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

October 16 , 1986

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Mr. Everett A. Wick
Division of Waste Management
Office of Nuclear Materials Safety and Safeguards
U.S. Nuclear Regulatory Commission
Washington, DC 20555

Re: Monthly Letter Status Report for September 1986 (FIN-A-4171-6)

Dear Mr. Wick:

Enclosed is the September 1986 monthly progress report for the project "Evaluation and Compilation of DOE Waste Package Test Data" (FIN-A-4171-6). The financial information is reported separately.

Sincerely,

Charles G. Interrante

Charles G. Interrante
Program Manager
Corrosion Group
Metallurgy Division

Enclosures

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Monthly Letter Report for September, 1986

Published October 1986

(FIN-A-4171-6)

Performing Organization: National Bureau of Standards
Gaithersburg, MD 20899

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

TASK 1 -- Review of Waste Package Data Base

Appended to this report are copies of five Draft Reviews not submitted earlier. Comments by NRC and its contractors are solicited.

1. HEDL-SA-3271P, "Low Temperature Spent Fuel Oxidation Under Tuff Repository Conditions," -- Review revised date 7/29/86

2. HEDL-7452, "Evaluation of the Potential for Spent Fuel Oxidation under Tuff Repository Conditions," -- Review revised date 8/6/86

3. UCRL-94222, "Important Radionuclides in High Level Nuclear Waste Disposal: Determination using a Comparison of the EPA and NRC Regulations," -- Review revised date 8/11/86

4. HEDL-7556, "Test Plan for Series 2 Thermogravimetric Analysis of Spent Fuel Oxidation," -- Review revised date 8/11/86

5. HEDL-7577, "Test Plan for Series 3 NNWSI Spent Fuel Leaching/Dissolution Tests," -- Review revised date 8/23/86

Purchase orders have been prepared for hardware and software to be used in the establishment of a computer assisted database for reviews. These orders will be processed at the beginning of FY 87 and we expect that the units will be fully operational by the end of October, 1986.

An intensive effort was undertaken to engage outside consultants who could assist NBS personnel in their work on the HLW project. These consultants must not only meet extremely rigorous standards established by NRC with respect to potential conflicts of interest but must also possess the expertise necessary to make a significant contribution to the program. To date, we have selected Dr. D. Cronin, Mr. B. Adams and Prof. M. Rosen who satisfy these criteria. Contracts are now being drawn up and we expect them to be working with us in the near future.

Revision of Structure of Keyword Checklist

A revised keyword checklist was developed from the keyword checklist submitted earlier. This revision has an improved hierarchy that more clearly directs both the reviewer and the user of the database. Some redundancies were eliminated, some keywords were added, and this improved hierarchy should facilitate searches of the most pertinent papers for a given topic. The original checklist seemed confusing to some reviewers because too many of the same terms appeared in several categories with no distinction made between them. For example, the same terms appeared under the heading of the material studied, under the environment of the study, under materials found in the environment, etc. and the category under which the keyword term should be entered was not clear to the reviewer. The lack of categorization also would have presented difficulties for the user of the computer database. Without a specific designation of the category to which a keyword belonged, the user would have retrieved every occurrence of a keyword with no distinction made as to its significance.

To eliminate the problem, categories for the various keywords were established and an outline tree was prepared listing the various groupings (see attached). In the attached tree, the categories which are underlined are those in which the keywords occur on the checklist. The bold face terms designate the broad topical categories to which the keywords belong. The original keyword list was carefully checked through to ensure that none of the keywords would be omitted from the new list. The revised keyword checklist allows for the addition of new keywords as well as the addition of new categories as needed. The NBS workers are currently reviewing and revising the checklist, before it is incorporated into our instructions for reviewers. Both the tree and the revised checklist are attached for your comments and suggestions.

SRP -- The following reports have been identified and assigned for review:

1. DOE/CH/10140-03(85-4), "Salt Repository Project Technical Progress Report for the Quarter, July 1-September 30, 1985, prepared for the U.S. Department of Energy by the Office of Nuclear Waste Isolation, Battelle Project Management Division

2. DOE/CH/10140-03(86-1), "Salt Repository Project Technical Progress Report for the Quarter, October 1-December 31, 1985, prepared for the U.S. Department of Energy by the Office of Nuclear Waste Isolation, Battelle Project Management Division

SRP -- The following report is under consideration and will be reviewed when a suitable reviewer is identified:

BMI/ONWI-612, "The Effects of Stabilizers on the Heat Transfer Characteristics of a Nuclear Waste Canister," K. Vafai and E. Ettenfagh, July 1986

BWIP -- Reviews have been initiated on the following reports this month:

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

2. B036177, "Analysis of the Effects of Radiation on the Chemical Environment of a Waste Package in a Nuclear Waste Repository in Basalt," W.J. Gray, March 1984

3. SD-BWI-DP-060, "Interim Data Document for the Advanced Conceptual Design of High-Level Waste Packages for a Repository in Basalt," W.J. Anderson, Nov. 1984

4. RHO-BW-SA-280P, "Conceptual Design of a Waste Package for Emplacement in Basalt," W.J. Anderson, Feb. 1983

5. RHO-BW-SA-316, "Irradiation-Corrosion Evaluation of Metals for Nuclear Waste Package Applications in Grande Ronde Basalt Groundwater," J.L. Nelson, Nov. 1983

6. SD-BWI-TI-235 (B032012), "Corrosion Evaluation of Candidate Iron-Based Nuclear Waste Package Alloys in Grande Ronde Basalt Groundwater," R.E. Westerman, Feb. 1984

7. HEDL-7612, "Corrosion of Copper-Based Materials in Gamma Radiation," W.H. Yunker, June 1986

NNWSI -- The following reports have been identified for review and will be reviewed as soon as a suitable reviewer has been identified:

1. UCRL-15825, "The Effect of Gamma Radiation on Groundwater Chemistry and Glass Leaching as Related to the NNWSI Repository Site," T. Abrajano, J. Bates, W. Ebert, and T. Gerding, Lawrence Livermore Laboratory, May 1986

2. UCRL-53735, "The Tuff Reaction Vessel Experiment," F. Bazan and J.H. Rego, Lawrence Livermore Laboratory, June 1986

TASK 2 -- Identification of Additional Data Required and Identification of Tests to Generate the Data.

Over the summer months, consideration has been given to the merits of asking the National Materials Advisory Board of the National Research Council to conduct a study that has potential for being of value to us in our work on Task 2. A copy of a preproposal dated September 26, 1986 on "Integrity of Engineered Barriers for High-Level Radioactive Waste," which we have developed with NMAB's Stan Wolf, is enclosed for you to review before meeting with us to discuss this in October.

NBS lead workers are continuing their studies concerning the types of additional data and verification tests needed to demonstrate that the DOE waste package designs will meet the performance objectives of 10 CFR 60. Presently, no parts of the NBS preliminary plans for laboratory tests to be conducted at the NBS under Task 3 of this contract have been approved. Two of the proposals submitted earlier are being redrafted and will be resubmitted in October.

TASK 4 -- General Technical Assistance

Dr. Richard Ricker visited Battelle in Columbus, Ohio on September 4 to 5, 1986 to meet with staff to discuss DOE/NRC High Level Waste Program. At these meetings, the following four presentations were given:

1. D. Stahl, "Long Term Performance of Materials Used for High-Level Waste Packaging"
2. J.A. Beavers, N.G. Thompson, and R.N. Parkins, "Container Materials"
3. A.J. Markworth, J. Kevin McCoy, and D.D. Macdonald, "Analysis of Container Corrosion"
4. H.J. Cialone, "Hydrogen Embrittlement of Containers for High-Level Waste-Containment"

The NBS was represented, by Dr. M. Linzer and a guest (of NBS) worker Prof. A. Sembira, at "Spectrum '86," the American Nuclear Society International Topical Meeting on "Waste Management and Decontamination and Decommissioning," which was held in Niagara Falls, NY, September 14 to 18, 1986. Because of the large number of papers, simultaneous sessions were held. The High Level Waste sessions, which were the focus of NBS' interest, featured talks on international vitrification projects, plans and system testing, off-gas behavior and processing. The work of the Catholic University groups, carried out under the direction of Dr. Macedo, appeared to be the highest quality of the research presented on glass leaching. A tour of the West Valley Demonstration Project on September 18 was very informative and a highlight of the meeting. Our trip report will be furnished after the abstracts of the papers for this meeting become available, probably early in December.

Draft Copy

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

DATA SOURCE:

(a) Organization Producing Data: Westinghouse Hanford Company, Richland, Washington 99352

(b) Author(s), Reference, Reference Availability: Einziger, R. E., and Woodley, R. E., "Low Temperature Spent Fuel Oxidation Under Tuff Repository Conditions", HEDL-SA-3271FP (1985).

DATE REVIEWED: 7/29/86

TYPE OF DATA: Experimental rate of oxidation of spent fuel (UO_2). Theoretical analysis of mechanism of oxidation to determine whether grain boundary or bulk diffusion.

MATERIALS/COMPONENTS: PWR spent fuel (UO_2) from PWR Turkey Point unit 3.

TEST CONDITIONS: PWR (UO_2) spent fuel in moist air (16000 ppm) and dry air (3 ppm) using fragments or pulverized fuel at temperatures of 200-225°C up to 737 hrs.

