

Low-Temperature Oven Method for Spent Fuel Oxidation Testing

- Activity Plan (D-20-45) -

for

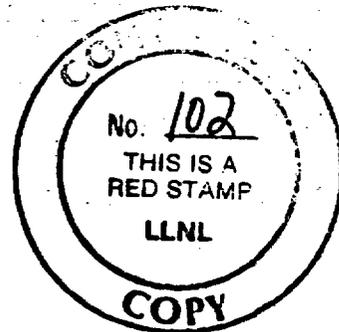
YMP WBS Element 1.2.2.3.1.1

LLNL-YMP Spent Fuel Waste Form Testing

Lawrence Livermore National Laboratory

Revision 0

August 16, 1989



APPROVALS:

*L. J. Jardine* 8/28/89  
L. J. Jardine, Project Leader Date

*David W. Short* 8/23/89  
D. W. Short, Quality Assurance Manager Date

*H. F. Shaw* 8/23/89  
H. F. Shaw, Technical Area Leader Date

9005100166 900528  
PDR WASTE  
WM-11 PDC



**CHANGE NOTICE**

CN No.: CN:1 D-20-45

Affected Document: **Low-Temperature Oven Method for Spent Fuel Oxidation Testing  
Activity Plan D-20-45, Rev. 0**

Prepared by: **E. C. DeLeon**

Approved by: [Signature] 9/28/89  
(Technical Area Leader) (Date)

Approved by: [Signature] 9/28/89  
(YMP QA Manager) (Date)

Training Required:  
Yes  No

Approved by: [Signature] 9/28/89  
(YMP Project Leader) (Date)

Currently Read as Follows:

Paragraph 3.3, 1st sentence, "... Section 2.0 of the test plan ..."

Changed to Read:

Paragraph 3.3, 1st sentence, "... Section 2 of the test plan ..."

**NOTE: THIS CHANGE NOTICE IS TO BE FILED AT THE FRONT OF THE AFFECTED DOCUMENT**

**Appendix B - Test Plan for Long-Term, Low-Temperature Oxidation of BWR  
Spent Fuel**

TEST PLAN FOR LONG-TERM,  
LOW-TEMPERATURE OXIDATION  
OF BWR SPENT FUEL

R. E. Einziger

December 1988

Prepared for  
Lawrence Livermore National Laboratory  
under a Related Services Agreement  
with the U.S. Department of Energy  
Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory  
Richland, Washington 99352

## CONTENTS

SUMMARY .....	iii
1.0 INTRODUCTION .....	1.1
1.1 OBJECTIVE OF DRY BATH TESTING .....	1.1
1.2 BACKGROUND .....	1.2
2.0 TEST DESCRIPTION .....	2.1
2.1 TEST MATRIX .....	2.1
2.1.1 Test Temperature .....	2.1
2.1.2 Fuel Particle Size .....	2.2
2.1.3 Moisture in the Atmosphere .....	2.4
2.1.4 Number of Samples and Test Duration .....	2.4
2.1.5 Radiation Fields .....	2.5
2.2 SAMPLE SELECTION, PREPARATION, AND IDENTIFICATION .....	2.6
2.3 EQUIPMENT DESCRIPTION .....	2.9
2.4 TEST OPERATION .....	2.10
2.4.1 Initial Startup .....	2.10
2.4.2 Interim Examination .....	2.10
2.4.3 Final Examination .....	2.11
2.5 POST-TEST SAMPLE EVALUATION .....	2.11
2.5.1 Ceramography .....	2.11
2.5.2 X-Ray Diffraction .....	2.12
2.5.3 Scanning Electron Microscopy .....	2.12
2.5.4 Transmission Electron Microscopy and Electron Diffraction .....	2.12
2.6 ANALYSIS OF DATA .....	2.14
2.7 PROCEDURES .....	2.14

fuel in the repository. Earlier testing<sup>(11)</sup> indicated that oxidation may be a two-step process consisting of oxygen diffusion down the grain boundary, followed by diffusion of oxygen into the grains; the second step is the rate-controlling mechanism. If the grain boundary diffusion is rapid with respect to the test duration, such as in the tests at 200°C and 225°C, then samples consisting of fragments are suitable as test specimens.

If diffusion down the grain boundaries takes longer than the test duration, then measuring weight gain of fuel fragments will not reflect the rate-limiting diffusion into the grains.<sup>(12)</sup> The grain boundary diffusion step can be significantly enhanced if the fragments are pulverized into smaller particles.

Thermogravimetric analysis testing of PWR fuel<sup>(13)</sup> indicated that pulverization does little to enhance oxidation above 175°C. At 140°C there appeared to be some enhancement of the oxidation. This observation was supported by the results of the dry bath oxidation tests on PWR fuel,<sup>(8)</sup> which showed no enhancement at 175°C but a distinct spread of over a factor of 2 in the rate of weight gain due to oxidation for the different particle sizes at 130°C and 110°C. A lengthy argument based on the onset of bulk diffusion was made in the Series 1 test plan<sup>(10)</sup> to justify the selection of particle size. The reasoning presented there still holds. The particle sizing for the pulverized fuel used in the Series 1 PWR fuel oxidation tests will again be used in the BWR fuel oxidation tests.

Fuel can easily be sieved or pulverized so that each size fraction has the same chemical composition and radiation spectrum. In the 175°C experiment, as-irradiated fragments will be used primarily. Pulverized samples, primarily in the -10/+24 Tyler mesh range, will be used in the 130°C test, and pulverized samples, primarily in the -24/+60 Tyler mesh range, will be used in the 110°C test. The maximum diameter of particles passing through the number 10, 24, and 60 Tyler meshes are 1.7 mm, 0.71 mm, and 0.25 mm, respectively. The complete matrix is in Table 2.1.

