

November 28, 1989



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

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

Re: Quarterly Progress Report for July, August, September 1989
(FIN-A-4171-9)

Dear Mr. Peterson:

Enclosed is the July, August, September 1989 quarterly progress report for the project "Evaluation and Compilation of DOE Waste Package Test Data" (FIN-A-4171-9). The financial information is reported separately.

Sincerely,

Charles G. Interrante
Charles G. Interrante
Program Manager
Corrosion Group
Metallurgy Division

Enclosures

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8911300324 891128
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A-4171 PDC

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A-417
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Quarterly Letter Report for July, August, September 1989

Published November 1989

(FIN-A-4171-9)

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 Quarter</u>	<u>Previous Quarter</u>
Number of citations	1122	1086
Number of completed reviews	85	78

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

1. ANL-88-14, "The Reaction of Glass During Gamma Irradiation in a Saturated Tuff Environment, Part 3: Long-Term Experiments at 1×10^4 rad/hr," February 1988.
2. Konynenburg, Van, R. A., Smith, C. F., Culham, H. W., and Otto, C. H., "Behavior of Carbon-14 in Waste Packages for Spent Fuel in a Repository in Tuff," UCRL-90855, November 1984.

3. PNL-5157, "Final Report of the Defense High-Level Waste Leaching Mechanisms Program," August 1984. A summary review that incorporates information given in each of seven previously conducted reviews.

As required under Task 1, a revised draft of the interpretive paper on "Mechanisms of Localized Aqueous Corrosion of Copper and its Alloys," was submitted on September 6, 1989.

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."

Yucca Mountain Project

- 2 Reports currently under review (Category 1).
- 28 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.
- 8 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 the quarter (July, August, September 1989) include Metadex, NTIS, DOE Energy, and Compendex Plus. Examples of the search conducted for each of these databases are in this report (see p. 26).

STATUS OF REVIEWS OF YUCCA MOUNTAIN PROJECT REPORTS

Yucca Mountain Project --

Category 1 -- Reports currently being reviewed

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

2. Beavers, J. A. and Thompson, N. G., "Container Corrosion in High Level Nuclear Waste Repositories," - First Semi-Annual Report/Year 2, September 1988 to February 1988.

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	<u>2/21/89</u>	<u> </u>	<u> </u>	<u> </u>
Beavers, 1988	<u>5/11/89</u>	<u>6/23/89</u>	<u>6/24/89</u>	<u> </u>

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

None this month.

Category 3 -- File and cross reference

None this month.

OTHER REPORTS ON VITRIFIED WASTE FORM --

Category 1 -- Reports currently being reviewed

None this month.

Category 2 -- Review as time permits

None this month.

Category 3 -- File and cross reference

None this month.

TASK 3 -- LABORATORY TESTING

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

July, August, September 1989:

Plots of various types were made using the data collected on specimen ST8. The types of plots made include plots of (1) stress-intensity factor (K) vs. elapsed time (s), (2) crack length (a) vs s, and compliance (C) vs s. In making these plots, various errors in the

computer codes used to represent constants and formulae were revealed and corrected:

1. Values of all pertinent constants, e.g. specimen width, notch width, crack-mouth-opening displacement (before and after wedge loading) were checked.

2. Values of all computed parameters, e.g. compliance (C), crack-extension rate (da/dt), and stress-intensity factor (K) were compared with values obtained from the results of independent calculations made by a different programmer who using a HP-9830 computer, which is programmed in a different language from that used for the computations in question. This other code had been checked by hand calculations in earlier studies, and so it was regarded as a reliable reference.

3. Corrections were made in the formulae used to convert the measured values of microvolts into units of crack length (a). This effort is a follow-up to the "problem solving" discussed as item 2 in the previous report on this work. In addition, it was discovered that values of the apparent crack length, as computed from these formulae, require a linear correction to be consistent with optical measurements of crack length, and this correction will be applied in the near future, as the math for doing the correction was completed but implementation into the computer code was not made during this period.

4. Values used for the intercept (b) and slope (m) were each off by an order of magnitude from the values determined in the calibration, which was made over a year ago.

5. In the formula used for computation of stress-intensity factor, K, an error in placement of a parenthesis made a large error in the calculated values of K.

6. A method was devised for computation of increments of strain energy released during discreet periods of crack extension for which the amount of acoustic energy detected is known. This comparison is the one needed to determine whether or not the relation between these two parameters is suitable for use in analyses of fracture mechanics test data. Implementation into the computer code is expected to be done during the next reporting period.

B. Title of Study: Effect of Resistivity and Transport on Corrosion of Waste Package Materials.

Principal Investigator: Edward Escalante

July, August, and September 1989:

Characterization of the after-exposure surface of the steel specimens verifies that, under aerated conditions, as the conductivity of the environment decreases, corrosion becomes more localized. This is particularly pertinent to the Yucca Mountain environment, since the site is expected to be well aerated with low conductivity. As has been

stated in the past, steel is used for these experiments because its corrosion rate is considerably greater than the waste container candidate materials, and will provide measurable weight loss data in a relatively short term experiment (90 days).

C. Title of Study: Pitting Corrosion of Steel Used for Nuclear Waste Storage.

Principal Investigator: Anna C. Fraker

Studies of literature and additional data and specimen analysis in preparation for writing a paper continue. The report that was submitted to NRC earlier will be put in the form of an NIST Internal Report.

D. Title of Study: Corrosion Behavior of Zircaloy Nuclear Fuel Cladding.

Principal Investigator: Anna C. Fraker

July, August, September 1989:

The purpose of this study is to provide information and data on the corrosion behavior of Zircaloy that can be used to determine the long-term durability of nuclear fuel cladding made of this material. Previous reports on this project have described laboratory tests consisting of data obtained from polarization measurements. These tests were conducted on bulk Zircaloy-2 and Zircaloy-4 and on cladding tubes of both of these alloys in 95°C, simulated J-13 well water (representative of water present at the waste storage site at Yucca Mountain, Nevada). In general, the tests have shown negligible corrosion rates, some variation from test to test regarding localized corrosion, and a tendency for crevice corrosion (localized) at the interface of the masking material and the Zircaloy. Approximately thirty tests have been conducted, and some tests have extended over longer time periods.