METHODS OF DATA COLLECTION/ANALYSIS: Oxidation rate by weight gain vs time at fixed temperature and atmospheric pressure in air. Thermogravimetric and ceramographic analysis. (Ceramographic analysis involves study of micrographs of non-metallic materials).

AMOUNT OF DATA: 7 figures and 4 tables. Figures; Phase diagram of O/U from O/U of 2 to 3 and temperatures from 0 to 1000°C., Schematic of oxidation rate vs 1/T when oxidation mechanism changes at temperature below experimental range., Schematic of Thermogravimetric Analysis (TGA) system., TGA test sample weight changes, % weight change and change in $^{16}O/M$ or $^{18}O/M$ vs time in hours., 2 figures of polished cross sections of UO_2, Plot of $[1-(1-C)^{1/3}]$ vs $t^{1/2}$ showing that (bulk) diffusion limits the oxidation rate. Tables; Free-energy change for oxidation of UO_2 to U_3O_8 and U_3O_8 to UO_3, Characteristics of Turkey Point unit-3 fuel., Experimental test parameters., Results of preliminary oxidation tests.

UNCERTAINTIES IN DATA: Not quantitatively discussed.

DEFICIENCIES/LIMITATIONS IN DATABASE: The authors state that the conclusions are preliminary. Additional testing, for longer times and at lower temperatures, is needed before spent-fuel-oxidation mechanisms are established and extrapolation of oxidation behavior in a tuff repository can be made.

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

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

Related to issue 2.3.6 in the ISTP for the Nevada Nuclear Waste Storage Investigation (NNWSI) Project.

(b) New Licensing Issues

(c) General Comments

KEY WORDS: Experimental data, TGA, Water vapor, Spent fuel, Oxidation rate, Degradation (spent fuel), Mechanism of oxidation

GENERAL COMMENTS: This work demonstrates that oxidation of spent fuel in breached fuel canisters may take place, but the test temperatures are thought to be somewhat higher than the estimated repository temperatures. The repository temperature is estimated to be 145-160°C after 300 years. I have several criticisms of this work. Athough the work is preliminary, there is no error analysis of the temperature dependence of the activation energy. I do not see how the oxidation rate measure on bare fuel can be related to fuel in intact zircaloy cladding. The air flow rates used in the measurements of 400 cc/min are probably too high. It is possible that in the repository the rate of arrival of oxygen at the surface could be the rate determining step. If the leach rate of higher uranium oxides is an important consideration measurements of this leach rate should be obtained.

Draft Copy

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

DATA SOURCE:

(a) Organization Producing Data: Hanford Engineering Developement Laboratory, P. O. Box 1970, Richland, Washington 99352

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

DATE REVIEWED: 8/6/86

TYPE OF DATA: Review of literature data concerning oxidation of spent fuel in breached zircaloy cladding.

MATERIALS/COMPONENTS: Spent fuel.

TEST CONDITIONS: Conditions possible in a tuff repository which would impinge on the oxidation of spent fuel in rods with breached cladding are discussed. These include water, moist air or inert gas, temperature, and radiation field, as a function of time.

METHODS OF DATA COLLECTION/ANALYSIS: Scenarios based on literature data and assumed conditions in the repository are reviewed.

AMOUNT OF DATA: Twelve figures and 5 tables. Figures; Phase diagram, O/U ratio from 2 to 3, temperature 0-1000°C; Log P (O_2) (atm), (-10 to -50) vs T°C (0 -300); Time to spallation (1-10000 hrs.) vs 1/T(K) (360-190°C), measure of oxidation rate of UO_2; Summary of conditions for fuel oxidation experiments; Percent weight increase (0-4) vs time (0-700) for oxidation of CANDU irradiated or unirradiated fuel at 230 and 250°C; Log rate of weight increase in %/hr (10^{-5},-1) vs 1/T(K) (395-180°C) for spent fuel and UO_2 in air; Initial potential tuff repository environmental and waste package condition; Summary of time dependence of tuff repository environmental and waste package conditions, 10-10^4 yr vs atm, cannister condition, rod integrity, fuel temperature and irradiation field; Rate of oxidation of spent fuel vs 1/T for two possible oxidation mechanisms with lower temperature mechanism having lower activation energy (schematic); Spent fuel scenarios 0-1000 yrs, showing possible pathways dependent on initial conditions; Cumulative damage fraction vs year breach occurs (used to estimate if breached cladding will split in 300 yrs); Spent fuel disposal scenarios beyond 300 years dependent on radiation field, temperature, atmosphere, moisture, and cannister condition; Tables; Free energy changes for oxidation of UO_2 and U_3O_8 from 25-250°C; List of bare fuel oxidation experiments and conditions from literature; List of literature oxidation experiments on defected rods; Gamma field as function of time; Summary of fuel oxidation evaluation for time and atmosphere conditions.