**TABLE 2.2. Characteristics of ATM-105 Cooper BWR  
Fuel from General Electric**

<u>Fuel Type</u>	<u>BWR 7 x 7</u>
Assembly Identification	CZ-346
Discharge Date	(a)
Nominal Burnup	~26 Mwd/kgHM
Fission Gas Release	(b)
Initial Enrichment	2.93%
Initial Pellet Density	(a)
Initial Rod Diameter	(a)
Cladding Material	Zircaloy-2
Cladding Thickness	(a)
Rod Identification	ADD-2974

- (a) Information to be provided in a characterization report to be released at a later date by MCC.  
(b) To be measured by MCC.

The MCC will characterize the rods. Prior to cutting, the rods will be punctured for both chemical and isotopic fission gas sampling. Gross and spectral gamma scanning will be used to determine the burnup profile. Burnup analyses will be conducted at three locations in the bottom half of the rod. Transverse and longitudinal ceramography examinations will be performed adjacent to the burnup samples to determine grain size and to look for unusual features.

These tests are not concerned with burnup effects, so 18 in. of fuel with nearly equal burnup as indicated by the gamma scans will be used for testing. At ~10 g per sample and 31 g of fuel per in., there is sufficient fuel for ~55 samples. The fuel segments will be cut dry into 4- to 6-in.-long pieces for handling purposes. The cladding will be split with a carbide end mill and then pried open for removal of the fuel fragments. Six fragments will be set aside for possible characterization, ~25 fragments will be used in TGA testing, and the remainder of fuel will be used for dry bath test samples.

### 2.4.3 Final Examination

At designated intervals, selected samples will be removed from the tests for interim examinations, for further examination, and for use as leaching samples. After weighing, the fuel will be poured into a labeled metal can for transfer out of the cell. The empty crucible will be ultrasonically rinsed with ethyl alcohol and reweighed.

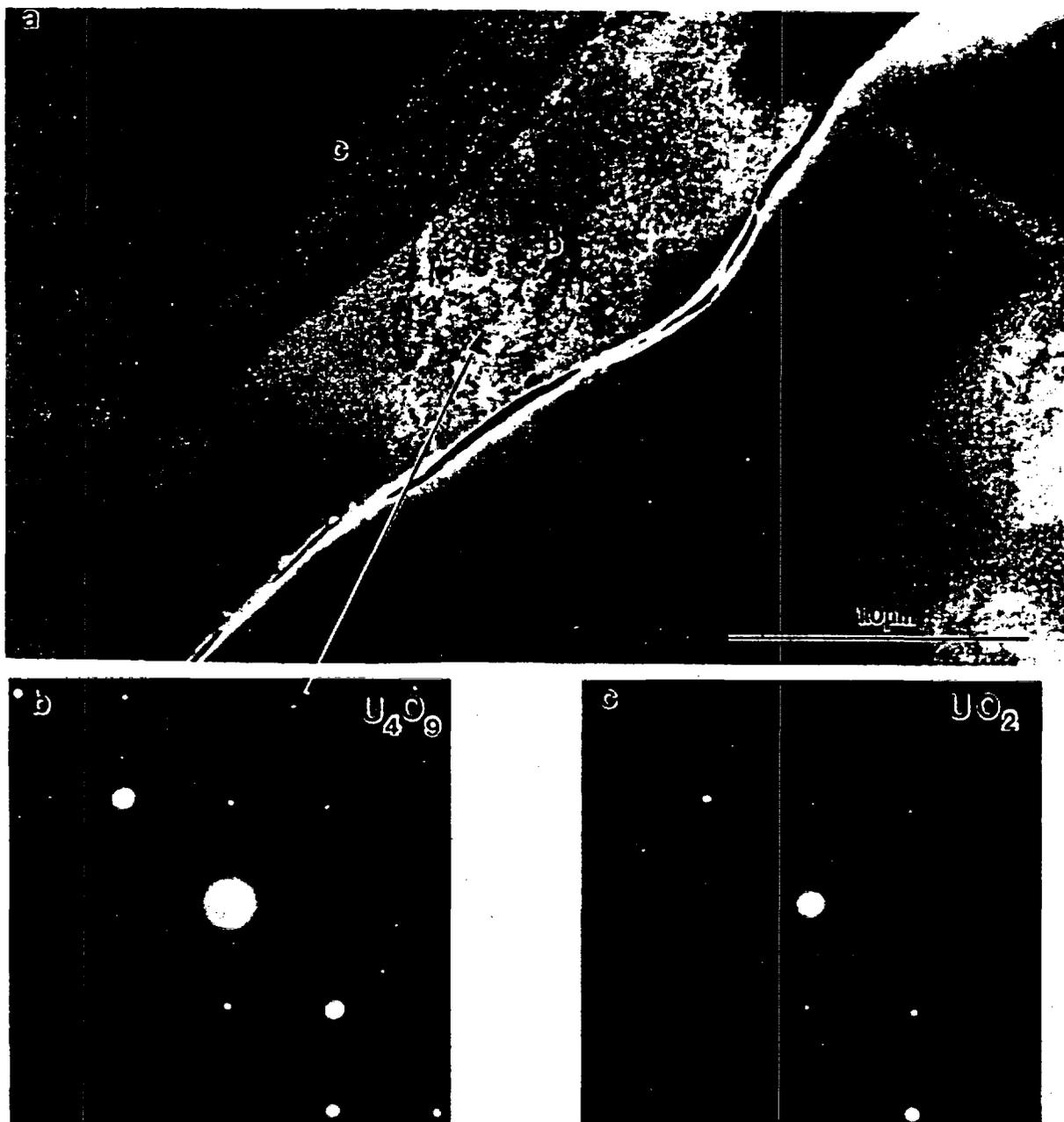
## 2.5 POST-TEST SAMPLE EVALUATION

The weight change of the samples will be checked at intervals during the test, but weight measurements cannot determine the uniformity of oxidation or the location of the oxidation process in the sample. Only limited information can be determined about the oxide phases. For instance, complete conversion of  $UO_2$  to  $U_3O_7$  yields a weight gain of 1.98%, and complete conversion to  $U_3O_8$  yields a weight gain of 3.96%. If the sample weight gain is found to be greater than 1.98%, it might be expected that at least some  $U_3O_8$  has formed. However, the sample could be almost uniformly  $U_3O_7$  with a minor amount of  $U_3O_8$ , or it could be partly  $UO_2$  and partly  $U_3O_8$ . In all likelihood, a combination of the phases will occur. Following the test, selected samples will be examined ceramographically by SEM, XRD and TEM/electron diffraction to obtain additional phase information. All samples consisting of fragments will be visually examined to see if there is any spallation.