The work of this period has been directed toward publishing the literature review that was done for this study and preparing a report on the laboratory tests conducted to date. The literature review was published in August as NISTIR-4114, and a draft of the laboratory was prepared. A paper is in preparation for the proceedings of the Materials Research Society Meeting in November, 1989 where a poster on the laboratory work will be presented. There will be some additional analysis of the data and replotting of the data, and the draft report will be put in final form.

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

DATA SOURCE

(a) Organization Producing Data

Argonne National Laboratory, Argonne, Illinois

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

Abrajano, T. A., Bates, J. K., Gerding, T. J., and Ebert, W. L.
"The Reaction of Glass During Gamma Irradiation in a Tuff Environment"
ANL-88-14, February 1988

DATE: 9/22/89

CONTENTS

This 124-page report consists of an abstract, an executive summary, an introduction, a summary and conclusions, an acknowledgement, 30 figures, 12 tables, and the following content:

CONTENT	NUMBER OF PAGES
Experimental	10
Results	48
Discussion	37
Appendix I, II, III, IV	10
References (35)	4

PURPOSE

The experiments were performed as part of an effort by the Nevada Nuclear Waste Storage Investigations Project to assess the importance of radiation effects on repository performance and waste glass corrosion. The gamma radiation field used in this work was $1.0 \pm 0.2 \times 10^4$ rd/h.

"Savannah River Laboratory 165 type glass was leached with equilibrated J-13 groundwater at 90°C for times up to 182 days." "To understand the effect of radiation on groundwater chemistry and waste form durability, NNWSI is performing an extensive set of radiation experiments as a function of dose rate (2×10^5 , 1×10^4 , 1×10^3 , and 0 rd/h." "This report presents the results of the tests done at 1×10^4 rd/h and discusses the relevance of these tests to repository conditions."

KEY WORDS

Experimental data, leaching, microscopy (ligh, electon, SEM, etc.), spectroscopy, surface film, laboratory, J-13 water steam, J-13 water, Cu, Fe, tuff, alpha radiation field, gamma radiation field, ambient pressure, ambient temperature, basic (alkaline) solution (ph >7), static (no flow), stainless steel, 304L stainless steel, welded, glass (defense waste reference glass), defense high-level waste (DHLW), ^{137}Cs , Sr, ^{237}Np , ^{239}Pu , ^{241}Am , J-13 steam, J-13 water, tuff, leaching (radiation enhancement).

TEST CONDITIONS

Materials:

- glass: SRL U (SRL 165 black frit to which uranium, cesium, and strontium had been added).
SRL A (SRL U glass to which ^{237}Np , ^{239}Pu , and ^{241}Am had been added.)
- vessel: Type 304L stainless steel.
- rock: (1) tuff from USW-G1, 1235.1-ft. and 1232.2-ft. levels.
(2) tuff supplied by NNWSI (UE-25h #1, 173.0-ft to 173.6-ft. levels).
(3) tuff from USW-62, 1235.1~1232.2 ft.-level.

Specimen Preparation:

- glass: One of the two disks used in each test had one side ground to 600 grit. Therefore, three different types of glass surfaces were present in each test; both faces of one disk and the bottom side of the other disk are as cut, the top side of the other disk is ground to 600 grit, and the sides of the disks are as cored. The glass surface area to solution volume ratio was nominally set at 0.3 cm^{-1} .
- rock: polished tuff core wafer.

Environment: EJ-13, EJ-13 plus tuff, EJ-13 vapor and EJ-13 vapor plus tuff at a nominal temperature of 90°C and at a gamma-dose rate of $1.0 \times 10^4 \text{ rd/h}$ (total dose up to $4.4 \times 10^7 \text{ rads}$) for 14 to 182 days. The vessels have a capacity of 21.4 cm^3 and were sealed a compression fitting and a silicon rubber gasket.

UNCERTAINTIES

- gamma dose rate : $\pm 0.2 \times 10^4 \text{ rd/h}$.
- temperature : $\pm 0.5^{\circ}\text{C}$.
- selected activities in SRL A glasses : SD (standard deviation) from 0.031 to 27.77 (% SD from 8.98% to 26%).

- "1 σ " (not specified, we assume this to be one of SD definition from the probabilities distribution function).
- "Variation of Np and Am between glass disks were within 20 and 10 percent of their mean, respectively. The experimentally determined analytical precision of the 2-counting facility was less than 10%."
- "Water loss for experiments performed for less than 182 days did not exceed 0.10 g, whereas the maximum water loss noted for the 182-day experiment was 0.25 g."
- "A 2-KeV neon ion beam was used for SIMS. The sampled area is of the order of 1 mm²."
- Analytical accuracy and precision from repeated analyses: "3~10% (cation), 5% (anion), 5% (uranium), 10% (dissolved gases), and 0.02 unit (pH)."
- Experimental accuracy and precision: "Because of statistical limitations, several sources of error are identified."
 - "compositional inhomogeneity of glass and tuff"
 - "the contamination of the solution with Si from the gasket material"
 - "the contamination of the solution after termination of the run. A speculative scenario is that refluxing gases vaporized during the 90°C acidification could have condensed on some portions of the Si gasket, reacted with the gasket and dripped back to the solution."
- "opted to present results of duplicate experiments to give the reader a better feel for the experimental uncertainties, rather than the overall precision (i.e., experimental and analytical) to be $\pm 7\%$ for Na, $\pm 4\%$ for B, $\pm 5\%$ for all anions, $\pm 15\%$ for the actinides and $\pm 10\%$ for all other cations, which were estimated in the previous report."
- "the overall variations in the concentrations of anions (F⁻, Cl⁻, NO₂⁻, NO₃⁻ and SO₄⁻²) are only between 10 to 20 percent (1 σ) of their mean values for the EJ-13 only experiments." "Greater variations were observed in the concentrations of NO₂⁻ and NO₃⁻ for the EJ-13+ tuff experiments, but even these are within 30% of their mean abundances in all the tests."
- There are cross-hatched regions in figures showing the uncertainties: time vs pH, NO₃⁻/NO₂⁻ vs pH.
- There was an "analytical artifact in Nuclear Resonance Profiles arising from the presence of irregular morphology on the reacted glass surfaces."
- In SIMS analyses, "sputtering depths measured using both methods 2 and 3 agree within 10%" (method 2 : by surface profilometry (Dektac) of glasses sputtered for known periods of time; method 3 : by sputtering layers of known thickness as measured by SEM).