UNCERTAINTIES IN DATA: Error bars on oxidation-rate data of up to 20%. There may be real differences in fuels dependent on previous history. With respect to correlating different methods of measuring oxidation rate the authors state, "It is difficult to relate the measures to one another since, in many cases, insufficient data are measured."

DEFICIENCIES/LIMITATIONS IN DATABASE: The effect of radiolysis of moisture on spent-fuel-oxidation rates and oxidation products formed in an inert atmosphere need to be determined. Low-temperature oxidation studies of UO_2 to determine low-activation-energy processes are needed.

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

(a) Relationship to Waste Package Performance Issues Already Identified

Related to issue 2.3.6 in the ISTP for the Nevada Nuclear Waste Storage Investigation (NNWSI) Project.

(b) New Licensing Issues

(c) General Comments

KEY WORDS: literature review, tuff, water vapor, zircaloy, spent fuel, spalling, oxidation of spent fuel

GENERAL COMMENTS: This work focusses on the oxidation of UO_2 in breached cladding when stored in an air filled repository. The essential points are that oxidation of the spent fuel, which forms oxides of lower density, may lead to splitting of the cladding. The contention is that higher oxides of uranium may leach at higher rates than UO_2, although data corroborating this contention is apparently minimal. The authors state that only about 0.01% of the fuel rods contain defects in the form of small pinholes or splits. As oxidation of the spent fuel can only occur in rods with defective cladding, clearly this problem is closely related to the cladding lifetime.

Draft Copy

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

DATA SOURCE: Prepared for submittal to Nuclear and Chemical Waste Management.

(a) Organization Producing Data: Earth Sciences Department
Lawrence Livermore National Laboratory, Livermore, CA 94550

(b) Author(s), Reference, Reference Availability: Oversby, Virginia M., "Important Radionuclides in High Level Nuclear Waste Disposal: Determination using a Comparison of the EPA and NRC Regulations", UCRL-94222, Preprint, February, 1986.

DATE REVIEWED: August 11, 1986

TYPE OF DATA: Analysis of amount and decay of radionuclides in spent fuel.

MATERIALS/COMPONENTS: PWR spent fuel.

TEST CONDITIONS: Calculations based on data for PWR spent fuel.

METHODS OF DATA COLLECTION/ANALYSIS: The analysis identifies those radionuclides most likely to be important in meeting NRC (10 CFR Part 60) and EPA (40 CFR Part 191) regulations pertaining to licensing and performance objectives for nuclear waste repositories.

AMOUNT OF DATA: Eight tables; Table 1 gives the inventory of significant radionuclides in PWR spent fuel at 10, 100, 1,000, and 10,000 years in Ci/MTIHM (Curies/Metric Ton of Initial Heavy Metal). Also included is the inventory at 300 years for those nuclides changing greatly in abundance between 100 and 1000 years; Table 2, Performance objective for control of release rates, lists nuclide, inventory at 1000 yr, 1/100,000 of 1000 yr inventory, and level to which release rate must be controlled in Ci/MTHM per yr, ratio of controlled release rate to 1/100,000 inventory; Table 3, Comparison of release rates allowed in 10CFR60 to the cumulative release limits in 40CFR191, lists nuclide, Table 1 EPA limit in Ci/MTIHM (from Table 1 EPA 40CFR191), release under 10CFR60 in Ci/9000 yr, ratio of NRC/EPA, factor to reach R=0.035, (.035 is used to insure that no more than .035 of the total EPA limit is due to each radionuclide); Table 4, Comparison of the 1,000 year inventory of radionuclides to the EPA release for low probability events, lists nuclide, 10 times EPA limit table 1 and inventory at 1000 years in Ci/MTIHM, ratio of inventory to 10 x EPA table 1, factor to reduce ratio to R=.035; Table 5, Reduction factors for maximum NRC allowed releases, radionuclides listed by order according to the amount by which their release exceeds the amount that could be released to the accessible environment, assuming that each radionuclide accounts for no more than 0.035 of the total EPA limit; Table 6, Comparison of releases controlled by matrix dissolution at 1/100,000 per year for 9000 years with EPA cumulative release limits; lists nuclide, release for 9000 yr and EPA Table 1 limit in Ci/MTHM, ratio of release to EPA limit, factor to reduce ratio to .035; Table 7, Reduction factors for release at the rate of 1/100,000 per year for all radionuclides plus a 1% spiked release for CS, C, I, and Tc in the 1000 yr inventory, lists nuclides and factors by which release at edge of engineered barrier system exceeds EPA limit assuming no radionuclide accounts for more than .035 of EPA limit; Table 8, Same as table 6 except matrix dissolution is at 1/1,000,000. All values except the EPA (Table 1) limit are reduced by a factor of 10.