The goal of the waste form testing program is to determine the radionuclide release characteristics of the spent fuel waste form. Part of this task is to determine the change in leaching characteristics with oxidation state. After post-test evaluations, the test material will be stored in sealed containers and labeled with the sample identification. These samples will be available for leach testing.

### 2.5.1. Ceramography

Ceramography will be used to visually determine the gross grain boundary degradation and to evaluate the extent of the oxidation into the fragment from the external surfaces. In addition, it will serve as a sampling guide for taking TEM specimens.



**FIGURE 2.2.** Grain Boundary Region in Oxidized Fuel. Sample G7-14-3-#8 (155°C test, O/M = 2.05). (a) Darkfield TEM taken with diffuse-scattered intensity from U<sub>4</sub>O<sub>9</sub>, showing enhanced intensity from U<sub>4</sub>O<sub>9</sub> on one side of grain boundary. (b) Selected-area diffraction pattern from U<sub>4</sub>O<sub>9</sub> region in (a), showing diffuse scattering from short-range ordered oxygen interstitials. Near (001) orientation. Arrow indicates aperture position used to form darkfield image (a). (c) SAD pattern for UO<sub>2</sub> region in (a). Same crystal orientation as (b).

DISTRIBUTION

No. of  
Copies

No. of  
Copies

OFFSITE

10 DOE/Office of Scientific and  
Technical Information

13 DOE-HQ/Office of Civilian  
Radioactive Waste Management  
Forrestal Building  
Washington, DC 20585  
ATTN: C. E. Kay,  
Acting Director  
D. H. Alexander  
J. C. Bresee  
S. J. Brocoum  
V. J. Cassella  
M. W. Frei  
B. G. Gale  
T. H. Isaacs  
S. H. Kale  
G. Parker  
S. Rousso  
J. D. Saltzman  
R. Stein

24 DOE/Nevada Operations Office  
P.O. Box 98518  
Las Vegas, NV 89193-8518  
ATTN: M. Blanchard, YMPO  
M. Cloninger  
P. K. Fitzsimmons  
J. L. Fogg (12)  
C. P. Gertz, YMPO  
MS-523 (5)  
L.P. Skousen, YMPO  
M. Valentine, YMPO  
C. L. West  
  
A. T. Tamura  
DOE/Office of Scientific  
and Technical Information  
Science and Technology Division  
P.O. Box 62  
Oak Ridge, TN 37831

RW-1  
RW-332  
RW-10  
RW-221  
RW-123  
RW-22  
RW-23  
RW-40  
RW-20  
RW-333  
RW-10  
RW-20  
RW-30

2 Argonne National Laboratory  
Chemical Technology Division  
9700 S. Cass Avenue, Bldg. 205  
Argonne, IL 60439  
ATTN: J. Bates  
E. Veleckis

Battelle Columbus Laboratory  
505 King Avenue  
Columbus, OH 43201  
ONWI Library

D. Schweitzer  
Brookhaven National Laboratory  
Associated Universities, Inc.  
Upton, NY 11973

Director  
Center for Nuclear Waste  
and Regulatory Analyses  
6220 Culebra Road  
Drawer 28510  
San Antonio, TX 78284

Director, Community Planning  
City of Boulder City  
P.O. Box 367  
Boulder City, NV 89005

5 J. Foremaster  
City of Caliente  
P.O. Box 158  
Caliente, NV 89008

City Manager  
City of Henderson  
Henderson, NV 89015

Economic Development Dept.  
City of Las Vegas  
400 E. Steward Avenue  
Las Vegas, NV 89101

No. of  
Copies

No. of  
Copies

ONSITE

DOE Richland Operations Office

D. C. Langstaff  
P. H. Turner  
J. J. Sutey

39 Pacific Northwest Laboratories

M. J. Apted  
W. W. Ballard  
H. C. Buchanan

T. T. Claudson  
R. E. Einziger (20)  
E. R. Gilbert  
W. J. Gray  
A. B. Johnson  
S. C. Marschman  
J. E. Mendel  
P. W. Reimus  
H. D. Smith  
L. E. Thomas  
C. N. Wilson  
Technical Report Files (5)  
Publishing Coordination

**Appendix C - Addendum to Test Plan for Long-Term, Low-Temperature  
Oxidation of High Burnup Spent Fuel**



**Battelle**

Pacific Northwest Laboratories  
P.O. Box 999  
Richland, Washington U.S.A. 99352  
Telephone (509)  
Telex 15-2874

August 4, 1989

Dr. Ray Stout, L-201  
Lawrence Livermore National Laboratory  
P.O. Box 808  
Livermore, CA 94550

Dear Ray:

Attached is a revised copy of the "Addendum to Test Plan for Long-Term, Low-Temperature Oxidation of High Burnup Spent Fuel."

If you have any questions, please contact me on FTS 444-3453.