DEFICIENCIES/LIMITATIONS IN DATABASE

- "Some comments on the relevance of the results to actual repository scenarios are" "not intended to be exhaustive, as there are certain experimental parameters that have been of standard use in NNWSI repository-oriented tests which are more elaborately discussed elsewhere, e.g., NNWSI Site Characterization Plan." Such parameters are temperature, glass composition, EJ-13 solution, radiation levels and dose rate, H₂O liquid volume, various configuration of glass

associated with tuff, homogeneity of host rocks, pressures of gas phase and water steam, the ratio of gas volume to liquid volume, pH drift, availability of buffering species such as HCO_3^- , and the ratio of glass surface area to solution volume. "A thorough analysis of even some of the experimental parameters adapted in the present program may be premature because more experiments are in progress in our laboratory to further understand the interactions which occur in the reaction vessels and to refine the experimental methodologies." "Several experiments designed to further understand NNWSI repository conditions after waste package emplacement are actively being pursued in other laboratories, foremost of which is LLNL."

- "The gamma dose rate used in the present work is higher than the dose rates appropriate for a SRL glass canister breached sometime during the containment period." "Accelerating the effect of γ -radiolysis by increasing the dose rate without accelerating other interactions (e.g., waste glass corrosion) can unduly influence the glass corrosion mechanism and elemental release pattern, e.g., actinides."
- The Si concentration in solution is going to be higher in systems with high SA/V ratio compared with systems with low SA/V ratio." "Practically, there is no evidence that the leachate is independent of SA/V ratio."
- "(SA/V . t) scaling can be used to predict long-term release rates only if fractional leach rates are a function of solution concentration only."

CONCLUSIONS OF AUTHOR

The measured pH's in both the blank and actual runs followed a trend of decreasing values to 56 or 91 days and increasing values after 91 days. We attribute this variation to the interplay between acidification by nitric acid production (radiolysis of N_2) and basification by $\text{CO}_2(\text{g})$ loss from solution. In the presence of SRL glass, exchange reactions between the hydronium ions or protons from solution and mobile cations (e.g., Li and Na) from the glass further basified the solution. However, the overall effect of glass corrosion on solution pH in the present experiments is not dramatic, and a narrow pH range of 6.7 to 7.5 prevailed for the whole duration of the experiments.

The only significant anion variation observed in the leaching experiments is the systematically lower $\text{NO}_3^-/\text{NO}_2^-$ ratios from EJ-13 only to EJ-13 + tuff to EJ-13 + tuff + glass experiments. This observation could be attributed to the catalytic effect of reduced species on silicate surfaces upon radiolytic reactions that ultimately lead to nitrate breakdown [VON KONYNENBURG].

The general normalized release pattern, $Li \geq Na \geq B \geq Si$, was exhibited by the solution data. A single parabolic function could be fitted to Li and Na (to 91 days) regardless of the presence or absence of tuff. The release rate of B and Si also slowed down with time.

The nuclear resonance profiles for H and the SIMS profiles for Li, Na, B, Al, Ca, Mg, Mn, and Fe indicated two regions in the reacted layer. The region closer to the unreacted glass, referred to as the transition zone, exhibits well-defined S-shaped profiles for H, Li, Na, and B indicative of concentration-dependent diffusion coefficients for these elements during interdiffusion. The region away from the glass, referred to as the gel layer, exhibits relatively flat profiles for these elements.

Speculations on the mechanisms of release of individual glass species were based on the above information. Lithium, and Na are released from the glass via ion-exchange (with proton or hydronium) across the transition zone while B diffuses from the transition zone after pathways for diffusion are established. The transition zone behaves as a transport barrier to these species. On the other hand, the dominant mode of Si release is etching of the gel layer. Some Si may be released from within the gel layer as speculated by [GRAMBOW], but this amount is probably a minor contribution to the overall Si-release. Transport of H, Li, Na, and B across the gel layer is rapid compared with diffusion across the transition zone.

The depth profiles showed that the transition zone grew at a decreasing rate with time, a finding which agrees with the leachate data. Approximate calculations of the rates of gelation and etching suggest that both rates also decrease with time. We attribute these decreased rates with time to saturation effects.

The release of other major glass species were considered within the framework of the mechanism discussed above. The variation of the normalized solution concentrations of Mg and Sr with time is consistent with their release via etching. A similar release mechanism may explain the release pattern of Ca, but some influence of secondary phase precipitation is also indicated. The SIMS profile for Ca and Fe shows an enrichment in the gel layer. Early precipitation of Al phase(s) is indicated by the solution data.

The actinide elements, Pu and Am showed total normalized release patterns that are approximately linear with time to at least 91 days. We tentatively ascribe this release pattern to a combination of matrix breakdown and reconstitution in the gel layer, although preliminary ion microprobe profiles suggest that Pu and Np behavior may be more complex. The significant correlation between pH with Pu, Np, and Am concentrations in unfiltered nonacidified samples suggests that solubility is the key parameter controlling their concentration levels in solution.

The presence of tuff did not appear to affect the mechanism of release of the major glass species. Because tuff also dissolved in solution, a faster approach to saturation of secondary products was noted where tuff was present. The present data strongly indicate that some sorption of actinides to the tuff surface has occurred. Larger total actinide release was noted in

the case of Pu and Am in tuff-containing experiments. This observation is attributed to the lower integrated pH values attained in the tuff-containing experiments in the present program.

Some preliminary comments were made on the applicability and limitations of the present results to predicting the nature of actual interactions in the NNWSI repository. Specific attention was given to test configuration, test component materials, R, γ -dose rate, and SA/V ratio. The present results, together with those of the previous [BATES-1] and ongoing experiments, will be evaluated in the future to provide more definitive assessment of the repository relevance of these experiments.