UNCERTAINTIES IN DATA: Not discussed.

DEFICIENCIES/LIMITATIONS IN DATABASE: None mentioned

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

(a) Relationship to Waste Package Performance Issues Already Identified

Related to issues 3.3.1 and 3.3.1.2 in the ISTP for the Nevada Nuclear Waste Storage Investigation (NNWSI) Project.

(b) New Licensing Issues

(c) General Comments

KEY WORDS: EPA regulations, NRC regulations, repositories, commercial high level waste, spent fuel, (power reactors)

GENERAL COMMENTS: This report analyzes the radioactive waste inventory of a generic repository with respect to EPA regulations governing the total release to the accesible environment and the NRC regulation governing rate of release from the repository. Seventeen elements are identified as materials for which data on solubility and sorption would be needed for use in cite performance assessment but by far the most important elements are americium and plutonium. Note that the EPA regulation puts a limitation on release to the accesible environment in terms of Ci/MTIHM starting 1000 years after closure of the repository, while the NRC regulation requires that less than 1/100,000 per year of the inventory at 1000 years, is released from the engineered barrier system. Under these regulations, release into the environment is only allowed after 1000 years so that ^{137}Cs which presents problems during initial handling of spent fuel has essentially decayed away by the time these regulations come into effect. Note: In this report the term Ci/MTIHM is used interchangeably with the term Ci/MTHM.

Draft Copy

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

DATA SOURCE:

(a) Organization Producing Data: Westinghouse Hanford Company
Richland, Washington 99352

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

DATE REVIEWED: 8/5/86

TYPE OF DATA: Planned work involving oxidation of spent fuel.

MATERIALS/COMPONENTS: Turkey Point spent fuel.

TEST CONDITIONS: Oxidation in air or N_2 and $^{18}O_2$ at 175 and 155°C with 3, 1000, or 16000 ppm water vapor.

METHODS OF DATA COLLECTION/ANALYSIS: Rate of oxidation measurements will be made by thermogravimetric analysis (TGA). Samples to be characterized by ceramography, XRD, SEM, and ion microprobe.

AMOUNT OF DATA: Three tables and 6 figures. Figures; Flow sheet showing technical approach to measurements; Selection of test data; Temperature dependence of the bulk diffusion-controlled temperature by assuming Arrhenius behavior of higher temperature oxidation rate constant, rate constant, 10^{-3} to $3x10^{-4}$ vs 1/473-1/498; Relationship of test time to oxidation behavior, schematic diagram showing regions of grain boundary and bulk diffusion at 225 and 140°C; Schematic of TGA system; Plot of $[1-(1-C)^{1/3}]$ vs $t^{1/2}$; showing grain-boundary and bulk diffusion ranges at 200°C (from previous experimental work); Tables; TGA series 2 test matrix, delineates temperatures, durations, and moisture contents for proposed measurements; Moisture levels for testing, dew point vs ppm water vapor concentration; Characteristics of H. B. Robinson unit 2 and Turkey Point unit 3 spent fuel.

UNCERTAINTIES IN DATA: Not discussed.

DEFICIENCIES/LIMITATIONS IN DATABASE: The authors point out that low-temperature tests may indicate that oxidation takes place by a specific mechanism, such as grain-boundary diffusion or bulk diffusion but that this mechanism may break down after a long period of time. They hope to overcome this limitation by running tests for a 3-month period.

Draft Copy

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

(a) Relationship to Waste Package Performance Issues Already Identified

Related to issue 2.3.6 in the ISTP for the Nevada Nuclear Waste Storage Investigation (NNWSI) Project.

(b) New Licensing Issues

(c) General Comments

KEY WORDS: oxidation of spent fuel, oxidation rate, degradation of spent fuel, mechanism of oxidation.

GENERAL COMMENTS: The impetus behind these proposed measurements is that oxidation of UO_2 in defective fuel rods may lead to the rupture of the zircaloy cladding due to the volume expansion that occurs on oxidation. In addition, the leach rate of the higher oxides of uranium is apparently unknown but is thought to be higher than UO_2. I believe the authors may be overly confident concerning the accuracy of their data because scatter in rate data by factors of 10 for measurements in different laboratories is typically found in the literature. I would also suggest that less sophisticated but longer term experiments (years), which could involve multiple samples at the same time, might be more useful than those proposed. It is difficult to imagine how oxidation of UO_2 will take place in fuel rods until defects occur in the cladding. More definitive data on lifetime of the cladding and the leach rate of higher uranium oxides is important to this problem.