Sincerely,

Dr. Robert E. Einziger, Manager  
Materials & Chemical Systems Performance Section

REE:neb  
Attachment

cc: SC Marschman - PNL

**Addendum to Test Plan for Long-Term,  
Low-Temperature Oxidation of High Burnup  
Spent Fuel**

**R. E. Einziger**

**January 1989**

## 1.0 INTRODUCTION

The Yucca Mountain Project Office (YMPO) is evaluating a site at Yucca Mountain, Nevada, to determine the suitability of this site for a high-level nuclear waste repository. The horizon that is under investigation for repository development is the Topopah Spring Member of the Paintbrush Tuff, a welded, devitrified ash flow tuff. At Yucca Mountain, this unit lies in the unsaturated zone; the water table is hundreds of meters below the reference repository horizon. Lawrence Livermore National Laboratory (LLNL) is developing designs for waste packages and testing the performance of waste forms and metal barriers under expected repository conditions for the YMPO Project.

The Environmental Protection Agency (EPA) (40 CFR 191 1982) and the Nuclear Regulatory Commission (NRC) (10 CFR 60 1983) have imposed requirements limiting potential radionuclide release from a high-level nuclear waste repository. The potential change in the oxidation state of spent fuel during its residence in a repository must be known to evaluate radionuclide retention capabilities of the repository. Analyses indicate that  $UO_2$  will oxidize to higher states under the temperature and atmospheric conditions expected in a tuff repository (Einziger and Woodley 1985). If oxidation progresses sufficiently, cladding that contained breaches might split open, or significant quantities of higher oxides with potentially higher leach rates might form.

An integrated technical approach (Einziger 1985) was developed to study spent fuel oxidation at low temperatures characteristic of the post-container breach period. The objective of the long-term oxidation testing is to verify at low temperatures the predictions based on the thermogravimetric analysis (TGA) results of the influence of important fuel characteristics (i.e., gas release, burnup, fuel type, etc.) and atmospheric variables (i.e., moisture content, radiation field) on oxidation rates and mechanisms. Series 1 tests were designed to verify the effects of atmospheric moisture, temperature, and particle size on the oxidation rate and phase formation in pressurized-water reactor (PWR) fuel. Series 2 tests had the same goals as Series 1 except

that boiling-water reactor (BWR) spent fuel was used. These tests will also provide fuel at various stages of oxidation for comparative dissolution tests with as-irradiated fuel. Both of these series had detailed test plans (Einziger 1986 and 1987). This test plan addendum to the Series 2 Test Plan is for Series 3, and will present the details for the long-term, low-temperature oxidation tests of high burnup PWR spent fuel that has high and low gas release.

## 2.0 ADDENDUM

Series 3 will use the same equipment, test temperatures, fuel particulate sizes, atmospheres, radiation fields, sample preparation, sample identification, pre-test characterization, test operation, post-test evaluation, analyses of data, procedures, reporting, data recording, and quality assurance as Series 2. The reader is referred to the Series 2 test plan (Einziger 1987) for a detailed description of these items. The main differences in the Series 2 and Series 3 tests are the type of fuel being tested and number of samples in the test matrix.

### 2.1 Fuel Characteristics

Series 3 will use high burnup fuel with high and low gas releases. High burnup PWR fuels manufactured by Combustion Engineering and irradiated in the Calvert Cliffs Unit reactor were acquired by the Materials Characterization Center (MCC) for use as ATM-104 and ATM-106. The fuel characteristics are given in Table 2.2. ATM-104 is thought to represent a high burnup, low fission gas release PWR fuel. ATM-106 is thought to represent a high burnup, high fission gas release BWR fuel.

**TABLE 2.1 Characteristics of High Burnup Calvert Cliffs PWR Fuel From Combustion Engineering**

<u>Fuel Type</u>	<u>ATM-IC4</u>	<u>ATM-106</u>
Assembly Identification	D047	BT03
Discharge Date	April 17, 1982	October 18, 1980
Nominal Burnup	~43Gwd/MTU	~43 Mwd/kgM
Fission Gas Release	1.1%	18%
Initial Enrichment	3.038%	2.453%
Initial Pellet Density	94-96% TD	92-24% TD
Initial Rod Diameter	1.118 cm	1.118 cm
Cladding Material	Zircaloy-4	Zircaloy-4
Cladding Thickness	0.066 cm	0.066 cm
Rod Identification	MKP-701	NBD131
Sample Location	*	*

\* Location of samples is as yet undetermined, the location will be recorded in the laboratory record book (LRB).

The MCC has characterized the rods (Guenther 1988 and 1987). Prior to cutting, the rods were punctured for both chemical and isotopic fission gas sampling. Gross and spectral gamma scanning were used to determine the burnup profile. Burnup analyses was conducted at three locations in the bottom half of the rod. Transverse and longitudinal ceramography examinations were performed adjacent to the burnup samples to determine grain size and to look for unusual features.

## 2.2 Test Matrix

The initial test matrix for the Series 3 tests, consisting of two sets of 27 samples each, is given in Table 2.2. The matrix in Table 2.2 will be repeated for the ATM-104 and ATM-106 fuel. The justification for the choice of variables that follows is given either in the Series 2 test plan (Einziger 1987) or below. The matrix may be adjusted to reflect the latest data from ongoing tests and data obtained as the test progresses.

**TABLE 2.2 Test Matrices for Series 3 (High Burnup Fuel) Dry Bath Oxidation Tests**

Temperature, °C	Atmospheric Moisture Levels, dew point°C	Number of Samples <sup>(a)</sup> of Size Indicated <sup>(d)</sup>			Test Duration yr (no. of samples)
		Fragments	-10/+24 Mesh	-24/+60 Mesh	
175	80	2			2 (2)
	-55	5	1	1	(b), (c)
130	80		2		2 (2)
	-55	1	5	1	(b), (c)
110	80			2	2 (2)
	-55	1	1	5	(b), (c)

- (a) Each Sample weighs ~10g
- (b) Single samples will go the full 2 yr.
- (c) Three of the multiple samples will go the full 2 yr; others will be removed for between 6,000 and 12,000 h for examination.
- (d) Maximum diameter of particle passing through each Tyler screen is:  
10 mesh = 1.7 mm, 24 mesh = 0.71 mm, 60 mesh = 0.25 mm.