RELATED REPORTS

1. Bates, J. K., Fischer, D. F., and Gerding, T. J., "The Reaction of Glass During Gamma Irradiation in a Saturated Tuff Environment, Part I: SRL 165," ANL-85-62 (1986).
2. Bates, J. K., Ebert, W. L., Abrajano, T. A., Mazer, J. M., and Gerding, T. J., "NNWSI Waste Form Testing at Argonne National Laboratory," Semiannual Reports for July-December 1985 until Present, UCRL-1580, in press.
3. Bates, J. K. and Gerding, T. J., "One Year Results of the NNWSI Unsaturated Test Procedure: SRL 165 Glass Application," ANL-85-41 (1986).

COMMENTS OF REVIEWER

1. This report presents unique data of radioactive glasses leaching which cannot be obtained easily in many other laboratories. Therefore, the data presented here are very valuable for HLW licensing applications.
2. The authors provide the readers with their elaborate analysis of the experimental errors. They could easily identify the analytical errors. However, the experimental errors have been concluded to be very difficult to be assessed, leaving them for a future task. The reviewers recommend an adoption of statistical theories in such difficult circumstances. An example of those theories is the extreme value theory.
3. Much longer tests, or valid accelerated tests, are recommended to be performed. The main reason for this recommendation is the incongruency of leach rates accompanied by pH drift or by the diffusional movement of the altered surface layer, which makes it difficult to predict the long-term performance of waste glasses. Without the long-term prediction, it is likely to make serious mistakes in the formulation of waste glasses.
4. The authors point out that colloid formation has not been analyzed at this point. Considering the portion of the dissolved species in acidified solutions is substantial, this topic should be explored further. Many analytical tools do not detect un-ionized species, and the dispersion of such particles to biospheres will enhance the possibilities of violating the

government rules. Likewise, the sorpted elements on host rocks or stainless steel should be reanalyzed in this regard too.

5. The purpose of surface analysis was very clearly stated in this report (p. 74), which is very unusual among so many works in the surface analysis of waste glasses. The reviewers further recommend that attempts be made to tailor glass compositions minimizing the leach rates based on these surface analyses.

6. While the generic leaching behavior of generic elements has been analyzed very extensively, their correlation with the analyses to radioactive elements is quite limited. The recommendation is that this be explored further for the licensing applications.

7. In many instances, the authors use solubility limits in analyzing leach data. We should be cautious about using the solubility limits because they are of thermodynamic equilibrium concepts. When precipitates are growing, those solution concentrations can be dynamically balanced ones. For instance, on page 91, actinides release rates was attributed to the pH dependence of the actinides solubilities. This can be explained by the pH dependence of reaction constants when viewed as the kinetic processes.

8. More elaborate efforts in cross-comparison of various data are necessary in interpreting data using multiple variables, rather than simple speculations using data from one variable. An example is the long-term decrease of leach rates. This behavior should be explored further by analyzing their dependence on other variables such as colloid formation, pH change, Eh change or altogether.

9. If the authors like to interpret their data in terms of solubilities, they are recommended to estimate the redox conditions more quantitatively. The importance of Eh condition in leaching has been well recognized in many related papers in glass leaching.

10. There have been minor errors found by the reviewers, which, in some cases, could be important in the future.

- On p. 68, the corrosion of stainless steel can be a source of proton generation too by the oxide formation with water molecules.
- On p. 70, the low O_2 and N_2 concentrations have been attributed to air leakage. If they are in equilibrium with air, there would be no leakage.
- The fitting experimental data to $t^{1/2}$ or t should be reconsidered. In many such formulations in chemical kinetics, there are constant terms involved, which change the curve behavior significantly.

- On p. 78, there were assumptions of using zeroth- or first-order kinetics. Some examples of such order kinetics from generic kinetics seems to be necessary to justify these assumptions.
- On p. 97, NNWSI site will not be saturated. To justify the present work in aqueous environments, this scenario should be re-reviewed, or at least a worst case scenario should be presented.
- On p. 11 and p. 33, there appear to be typographic errors: total dose 5×10^8 rad not rd/h, and $V_f^{-1}(S_A)^{-1}$ should be $V_f(S_A)^{-1}$ or $V_f^{-1}(S_A)$.

APPLICABILITY OF DATA TO LICENSING

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

- (a) Relationship to Waste Package Performance Issues Already Identified
 - 2.3.2 regarding the solubility of the waste form under potential repository conditions
 - 2.3.2.1.2 regarding rates of dissolution
- (b) New Licensing Issues
 - Effects of gamma radiation on waste form dissolution.
- (c) Comments related to Licensing

AUTHOR'S ABSTRACT

Savannah River Laboratory 165 type glass was leached with equilibrated J-13 groundwater at 90°C for times up to 182 days. These experiments were performed as part of an effort by the Nevada Nuclear Waste Storage Investigations Project to assess the importance of radiation effects on repository performance and waste glass corrosion. The gamma radiation field used in this work was $1.0 \pm 0.2 \times 10^4$ rd/h.

Glass dissolution is notably incongruent throughout the entire experimental periods and normalized releases follow the sequence $Li \geq Na \geq B \approx U \geq Si$. The normalized leach rates of these elements, as well as the measured growth rates of the reaction layers, decreased with time. The only significant variation observed in the abundance of anions is the systematic decrease in NO_3^-/NO_2^- ratio from the starting EJ-13 groundwater to the EJ-13 blank experiments to the tuff- and glass-containing experiments. A leaching model that is consistent with the observed solution data and depth profiles is presented. The applicability and limitation of the present results in predicting the actual interactions that may occur in the NNWSI repository are discussed.

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

DATA SOURCE

(a) Organization Producing Data

Lawrence Livermore National Laboratory, Livermore, CA. For the NNWSI Project, U.S. DOE.

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

Van Konynenburg, R. A., Smith, C. F., Culham, H. W., and Otto, C. H., Jr., "Behavior of Carbon-14 in Waste Packages for Spent Fuel in a Repository in Tuff," UCRL-90855, November 1984.

DATE REVIEWED: 8/7/89

PURPOSE

The authors state in the introduction that "Our laboratory was asked to perform mass spectrometric and radiometric analyses of gas samples taken from spent fuel storage canisters in order to determine whether any fuel rods had ruptured." "This paper discusses the initial results obtained and the ramifications for the storage of spent fuel in a potential repository in Tuff."