Draft Copy

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

DATA SOURCE:

(a) Organization Producing Data: Hanford Engineering Development Laboratory, P. O. Box 1070, Richland, WA 99352

(b) Author(s), Reference, Reference Availability: Wilson, C. N., " Test Plan for Series 3 NNWSI Spent Fuel Leaching/Dissolution Tests", HEDL-7577, April (1986)

DATE REVIEWED: August 23, 1986

TYPE OF DATA: Plan for leaching/dissolution study.

MATERIALS/COMPONENTS: Spent fuel from H. B. Robinson and Turkey Point PWR reactors. Tests include undefected, laser drilled defects and slit defects in the cladding, as well as bare fuel, in J-13 water.

TEST CONDITIONS: Four specimens at 85°C, one at 25°C from H. B. Robinson; one specimen from Turkey Point at 85°C, in 250 ml of J-13 water. Samples characterized with respect to cladding and position in fuel rod. Duration of test is to be 180 days with scheduled intermediate sampling. Two cycles are planned.

METHODS OF DATA COLLECTION/ANALYSIS: Planned schedule for volume and time of sample withdrawal. Filtered and unfiltered portions of solution will be obtained. Analysis includes pH, uranium, alpha and gamma spectrometry, ^{99}Tc, ^{90}Sr, ^{14}C, ^{126}Sn, ^{237}Np, and ^{129}I. Ion chromatography analysis for anions and inductively coupled emission spectrometry for cations will be performed on selected solution samples. Filters will be examined by SEM. Syringes will be stripped in 8 M HNO_3 and the solution analyzed for U and ^{241}Am by alpha spectrometry.

AMOUNT OF DATA: Four figures, 4 tables, and 2 appendixes. Figures: Two figures showing fuel rod segments superimposed on ^{137}Cs gamma scan; Sectioning diagram for Turkey Point fuel rod, series-3 test configuration for bare fuel tests; Defected or undefected cladding tests. Tables: Test specimens giving identification, specimen configuration and fuel type; Characteristics of H. B. Robinson and Turkey Point fuels; Sampling schedule for leaching study listing time, volume, sample type and analyses; Analysis Methods for radionuclides listing nuclide, method, detection limits in pCi/ml, ppb, and 10^{-5} inventory in pCi/ml; Appendix: Discussion of apparent misidentification of at least two pin segments of a fuel rod identified as ATM-101 and deficiency report; Detailed drawings of NWWSI series-3 spent-fuel leach test vessels.

UNCERTAINTIES IN DATA: Leach bath temperature will be uncertain by ± 2°C. Error limits for radionuclide concentrations are listed.

DEFICIENCIES/LIMITATIONS IN DATABASE: Not discussed.

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

(a) Relationship to Waste Package Performance Issues Already Identified

Related to issue 3.3 in the ISTP for the Nevada Nuclear Waste Storage Investigation (NNWSI) Project.

(b) New Licensing Issues

(c) General Comments

KEY WORDS: Leaching, J-13 water, spent fuel, H. B. Robinson fuel, Turkey Point fuel, NWWSI

GENERAL COMMENTS: This is a detailed report on plans for a third series of leach tests on spent PWR nuclear fuel. These tests differ from the series 2 tests in that the series 3 tests are to be run at 85°C in sealed type 304 (L?) stainless steel vessels. The tests are semi-static in that periodic solution samples will be taken for analysis and replaced with an equal volume of fresh J-13 water. I have several criticisms of this plan. Primarily, there is too much detail of how procedures are carried out and very little discussion of the purposes of the tests. The diagrams of the reaction vessels do not indicate what if anything is above the solution level. Later on it is stated that "Before removing the sample approximately 50 ml of air will be bubbled from deep in the vessel using a syringe. The bubbling has two purposes, to provide some limited convection mixing of the solution before sampling and to intoduce air into the test, which are otherwise sealed from the atmosphere." I think this is a poor procedure to introduce air into the test.

Keywo hecklist Tree 9/24/86

A. TECHNICAL DESCRIPTION OF REPORT

1. scope of work

2. applicability of data to licensing

3. model/methodology

4. measurement

B. ENVIRONMENT

1. testing sites

2. geographic location

3. aqueous environment

4. ionic or other chemical species present

5. solid materials environment

6. radiation environment

7. experimental/test conditions

C. MATERIALS TESTED

a. container and component materials

1. general material type

2. specific material designation

3. condition prior to test

4. test specimen specifications

b. radioactive waste (radionuclides and fuel materials)