### 2.3 Number of Samples and Test Duration

Tests under each set of temperature and atmospheric conditions will start with either two or seven samples containing fragments or pulverized fuel of different size fractions (see Test Matrix - Table 2.2). The mix of sample sizes was guided by the size fractions used in the sister drybath oxidation tests. The predominant sample type at each temperature is the smallest size fraction that is expected to yield substantial weight gains. The purpose of including a single sample of each of the remaining two size fractions is to confirm at lower temperature the particle size effects seen in the PWR fuel oxidation tests (Einziger and Buchanan 1987). No moisture effects are expected, so only two samples will be included in the high-moisture baths. These samples will be used for comparison with the five samples in the -55°C dew point atmosphere.

Including shutdown and startup time, each complete interim examination will require 5 days. To maximize the time-at-temperature, interim

examinations will be conducted approximately every 6 weeks to 2 months. This frequency of examination has worked satisfactorily with the other tests.

According to the test matrix (Table 2.2), either one, three, or five samples of any particular type (i.e., fragments, -10/+24 mesh, or -24/+60 mesh) will be run in a test. If five samples are used, two samples will be removed between 6,000 and 12,000 h. The three remaining samples will be tested for the duration, currently estimated to be 2 years. If two samples are used, both will remain for the duration of the test. If a single sample is used, it will remain for the full test duration. Additional samples may be placed under test, replacing those removed, if such action is indicated to be necessary by fuel examinations. The intervals for fuel sample removal may change as a result of the ceramographic, electro-optical, and leaching examinations.

### 3.0 REFERENCES

40 CFR 191, Dec. 29, 1982. Environmental Protection Agency, Proposed Rule, Vol. 47, No. 250.

10 CFR 60, June 30, 1983. "Disposal of High-Level Radioactive Wastes in Geologic Repositories--Licensing Procedures," Title 10, Chapter 1, Part 60.

Einzigler, R. E. and R. E. Woodley. 1985. Evaluation of the Potential for Spent Fuel Oxidation Under Tuff Repository Conditions. HEDL-7452, Hanford Engineering Development Laboratory, Richland, Washington.

Einzigler, R. E., 1985. Technical Test Description of Activities to Determine the Potential for Spent Fuel Oxidation in a Tuff Repository. HEDL-7540, Hanford Engineering Development Laboratory, Richland, Washington.

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

Einzigler, R. E., 1987. Test Plan for Long-Term, Low-Temperature Oxidation of BWR Spent Fuel. Pacific Northwest Laboratory

Guenther, R. J., D. E. Blahnik, T. K. Campbell, V. P. Jenquin, J. E. Mendel, and C. K. Thornhill, 1988. Characterization of Spent Fuel Approved Testing Material--ATM-106. PNL-5109-106, October.

Guenther, R. J., D. E. Blahnik, T. K. Campbell, U. P. Jenquin, J. E. Mendel, and C. K. Thornhill, 1987. Characterization of Spent Fuel Approved Testing Material--ATM-104. September.

Einzigler, R. E. and H. C. Buchanan, 1987. Interim Transition Report, Long-Term Low-Temperature Oxidation of PWR Spent Fuel. WHC-EP-007, Westinghouse Hanford Company, Richland, Washington.

**Appendix D - Estimated Staffing Requirements for Series 3 Oven Oxidation Tests\*.**

	<u><b>FTE</b></u>
Scientist (PNL)	.56
Secretarial (PNL)	.05
Technicians (PNL)	.08
Program Manager (PNL)	.42
Task Leader (LLNL)	<u>.10</u>
<b>Total FTE</b>	<u><b>1.21</b></u>

\* These estimates are based on currently planned Series 3 oven oxidation tests described in Appendix C.

Waste Form

1. "Plan for Spent Fuel Waste Form Testing for NNWSI" dated November 1987
2. Activity Plan titled "Low-Temperature Oven Method for Spent Fuel Oxidation Testing" Sub-activity D-20-45 Rev. 0

Applicable Implementing Procedures:

1. SFO-1-1 titled "Sample Preparation for Spent Fuel Oxidation Testing Using a Dry Bath Heating System" Rev. 0
2. SFO-1-2 titled "Measurement of Spent Fuel Oxidation Using a Dry Bath Heating System" Rev. 2
3. HTA-3-1 "Solids Analysis: Scanning Electron Microscopy" Rev. 4
4. HTA-3-2 "Solids Analysis: Transmission/Scanning Transmission Electron Microscopy" Rev. 4
5. HTA-3-3 "Solids Analysis; X-Ray Diffraction Analysis" Rev. 3

## CONTENTS.

1.	Purpose .....	1
2.	Scope .....	1
3.	Comment .....	1
4.	Introduction to the constant extension rate technique .....	2
5.	Apparatus .....	3
	5.1 List of equipment and materials .....	3
	5.2 Special environmental conditions .....	3
	5.3 Test Vessel .....	4
	5.4 Temperature Control .....	4
	5.5 Calibrations .....	4
6.	Test Specimen .....	5
	6.1 Specimen configuration and preparation .....	5
	6.2 Identification .....	6
	6.3 Dimensional .....	6
	6.4 Cleaning .....	6
7.	Materials Properties .....	6
8.	Test Procedure .....	7
	8.1 Number of Tests .....	7
	8.2 Test method .....	7
9.	Range, Accuracy and Precision .....	8
10.	Recording of results .....	9
11.	Personnel Responsibilities .....	10
12.	Quality Assurance Records .....	11
	References .....	12

## 1. PURPOSE

This procedure establishes the general method for using the Constant Extension Rate technique to evaluate the stress corrosion behavior of the six (6) metal barrier candidate materials.

## 2. SCOPE

This procedure is part of the electrochemical corrosion testing of the proposed metal barrier container materials for the Yucca Mountain Project (YMP). This procedure will be used during the conduct of Activity E-20-18h, 'Determination of Stress Corrosion Cracking Susceptibility Using The Constant Extension Rate Technique'. The procedure identifies the six (6) metal barrier candidate materials and describes the test specimens and their material properties. It discusses the test equipment to be used and the test procedures to be followed. Reporting of the test results is also discussed.