KEY WORDS

Data analysis, experimental data, gaseous radionuclides escape, spectroscopy, laboratory, air, N₂, O₂, Ar, He, CO₂, CO, N₂O, ¹³CO₂, ⁸⁵Kr, ¹⁴C, ¹⁴CO₂, ¹⁴C (not specified), ¹³C, high temperature, stainless steel, spent-fuel-rod with splits, 0.21 x 0.21 x 4.10 m, 0.645 Mg, spent fuel (PWR), gas escape, thermal instability.

CONTENTS

Text: 8 pages, 2 tables, and 22 references.

AMOUNT OF DATA

Table 1. Spectrometric and Radiometric Analyses.

Table 2. Calculated Gas Volumes and Releases.

TEST CONDITIONS

1. Materials: "15x15 PWR spent fuel assembly." "It consists of UO₂ fuel pellets inside Zircaloy-4 tubes, held in place by type 304 stainless steel orifice plates and Inconel 718 spacer grids." "After removal from the reactor the assembly was stored in a water pool for less than 2 years." Then, the assembly "was sealed inside a helium-filled canister, and was used in a silo-type fuel storage for about 4 years."
2. Specimen Preparation: "The assembly was removed from its canister and inspected, and two fuel rods were removed. The assembly was then placed in the Fuel Temperature Test (FTT) stand."
3. Environments: "The canister of FTT was filled with air and the temperature was raised to 275°C and was programmed to decrease slowly with time." The dose rate was 10⁴ rd/h.

UNCERTAINTIES IN DATA

"The CO₂ analyses for samples #26 and #34 are somewhat suspect because of air leaks into the samples during storage prior to analyses."

DEFICIENCIES/LIMITATIONS IN DATABASE

"A comparison of this value with the 10CFR60 release limit of 10⁻⁵ per year from the 'engineered system' shows that it will be important to carefully define the 'engineered system' and to take account of the time distribution of lifetimes of the canisters to meet this requirement."

CONCLUSIONS OF AUTHOR

1. About 1.5 mCi of ¹⁴C was rapidly oxidized and released as gas from the external surface of a PWR spent fuel assembly stored in air at 275°C and producing a radiation field of about 10⁴ rd/h.
2. An additional 0.3 mCi was released as gas after one of the 204 fuel rods breached, but it is likely that most of this also came from the external surface of the assembly. Radiometric ¹⁴C analysis should be performed directly on fuel rod gas to check this conclusion.
3. The initial gaseous release would not exceed the limit of 40CFR191. Further study is needed on slower releases of ¹⁴C.
4. This gaseous release is large enough that a broad definition of the 'engineered system' in 10CFR60 may be necessary, and account will have to be taken of the time distribution of lifetimes of the canisters, in order to meet the requirement of this regulation."

COMMENTS OF REVIEWER

1. There is a limited amount of experimental data on the release of the ^{14}C from spent fuel; therefore, the authors question the validity of conclusions derived from their own calculated results.
2. In discussing the present results in light of 10CFR60 (or 40CFR191), the authors do not consider the container of the waste package in the repositories as a possible source of ^{14}C . Only the fuel itself was considered. However, most of the metal from which a container would be fabricated, will contain nitrogen as an impurity element, and under the long service life of a repository, the container of the waste package is likely to have its nitrogen transformed into ^{14}C . Thus, the release of ^{14}C from the container is very likely to take place even before the failure of the container. This will occur whenever corrosion products are released to the environment. Oxygen and water are expected to be present around the package during the first 300-1000 years of its underground emplacement. Therefore, the reviewers suggest that this hypothesis should have been analyzed, in order to make the analysis more complete.
3. In agreement with these authors, the reviewers recommend that radiometric analysis of ^{14}C should be conducted directly on fuel-rod gases. However, the ^{14}C from the container materials must be considered in making estimates of the total release of ^{14}C from a waste package.
4. The authors' conclusion number 4 implies that truly significant releases may arise from the ^{14}C found in the spent fuel contained within waste packages. We would question the "significance" of these releases. A comparison would have to be made, for example, to determine if this waste-package-related release of this non-fission product would commonly be expected to be released from the smokestacks at nuclear power facilities. The question of relative health risks for these two cases should be assessed before concluding that this release is significantly harmful and therefore subject to special release rate limits.

RELATED HLW REPORTS

1. R. A. Van Konynenburg, C. F. Smith, H. W. Culham, H. D. Smith, "Carbon-14 in Waste Packages for Spent Fuel in a Tuff Repository," Lawrence Livermore National Laboratory, UCRL-94708, October 1986.

APPLICABILITY OF DATA TO LICENSING

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

(a) Relationship to Waste Package Performance Issues Already Identified

- 2.3 When, how, and at what rate will radionuclides be released from the waste form?

(b) New Licensing Issues

(c) Comments Related to Licensing

AUTHOR'S ABSTRACT

Analysis of gas from a heated air-filled canister containing a spent fuel assembly before and after rupture of a fuel rod shows that about 1.5 mCi of ^{14}C from the external surface of the assembly was rapidly oxidized and released as $^{14}\text{CO}_2$ in excess oxygen at 2753°C and 104 rad/hr. After rupture, an additional 0.3 mCi was released, probably also from the external surface. The total ^{14}C inventory in the entire 15x15 rod assembly including structural hardware is estimated to be 690 mCi. These measurements indicate that account will have to be taken of the time distribution of lifetimes of the canisters, and a broad definition of the "engineered system" may be necessary, in order to meet 10CFR60 requirements with spent fuel in a repository in tuff.

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

DATA SOURCE

(a) Organization Producing Report

Pacific Northwest Laboratory, Richland, Washington 99352.

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

Mendel, J., Compiler, "Final Report of the Defense High Level Waste Leaching Mechanisms Program," PNL-5157, August 1984. Available from NTIS.

DATE REVIEWED: 7/27/89

PURPOSE

Purpose of Program:

"The objective of the leaching mechanisms program was to determine the dominant leaching mechanisms for defense waste glass and to evaluate the effects of some major potential environmental parameters upon the leaching mechanisms."