1. source

2. radionuclides

c. environmental materials

1. aqueous

2. electrolytes present

3. solids

D. PROPERTIES AND FAILURE MODES STUDIES

1. mechanical and thermophysical properties

2. failure modes/phenomenon studied

A. TECHNICAL DESCRIPTION OF REPORT

1. scope of work
- __data analysis
- __design
- __experimental data
- __literature review
- __planned work
- __theory
- __other________________

2. applicability of data to licensing
- __key data
- __supporting data

3. model/methodology
- __Latin hypercube
- __Monte Carlo
- __PDF (probability distribution functions)
- __sampling
- __scoping test
- __other________________

4. measurement methods
- __adsorption
- __corrosion
- __electrochemical
- __linear-elastic fracture mechanics (LEFM)
- __microscopy
- __neutron diffraction
- __x-ray diffraction
- __sorption
- __spectroscopy
- __surface film
- __tensile testing
- __visual examination
- __weight change
- __other________________

B. ENVIRONMENT

1. testing sites
- __field site
- __simulated field site
- __laboratory

2. geographic location
- __Deaf Smith County
- __Hanford Reservation
- __Yucca Mountain
- __other________________

3. gaseous environment
- __air
- __other________________

3. aqueous environment
- __J-13 water
- __brine
- __brine (high ionic content)
- __brine (low ionic content)
- __deionized
- __basalt composition
- __granite composition
- __tuff composition
- __groundwater
- __simulated groundwater
- __deaerated distilled water
- __distilled water
- __aerated water
- __other____

4. ionic or other chemical species present
- __Cl
- __Cu
- __Fe
- __Ni
- __S
- __Keller's reagent
- __Carbon dioxide
- __other____

5. solid materials environment
- __basalt
- __granite
- __salt
- __tuff
- __bentonite
- __other____

6. radiation environment
- __alpha radiation field
- __gamma radiation field
- __other____

7. experimental/test conditions
- __ambient temperature
- __high temperature
- __ambient pressure
- __high pressure
- __hydrostatic head
- __lithostatic pressure
- __acidic solution (pH <7)
- __basic (alkaline) solution (pH >7)
- __neutral solution (pH = 7)
- __redox condition
- __static (no flow)
- __dynamic (flow rate given)
- __other____

C. MATERIALS TESTED

a. container and component materials

1. general material type

__brass
__bronze
__cast iron
__cladding
__copper base
__nickel base
__packing
__stainless steel
__steel
__carbon steel
__titanium base
__weld
__zircaloy
__zirconium base
__other ______________________________

2. specific material designation

__304 stainless steel
__304L stainless steel
__308L weld filler wire
__316L stainless steel
__317L stainless steel
__321 stainless steel
__347 stainless steel
__1020 carbon steel
__1025 carbon steel
__gray cast iron
__high nickel alloy 825
__nodular cast iron
__zircaloy-4
__other ______________________________

3. condition prior to test

__annealed
__annealed (austenitized and transformed)
__case hardened
__cast
__cold worked
__irradiated
__magnetized
__mill annealed
__sensitized
__sintered
__solution treated
__stress relieved
__textured
__welded
__wrought
__other ______________________________

4. test specimen specifications
[loading, geometry, and size designated by major category]
__standard tensile (round type)
__standard tensile (strip or strap type)
__slow strain rate
__standard compact
__modified compact
__size (circle): 1/4T, 3/8T, 1/2T, 3/4T, 1T, 2T, 3T,_____
__bolt or wedge loading
__tensile loading
__precracked
__prestressed (before exposure)
__prestressed (during exposure)
__other______________________________

b. radioactive waste (radionuclides and fuel materials)

1. source
__commercial high level waste (CHLW)
__defense high level waste (DHLW)
__spent fuel
__spent fuel (PWR reactor)
__spent fuel (BWR reactor)
__other______________________________

2. radionuclides
__Co60
__Np237
__Pu239
__other______________________________

c. environmental materials

1. aqueous
__J-13 water
__J-13 steam
__groundwater
__other______________________________

2. electrolytes present
__acetic
__chloride
__chloride (high ionic content)
__chloride (low ionic content)
__other______________________________

3. solids
__granite
__basalt
__salt
__tuff
__bentonite
__other______________________________

D. PROPERTIES AND FAILURE MODES STUDIED

1. mechanical and thermophysical properties

__bent beam tests
__creep strength
__density
__elongation
__heat capacity
__modulus of elasticity
__stress or strain
__tensile strength
__thermal conductivity
__thermal expansion
__yield strength
__other____