NACE Standard TM-01-77 [1] was used as a guide in the preparation of this procedure.

## 3. COMMENT

This procedure is prepared in accordance with procedures of the LLNL/YMP Quality Assurance Program Plan to permit acquisition of data that may be applicable to the selection criteria being completed under Activities E 20-15 'Establishment of Criteria for Metal Barrier Selection' and E 20-19 'Metal Barrier Selection'. This procedure will be used during the conduct of Activity E-20-18h, 'Determination of stress corrosion susceptibility using the constant extension rate technique'.

The six candidate materials are CDA 102 (Oxygen-free copper), CDA 613 (7% minimum bronze), CDA 715 (Cu-30Ni), stainless steels Type 304L and 316L, and Alloy 825 [2]. One of these materials could be used to fabricate the metal container for high level radioactive waste at the proposed repository in Yucca Mountain, Nevada. The test data shall be used in ranking the six candidate materials on the basis of stress corrosion cracking. The data shall also be used for model development. 60

#### 4. INTRODUCTION TO THE CONSTANT EXTENSION RATE TECHNIQUE.

The constant extension rate technique has been used as a means of determining the susceptibility of alloys to stress corrosion cracking (SCC) and for screening metal alloys for material selection. In essence, the constant extension rate technique comprises straining a specimen at a slow, constant extension rate under controlled environmental conditions.

This accelerated test method is consistent with the proposed general mechanisms of the SCC process, most of which involve plastic microstrain and film rupture. Based on experimental evidence, it has been suggested that the most important role of stress is the dynamic strain it produces [3]. Typical strain rates range from  $10^{-4}$  to  $10^{-6}$ /s. Strain rates in the critical range to promote SCC maintain the delicate balance at the crack tip between deformation, dissolution, film formation and diffusion.

There is a critical potential range for a material in an environment which promotes SCC, and exposure within this range is necessary for cracking to occur. The specificity of environments which promote SCC has long been recognized. Constant extension rate tests provide a technique to investigate the effects of solution composition or addition to a base solution on the severity of SCC. Temperature is also an important parameter. In some metal-environment systems SCC is more severe as temperature increases, whereas, in some other systems SCC is more severe at lower temperatures.

## 5. APPARATUS

5.1 List of equipment and materials. The equipment and materials listed below are needed for this procedure. Equivalent items may be substituted unless otherwise noted. Purchase of materials is to comply with 033-YMP-QP-4.0, "Procurement Control and Documentation."

Required M&TE include: Constant extension rate testing machines, (for example Cortest 34000 Floor Model and Instron Model 8562), load cells, linear velocity displacement transducer, pH meter, potentiostats, strip chart recorder, extensometer, digital calipers, thermocouples and saturated calomel or mercury/mercury sulfate reference electrodes. An Olympus metallurgical measuring microscope and PAR model 273 potentiostat will be ordered in the near term. The DOE identification numbers and the calibration records will be identified in the scientific notebook. All samples tested must be procured as specified in quality procedure 033-YMP-QP-4.0, 'Procurement' and controlled and identified as specified in quality procedure 033-YMP-QP-8.0, 'Items, Samples and Data'.

5.2 Special environmental conditions. Electrolytes used for testing will be prepared so as to maintain the same relative concentrations of ions as found in water from well J-13, if possible. Absolute concentrations may be greater or less than those found in water from J-13 (reference condition). Measurements in other aqueous environments (NaCl solutions, etc) will be made if necessary. Tests may also be conducted in a vapor-phase environment containing NO<sub>x</sub> species; the environmental variables will include temperature, partial pressure of water and partial pressure of NO<sub>x</sub> species. Solutions may be refreshed when necessary.

All chemicals shall be reagent or chemically pure (99.5% minimum purity).

The water shall be distilled or deionized and of quality equal to or greater than ASTM Type IV (ASTM D1193) [4].

### 5.3 Test vessel.

The size, shape, and entry ports of the test vessel will be determined by the actual test specimens, auxiliary electrodes and test fixtures used to stress the specimens.

Test vessels shall be constructed from materials that are inert to the test environment. For tests at temperatures above 95°C autoclaves should be used.

5.4 Temperature control. Tests at elevated solution temperature should be maintained within  $\pm 5.0^{\circ}\text{C}$ .

5.5 Calibrations. All M&TE must be calibrated as specified in quality procedure 033-YMP-QP-12.0. The Calibration records will be identified in the scientific notebook.

## 6.0 TEST SPECIMEN.

### 6.1 Specimen configuration and preparation.

6.1.1 The selection of the test specimens that can be used is often restricted by the size and shape of the material available for testing. For tests at elevated temperatures the selection of test specimen is also governed by the type of autoclave available. The orientation of the specimen can affect the results and should be noted.

6.1.2 The gage length of the tension specimen (Fig.1) shall be 0.1in (2.54mm) in diameter and 1.00 in (25.4mm) long. Specimens with gage of 0.25in (6.35mm) in diameter and 1.0in (25.4mm) long may also be used. The dimensional tolerances shown in Fig.1 are to be followed in specimen preparation.

6.1.3 The radius of curvature at the ends of the gage sections shall be at least 0.25in. (6.35mm) to minimize stress concentrations.

6.1.4 Ends of the specimen must be long enough to accommodate seals for the test container and to make connections to the stressing fixture.

6.1.5 Machining of the specimens must be done carefully to avoid overheating and cold working in the gage section.

6.2 Identification. Stamping or vibratory stenciling may be used on the ends of the specimen, but not in the gage section. Specimens may also be identified by placing them in clearly marked bags. TIP-CM-06 'Identification and Control of Metal Specimens', establishes general methods for proper identification, care, and documentation of metal test specimens.