Purpose of Review:

The NIST has reviewed the "Final Report of the Defense High-Level Waste Leaching Mechanisms Program", PNL-5157, which is a 1984 literature review on glass leaching behavior of nuclear-waste glasses that have been proposed for use in the vitrification of high level waste (HLW). The report consists of seven chapters and each chapter was prepared by one or more of seven reviewers. The PNL-5157 report was made under the direction of the Materials Characterization Center (MCC). Chapter topics are given below:

- (1) Mechanism of Defense Waste Glass Corrosion
- (2) Surface Layer in Leached Borosilicate Glass HLW Forms
- (3) Environmental Interactions
- (4) Dissolution of Specific Radionuclides
- (5) Radiation Effects
- (6) Phenomenological Models of Nuclear Waste Glass Leaching
- (7) A Computer Code PROTOCOL for a Numerical Simulation of Glass Dissolution

For each chapter, the NIST has conducted a review to identify the following categories of information: (1) the type of literature, (2) materials used, (3) test conditions, (4) analysis methods, (5) uncertainties in data, (6) deficiencies/limitations in data base, and (7) conclusions. A summary of

comments given by NIST reviewers of these chapters, with appropriate recommendations, is presented below in the order indicated above for the seven sections of the NIST reviews. For more details on any one chapter, see the NIST review of that chapter. Since the publication of this report (PNL-5157) in 1984; MCC has obtained additional results. The reader is referred to their reports and to reports from SRI, WVDP, Vitreous State Laboratory at the Catholic University of America.

KEY WORDS

Literature review, leaching, solubility, PROTOCOL, microscopy (light, electron, SEM, etc.), spectroscopy, surface film, weight change, x-ray diffraction, fluorometric uranium, laboratory, Yucca Mountain, deionized, simulated groundwater, HCl, NH₄OH, Stripa, Cl⁻, F⁻, CO₃²⁻, HCO₃⁻, SO₄²⁻, NO₃⁻, OH⁻, NH₄⁺, tuff, alpha radiation field, gamma radiation field, beta, acidic solution (pH <7), ambient pressure, ambient temperature, basic (alkaline) solution (pH >7), dynamic (flow rate given) 0.1 to 10 ml/h, high pressure, high temperature, neutral solution (pH = 7), static (no flow), glass (defense waste reference glass), defense high-level waste (DHLW), Np237, Pu239, U, groundwater, DI water, leaching (radiation enhancement), matrix dissolution (glass).

CONTENTS

This report consists of an introduction, a summary and conclusions, and the following contents:

Contents	Number of Pages	Number of Figures	Number of Tables	Number of References
Chapter 1	63	28	21	90
Chapter 2	37	34	7	30
Chapter 3	43	12	13	98
Chapter 4	49	52	7	41
Chapter 5	18	9	8	48
Chapter 6	16	8	0	44
Chapter 7	35	35	8	15
Appendix A: Reference Materials	2	0	2	2
Appendix B: Use of PROTOCOL	7	0	1	1
Appendix C: Leaching Mechanisms in Polyphase Ceramic High-Level Nuclear Waste Forms	28	22	11	11

AMOUNT OF DATA

The report "PNL-5157" comprehensively covers experimental efforts, modeling studies and a review of the literature related to leaching of nuclear waste glass, which is glass loaded with radioactive waste.

Emphases are on the simulated nuclear-waste glasses in the repository environments, including glasses containing ^{239}Pu , ^{237}Np or U. Control studies performed in commercial glasses were also reviewed. Finally, in relation to safe performance of glass over geological times, the nature of ancient and natural glasses was evaluated.

TEST CONDITIONS

This extensive review covers various glass shapes that had been tested. The forms of the glasses are either monolith or powders. The ratio of surface area to glass leachate volume, as covered in these tests, ranges from 10^{-5} to 2300-m^{-1} . For the surface of the monolith, preparation of the surface is very important. The surfaces were normally polished with Si-C papers, but inadequate documentation on surface preparation given in this document seems to have left this important variable at times uncontrolled.

This 1984 publication (PNL-5157) did not refer to the definitive study by Kingston et al. (1984)¹, but the MCC has revised MCC-1 and the revision includes changes that can be attributed to this important contribution. That revision to MCC-1 was made after the 1984 publication of PNL-5157.

The flow conditions for leachates are either static or dynamic conditions. In dynamic tests, continuous flow of the leachate was used to simulation high flow rates of groundwater. In another test, pulsed replenishment of the leachate was used to simulate the low flow rates expected to be present in some candidate repositories. Normally, the flow rates tested ranged from 0.1 to 10 ml/h. The testing temperatures ranged from 40 to 93°C, at ambient pressures, for times of up to 12 months.

The pH and Eh conditions of the leachate are respectively from one to 11 and from -10 to -80 mv. The leachates are deionized (DI) water, hydrochloric acid (HCl), ammonium hydroxide (NH_4OH) or simulated groundwater, including Hanford, Tuff and Stripa constituents. Leachates are either air-saturated, nitrogen-saturated or silicated. Environmental effects due to waste package components (other than glass) were also considered. These components include iron, stainless steels, and other secondary components such as backfill materials, each of which might be present in the leachates. In the case of radiation tests, alpha, beta and gamma were all considered for the structural damage effects as well as for radiolytic effects. Besides using these experimental conditions, one- and/or two-dimensional simplification was used in the modeling as a geometric description of the leaching system.

The leach rates are normally measured as the total mass loss per unit area per unit time (mass/A/t). The total mass loss was determined by atomic

¹ H.M. Kingston, D.J. Cronin, and M.S. Epstein, "Investigation of a Precise Static Leach Test for the Testing of Simulated Nuclear Waste Materials", Nuclear and Chemical Waste Management, v5, p3-15, 1984.

absorption (AA) or inductive coupled plasma spectroscopy (ICP); in addition, many other techniques were used for surface analyses, including SEM, SIMS, XPS, SIPS, AES, SAM and XRD. For radiation studies, actinide doping methods or external heavy ion and/or neutron irradiation were utilized. In the modeling efforts, solute diffusion was extensively used, and modifications were made in conventional leach models by taking into account the leachate saturation and the subsequent precipitates.