2. failure modes/phenomenon studied

__buckling
__corrosion (crevice)
__corrosion (general)
__corrosion (intergranular)
__corrosion (local)
__corrosion (microbial)
__corrosion (pitting)
__corrosion (stray current)
__corrosion (galvanic)
__corrosion (stress cracking) SCC
__creep
__creep buckling
__dealloying
__debonding
__deformation (elastic)
__deformation (plastic)
__degradation (spent fuel)
__devitrification (glass)
__fatigue (corrosion)
__fatigue (high cycle)
__fatigue (low cycle)
__fatigue (thermal)
__fracture (brittle)
__fretting
__hydration (glass)
__hydrogen attack
__hydrogen embrittlement
__leaching (radiation enhancement)
__leaching (spent fuel)
__matrix dissolution (glass)
__passivity
__poisoning (chemical)
__radiation effects
__rupture (ductile)
__rupture (stress)
__sensitization
__spalling
__thermal instability
__cracking (stress corrosion) SCC
__cracking (environmentally assisted)
__cracking
__other____

NRC/NMAB Preproposal--September 26, 1986

Integrity of Engineered Barriers for High-Level Radioactive Waste

SCOPE

Assess the status and requirements of the waste packages proposed to date for use in the disposal of high-level-waste (HLW), as a materials system that consists of a series of engineered barriers, which must have the durability required to meet the NRC requirements given in U.S. Public Law 10 CFR-60 on Disposal of High Level Radioactive Waste in Geological Repositories: 1. The waste package should contain all radionuclides for at least 300 to 1000 years after permanent closure. 2. The annual release rate for each radionuclide shall be less than one part in ten thousand of the amount present after 1000 years.

Emphasis would on the scientific and technological issues that must be adequately addressed in any long-range program for disposal of radwaste in host media of tuff, basalt, and salt. In addition to identifying and addressing the most critical issues, the study would attempt to define spefific information needs required to adequately verify the expected performance, and hence the acceptability, of a proposed system for disposal of radwaste. These included information on mechanictic understanding, modelling, laboratory and field testing, the statistical design of laboratory tests, and long-term monitoring. Although current DOE activities would be seriously considered, the study would not be specifically aimed at developing a critique of that work. Rather, it would focus on the nature of the scientific and technical issues in light of current U.S. efforts on this problem.

OBJECTIVES

1. To evaluate the technical issues associated with development of an engineered barrier system for long-term diaposal of HLW.

2. To recommend key activities to respond to requirements set forth in 10 CFR-60. This evaluation would encompass all aspects and components of present conceptuals designs for waste package systems, such as (1) metallic materials (shell, overpack, container, or cannister), (2) non-metallic materials, those used as a radwaste matrix (glass or other) and as packing or backfill, and (3) the environments (aqueous, gaseous and radiation) with which they will interact.

3. To evaluate the conditions under which long- or short-term monitoring (modeling or testing) studies are likely to be needed to adequately verify that the predicted performance has a high probability of success.

4. To determine key elements of a valid technical demonstration of the expected performance of the various elements of any proposed engineered barrier system.

5. To recommend R&D directions required to advance current DOE efforts.

BENEFITS

1. Provide an informed evaluation, from a panel of internationally recognized U.S. experts whose work is not dominated by government agencies, of technical issues relevant to performance of radwaste engineered barrier systems.

2. Provide a sound scientific basis for the development of a technical framework for prediction of expected performance for the key components (metallic and glass) of a HLW package system.

PROBABLE OUTPUTS

1. A set of guidelines for the R&D directions that would be needed to assess and demonstrate the expected performance of a waste package as a series of engineered barriers designed to meet the requirements of 10 CFR-60.

2. Assessments of the effectiveness of various technical inputs such as results of accelerated tests, the relevance of statistically designed experiments, and the conditions that would require long-term (50-year) monitoring of any of the elements of a waste package system. The relevance of deterministic and probalistic approaches to the problem would be considered:

A. Deterministic engineering approach--current and proposed test methodologies; real-time and accelerated tests; data needs; statistical design of experiments and extrapolation capabilities; likelihood of success based upon present mechanistic understanding.

B. Probabilistic modelling approach--current mechanistic understanding; validity of variables and inputs to models; likelihood of success.

SAMPLE TECHNICAL ISSUES TO BE EXAMINED

1. Time dependence of failure mechanisms, such as general corrosion, with particular emphasis on localized corrosion and any short-circuiting mechanisms that would lead to premature breach of an engineered barrier, radiation-induced changes in the environment (e.g. radiolysis of water) and in the barrier materials (embrittlement, phase changes).

2. Long-term microstructural stability, in components of a waste package, that might affect performance of the engineered barrier

system, e.g. phase transformations or embrittlement; transport problems (bulk or surface) within the waste package; effects of processing variables, e.g. weldments in metallic components; modelling capabilities; data needs and current data availability.

3. Monitoring of the HLW package system and its components in the early stages of the repository lifetime, i.e., during initial period of 50 years of retrievable storage: NDE and other diagnostics as appropriate.