6.3 Dimensional check. The gage diameter of each specimen will be measured and recorded for later use in computing reduction in area.

6.4 Cleaning. Prior to testing, specimens will be degreased in acetone and rinsed in methonal.

#### J MATERIALS PROPERTIES.

Tensile testing in an inert environment shall be used to determine base material properties. Two or more specimens shall be pulled and the results averaged to determine the yield and ultimate strengths, percent elongation, time to failure and percent reduction in area.

Tensile properties will be obtained at all the strain rates that shall be used during slow strain rate testing.

## 8.0 GENERAL PROCEDURE.

8.1 Number of tests. A certain amount of data scatter is inevitable in corrosion test results. The amount of scatter depends on the uniformity of test material, the condition of the specimen surface, and the stability of the exposure conditions. All of these factors influence the precision of corrosion tests. Although, the constant extension rate method is a very reproducible technique, certain variables require duplicate tests. For example when the effect of strain rate is being investigated, duplicate tests are advisable, whereas when the effect of potential is considered (tests at a number of different potentials), only test results that exhibit data departing from the general trend need to be repeated.

## 8.2 Test method.

8.2.1 Adjust testing machine to the appropriate cross head speed.

8.2.2 Place a clean specimen in the test vessel, make the necessary seals and then assemble the vessel in the testing machine.

8.2.3 Manually apply a load to secure the fixturing about the test vessel. Ensure that the yield stress is not exceeded.

8.2.4 Fill the test cell immediately with the test environment and heat the test environment to the desired temperature, and apply a potential to the cell if necessary. The initial value of solution pH should be recorded and the final pH of the solution should also be recorded at the completion of the test.

8.2.5 Apply a load to the specimen using the testing machine at the desired cross head speed.

8.2.6 The termination of the test shall be at specimen failure or at a pre-determined strain value.

## 9.0 RANGE, ACCURACY AND PRECISION

The range to be investigated with regards to stress/strain data (or load/% extension) will extend from a minimal load and essentially zero strain up to the ultimate tensile strength for load, and maximum strain measurement at specimen failure. Load is being monitored via the load cell and strain measured via extension transducers. The load and strain data will not exceed the operating specifications of the load cell or extension transducers.

The range of potential to be explored will depend on data generated from sub-activity E-20-18a, 'Use of Linear-sweep Polarization To Determine Pitting Potential', but will include the open circuit potential. However, the range of potentials are limited to the operating specifications of the PAR Model 273 potentiostat ( $\pm 2$  volts). This range is adequate for the testes required to be conducted.

The range of temperature measurements will extend from ambient conditions to 150°C. The range of temperatures will not exceed the operating specifications of the thermocouples.

The range of the linear measurements obtained from the samples will exceed the operating specifications of the measuring devices such as digital calipers and measuring microscopes.

The precision of the stress strain data is a function of the bias and accuracy of the load cell and extension transducers as well as the recording devices used to produce the stress-strain data. In addition, the accuracy of the extension transducer is a function of the axial stiffness of the load frames and load cells.

The precision of the potential, temperature, and linear measurements is a function of the bias and accuracy of the potentiostat and saturated calomel or mercury/mercury sulfate electrodes, thermocouples, and digital caliper and measuring microscopes.

The accuracy of the various measurements will be recorded in the scientific notebooks.

## 10. RECORDING OF RESULTS.

One of the parameters that shall be used to assess SCC susceptibility is stress-elongation curves; therefore crack susceptibility can be expressed by maximum load, as well as the usual measures of ductility, elongation and reduction in area.

Time to fracture or fracture energy (area under load-extension curve) may also be used to indicate crack susceptibility.

The above results are normalized in terms of a ratio of the ameters measured in the corrodent to measurements taken in an inert environment under the same conditions. Thus increasing susceptibility is marked by an increasing departure from unity.

Metallographic examination provides the most positive determination of SCC. From metallographic examination the crack velocity can be determined by dividing the deepest crack detected in the specimen after the completion of the test by the time to failure measured from yield.

Direct microscopical examination of fracture surfaces with the scanning microscope is also helpful in determining the cause of fracture. Sub-activity E-20-18e 'Microstructural Characterization', describes the methods to be used for microscopical examination.

## 11. PERSONNEL RESPONSIBILITIES.

The Task leader (TL) whose activities warrant the use of this procedure is responsible for implementing the requirements of this procedure. The Task Leader may delegate this responsibility and authority to a Principal Investigator.

The individual determining the susceptibility to stress corrosion cracking is responsible for following the requirements of this procedure, documenting calibrations, and assuring that the latest revision of this document is followed.

The YMP Quality Assurance Manager (QA Manager) is responsible for monitoring work to assure proper implementation of this procedure and to assure its continued effectiveness.

Copies of this procedure are available from both the Task Leader and the Principal Investigator. Suggestions for revisions or any questions regarding the procedure can be directed to either the Task Leader or the Principal Investigator.

## 12. QUALITY ASSURANCE RECORDS.

The documents produced by this procedure are: (1) this procedure; (2) the scientific notebook; (3) the data base; and (4) Identification and control of the samples.

The scientific notebook will identify the calibration status and the calibration procedure (including revisions) used to perform each calibration.