UNCERTAINTIES IN DATA

The presented data were examined carefully for uncertainties including error bars (or tolerance limit of each data set), and the drifting of experimental conditions such as pH was noted. To estimate reproducibilities, the averages of at least two determinations were given. Normally, in PNL-5157, this reliability information is not very extensive, especially in modeling, as for example, for the reaction rate constants used in the model: As a consequence of using uncertain reaction-rate constants, the exponent of the power-law leach-kinetics varied from -0.2 to 0.6, which may be unacceptable for most purposes.

DEFICIENCIES/LIMITATIONS IN DATABASE

Each chapter rarely addresses the deficiencies/limitations in the data base (used for PNL-5157) in relation to any licensing application issues. These issues would emphasize the leaching behavior of waste glass over extended (geological) times. Such an example is the determination of thermodynamic equilibrium for the solubility consideration: The state of seemingly thermodynamic equilibrium can be simply a balance state between dissolution and precipitation of the waste glass under repository conditions. See below for more details, which are given in the Conclusions section of this review.

CONCLUSIONS

Refer to Conclusions for each chapter and to comments of this review.

COMMENTS:

Summary and Recommendations:

The following is a summary of views of the NIST review groups for 7 chapters of PNL-5157:

The leach rates are extremely low regardless of the glass compositions at pH of 7 to 9, and temperatures from 40 to 93°C and low flow rates. A comprehensive coherent picture of glass leaching is not now available. Unless the repository design must handle the worst case scenarios, it has been indicated, by both the PNL authors and the NIST reviewers, that it may not be necessary to have a coherent picture for glass leaching, i.e. a very thorough mechanistic understanding or an extensive quantification of the effects of glass composition on glass leaching.

It is assumed that for Yucca Mountain, the leachate will become saturated in each of the radionuclides. The levels of this saturation concentration for the various radionuclides tends to depend on repository (environmental) conditions, i.e. solution chemistry, and this means that the chemistry of the waste forms (the glass in PNL-5157) will play a comparatively minor role. Thus, leach rates will depend directly on flow rates at sufficiently low flow rates such as those that are expected for Yucca Mountain. As flow rates increase, the leach rates become dependent on the kinetics of dissolution, but the high amounts of water associated with this behavior are not expected for Yucca Mountain. These leachate effects are relatively well documented.

Other effects of solution chemistry include those associated with iron and/or backfill-materials. These materials may be present in various components of a repository. They tend to increase the leach rates by forming precipitates. Species dissolved in the groundwater tend to decelerate the leach rates, because their presence brings the leachates closer to their solubility limits.

While much of the above discussion assumes conditions in which saturation concentrations have not been met, complications do arise from saturated leachates. In addition, a list of other complications to the development of a mechanistic understanding of the leaching process would include the following:

- (1) As leaching progresses, the pH changes.
- (2) The presence of iron ions will be to alter the solution Eh and/or oxic conditions. This will affect the leach rates. The Eh conditions were found to be important for the long-lived Tc and transuranic species, because the various valence states of these species have different solubilities.
- (3) The surface layer becomes altered and plays an important role in the leaching process.
- (4) The surface layer grows in the amorphous state with pits and crystalline deposits and its formation and growth are most greatly affected by solution chemistry as discussed above, i.e. it forms from products coming from solution rather than those coming from the glass itself.
- (5) Colloidal-sized particles form in the leachates; this is particularly important for Tc and transuranic species.

The role of the altered layer has not been studied and understood adequately well to make it possible to prevent or greatly retard leaching by careful control of glass chemistry. We note that this layer has the potential for doing for glass in some environments what a passive film does on the surface of a metal alloy like stainless steel in oxidizing environments. It would seem that leach rates could be greatly decreased by use of tailored glass compositions, which promote (for specific environments) the formation of an alteration layer that prevents further leaching.

The level of the radiation field expected to be present in a waste package sufficient to produce structural damage in glass as well as in the host rocks, and whether the state of thermodynamic equilibrium is obtained for not, is an important question for the prediction of long-term glass leaching behavior. When precipitates go, either on glass surfaces or in solutions, the solutions are not necessarily in a state of thermodynamic equilibrium. They can be simply in a balanced state between dissolution and precipitation of the available solids in the solutions. This is a dynamic state that can be terminated (at a limiting condition) in a state of thermodynamic equilibrium, e.g. at the solubility limit for a precipitate; at that limit it is presumed that the system is in equilibrium. Both again accelerate leach rates.

The development of models and computer codes, as presented in PNL-5157 for glass leaching, includes the classical approach in a dilute solution, the interaction with the leachate concentration, solubility effects, and a model for dispersion to the biosphere. However, the efforts presented in this publication are successful only in forming a skeleton/foundation which needs further polishing. Of course, deeper understanding of detailed leaching phenomena will be required to do successful modeling.

Based on the above conclusions, the NIST reviewers recommend the following for future work:

1. There are sparse quantitative data on effects of glass composition and this makes it impossible to tailor the glass composition to provide low leach rates. Since many authors recognize that this tailoring may not be necessary due to the low flow rates under expected Yucca Mountain conditions, it is an urgent task to identify the worst case of repository conditions, especially the flow rates.
2. In the study of flow-rate effects, the practice of replenishment of leachates should be validated as a means to simulate a very slow flow rate. Also, the change in surface area of crushed glass should be considered in the determination of leach rates, as it is corroded away to the vanishing point.
3. The high pH at low flow rates needs to be studied as the glass decomposes readily under this condition.
4. Other self-inconsistencies should be clarified in deriving many conclusions from the vast, though incomplete, experimental data.
5. Attempts should be made to make protective surface layers that are resistant to leaching, especially in relation to glass composition and to the leachate composition.
6. From the viewpoints of both the structural damage and the radiolytic effects of radiation, efforts are needed to obtain comprehensive data on the effects of radiation on glass leaching.

7. For a coherent picture of the long-term behavior of site-specific glass leaching, it is strongly recommended that longer-term tests should be performed to give greater assurance to the validity of predictions.
8. In the modeling given within PNL-5157, an overview should be made to explain why so many models have been developed to represent the glass leaching process; each separate model seems to have been developed to fit a single experimental data set, so that a multiplicity of models is used to represent the behavior. The models developed should somehow be unified.
9. The existing models on glass leaching are very confusing in dealing with the thermodynamic equilibrium state and with kinetic (dynamic) data. They tend to use these two states in a single equation. There should be justification for using these two states simultaneously.
10. Extant on the effects of flow rates and on leachate compositions are sufficient to be modeled.
11. The effects of temperature can be readily modeled to account for the effects of temperature on accelerations of tests results.
12. Finally, in using PROTOCOL for a numerical evaluation of leach rates, a better description of math, logic, input, reliability and usages may be needed. PROTOCOL is not described well enough to permit the non-user to become familiar with its details.

