Reports are cited in the references; UCID-20172 (<sup>1</sup>Rothman, reviewed at NIST), HEDL-7455 Rev.1, HEDL-TC-2562, UCRL-53629, HEDL-TME 80-85, HEDL-TME 83-9 and HEDL-7455 (reviewed at NIST).

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

- (a) Relationship to Waste Package Performance Issues Already Identified
  - 2.3.6 What are the potential failure mechanisms for spent fuel cladding?
- (b) New Licensing Issues