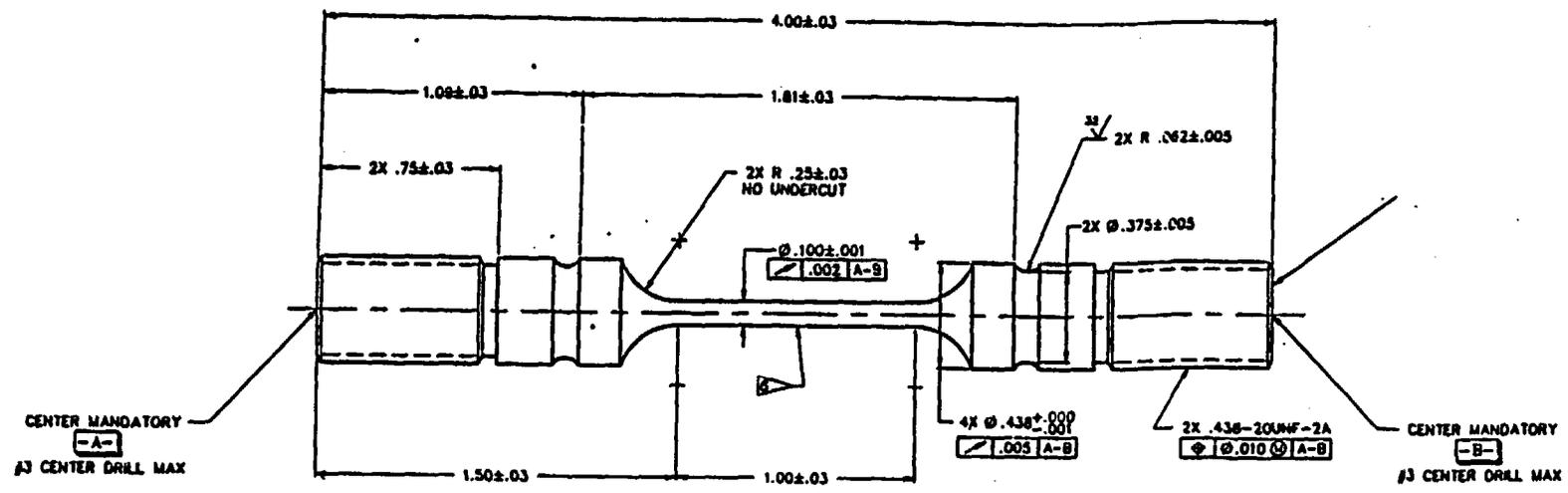
Determinations of stress corrosion cracking susceptibility will be recorded in the scientific notebook in accordance with procedure 033-YMP-QP 17.0, "Quality Assurance Records".

REFERENCES.

1. NACE Standard TM-01-77, "Testing of metals for resistance to sulfide stress cracking at ambient temperatures." NACE Houston, Texas 1977.
2. UCID-21362, 1989, Survey of Degradation Modes of Candidate Materials for High-Level Radioactive-Waste Disposal Containers, Lawrence Livermore National Laboratory, Livermore, California 94550.
3. STP 665 ASTM 1979 p5-25, R.N.Parkins "Stress Corrosion Cracking- The slow strain rate Technique,
4. Standard ASTM D1193, "Standard Specification for Reagent Water," American Society for Testing and Materials (ASTM), Philadelphia, Pennsylvania, 1989.

Figure I.  
Typical Constant Extension Rate Specimen

- NOTES  
UNLESS OTHERWISE SPECIFIED:
1. ALL DIMENSIONS ARE IN INCHES.
  2. DIMENSIONING AND TOLERANCING PER ANSI Y14.5M-1982.
  3. SURFACE TEXTURE PER ANSI B46.1-1978.
  4.  $\sqrt{R}$  ALL MACHINED SURFACES
  5. BREAK SHARP EDGES .010±.005 R OR CHAMFER
- $\nabla$  POLISH REDUCED SECTION TO REMOVE TOOL MARKS



Dwg No		PART / DATE / REV		(SEE DWG NO. 85-105213)		DESCRIPTION / MATERIAL		SPEC NO	ITEM
DWGN	P. F. SOUZA	3/83		CLASSIFICATION		W-87			
CHKD	Tom Connors	3/83		THE BOONER IS THE PROPERTY OF					
APPR	S. GORDON	3/83		THE UNIVERSITY OF CALIFORNIA					
DATE				LAWRENCE LIVERMORE NATIONAL LAB					
				REPRODUCTION PROHIBITED WITHOUT					
				PERMISSION OF THE MECHANICAL					
				ENGINEERING DEPARTMENT,					
				UNIVERSITY OF CALIFORNIA					
						DRAWING NO			
						85-105213			
						DATE			
						6873-10			
						SCALE			
						3/8"			
						SHEET 1 OF 1			

A	DATE	BY	CHKD	APPD	DATE	REVISION

## Metal Barriers

1. SIP No. SIP-NF-15 titled "Engineered Barrier System Field Tests-Prototype Tests" WBS 1.2.2.3.2 Rev. 0
2. Activity Plan titled "Establishment of Selection Criteria" Sub-activity E-20-15 Rev.0
3. Activity Plan titled "Parametric Studies: Use of Linear-Sweep Polarization to Determine Pitting Potentials" Sub-activity E-20-18a Rev. 0
4. Activity Plan titled "Parametric Studies of Metal Degradation and Microstructure: Measurement of Plane-Strain Fracture Toughness" Sub-activity E-20-18c Rev. 0
5. Activity Plan titled "Parametric Studies of Metal Degradation and Microstructure: Measurement of Threshold Stress Intensity for Stress Corrosion Cracking" Sub-activity E-20-18d Rev. 0

## Applicable implementing procedures:

1. TIP-CM-1 titled "Determination of Plane-Strain Fracture Toughness and the Threshold Stress Intensity for Stress Corrosion Cracking" Rev. 0
2. TIP-CM-2 titled "Operator Calibration of Scanning Transmission Electron Microscopes" Rev. 0
3. TIP-CM-3 titled "Operator Calibration of Scanning Electron Microscopes" Rev. 0
4. TIP-CM-4 titled "Operator Calibration of the Optical Metallograph" Rev. 0
5. TIP-CM-05 titled "Determination of Threshold Stress Intensity for Stress Corrosion Cracking Using Modified WOL Specimens" Rev. 0
6. TIP-CM-6 titled "Identification and Control of Metal Specimens" Rev. 0
7. TIP-CM-07 titled "Determination of Stress Corrosion Cracking Susceptibility Using the Constant Extension Rate Technique" Rev. 0