APPLICABILITY TO DATA TO LICENSING

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

(a) Relationship to Waste Package Performance Issues Already Identified

- | | |
|-----------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 2.3 | when, how, and at what rate, will radionuclides be released from the waste form. |
| 2.3.1 | regarding physical, chemical and mechanical properties of the waste form with time |
| 2.3.2 | regarding solubility of the waste form under repository conditions |
| 2.3.2.1 | regarding possible dissolution mechanisms of the waste form under repository conditions |
| 2.3.2.1.1 | which waste form dissolution mechanism or mechanisms are most likely? |
| 2.3.5 | how will packing, container materials (including overpacks, canisters, and any special corrosion-resistant alloys or spent fuel rod cladding, if applicable) and/or their alteration products interact with the waste form to cause its alteration and/or effect release of radionuclides? |
| 2.2.4.2 | concerning the effects of radiation on the corrosion failure modes and associated corrosion rates for the waste package container |

2.2.4.2.1 concerning the predicted rate of radiolytic generation of hydrogen, oxygen and other species due to gamma radiation in the vicinity of the waste package containers.

6.2 classical leaching model

6.2 role of protective layer in leaching

6.3 role of solution concentration in leaching

6.4 thermodynamic model of leaching in confined and stagnant solutions

6.5 dispersion of leach products

(b) New Licensing Issues

6.5 leaching kinetics of crystalline silicates as complementary tools for glass leaching.

6.6 Application of non-mechanistic model

(c) General Comments on Licensing

SDI006, UD 8915, SER. DD016

File(s) searched:

File 6:NTIS - 64-89/ISS15
(COPR. 1989 NTIS)

Sets selected:

Set	Items	Description
1	0	WASTE(W)PACKAGE?
2	4	CANISTER?
3	57	CORROSION
4	13	LEACHING
5	131	GLASS
6	2	VITRIFICATION
7	190	S3-S6/OR
8	0	HIGH(W)LEVEL(W)WASTE?
9	91	RADIOACTIVE(W)WASTE?
10	8	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
07jul 03:40EST PR 11/5/1-25 (items 1-1)

Total items to be printed: 1

26

SDIO06 SUMMARY

User:010543 , File 6
TITLE:DIALOG SDI PRINTS for UD=8915

PAGE: 2

SDIO06, UD 8915, SER. DD016

File(s) searched:

File 6:NTIS - 64-89/ISS15
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Sets selected:

Set	Items	Description
1	0	WASTE(W)PACKAGE?
2	4	CANISTER?
3	57	CORROSION
4	13	LEACHING
5	131	GLASS
6	2	VITRIFICATION
7	190	S3-S6/OR
8	0	HIGH(W)LEVEL(W)WASTE?
9	91	RADIOACTIVE(W)WASTE?
10	8	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
07Jul	03:40EST	PR 11/5/1-25 (Items 1-1)

Total Items to be printed: 1

26

SDI103, UD 8912, SER. DD017

File(s) searched:

File 103:DOE ENERGY - 83-89/MAY(ISS12)

Sets selected:

Set	Items	Description
1	416	WASTE(W)PACKAGE?
2	20	CANISTER?
3	229	CORROSION (1974 DEC)
4	73	LEACHING (1974 DEC)
5	174	GLASS (1974 DEC)
6	87	VITRIFICATION (1974 DEC)
7	429	S3-S6/OR
8	137	HIGH(W)LEVEL(W)WASTE?
9	366	RADIOACTIVE(W)WASTE?
10	119	NUCLEAR(W)WASTE?
11	20	(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
07Jul 17:38EST PR 11/5/1-25 (Items 1-20)

Total items to be printed: 20

SDIO32, UD 8908, SER. DD022

File(s) searched:

File 32:METADEx 66-89/Aug
(Copr. 1989 ASM International)

Sets selected:

Set	Items	Description
1	5	HIGH()LEVEL()WASTE? ? OR RADIOACTIVE()WASTE? OR NUCLEAR()WASTE?
2	2046	STEEL? ? OR ZIRCALOY? ? OR TITANIUM? ? OR COPPER
3	2	1*2
4	0	ANNA FRAKER, 223, B-254, X6009

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

Date Time Description
13JUL 07:45EST PR 3/5/1-25 (Items 1-2)

Total items to be printed: 2

SDIO32, UD 8910, SER. DD022

File(s) searched:

File 32:METADEx 66-89/OCT
(Copr. 1989 ASM International)

Sets selected:

Set	Items	Description
1	3	HIGH()LEVEL()WASTE? ? OR RADIOACTIVE()WASTE? OR NUCLEAR()WASTE?
2	1895	STEEL? ? OR ZIRCALOY? ? OR TITANIUM? ? OR COPPER
3	1	1*2
4	0	ANNA FRAKER, 223, B-254, X6009

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

Date Time Description
13sep 04:18EST PR 3/5/1-25 (items 1-1)

Total items to be printed: 1

29

SDI006, UD 8919, SER. DD016

File(s) searched:

File 6:NTIS - 64-89/ISS19
(COPR. 1989 NTIS)

Sets selected:

Set	Items	Description
1	5	WASTE(W)PACKAGE?
2	6	CANISTER?
3	44	CORROSION
4	8	LEACHING
5	84	GLASS
6	6	VITRIFICATION
7	129	S3-S6/OR
8	6	HIGH(W)LEVEL(W)WASTE?
9	82	RADIOACTIVE(W)WASTE?
10	13	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 Time Description
08sep 13:54EST PR 11/5/1-25 (items 1-3)

Total items to be printed: 3

30

SDI008, UD 8909, SER. DA016

File(s) searched:

File 8:COMPENDEX PLUS - 70-89/SEP Copr. Engineering Info
Inc. 1989)

Sets selected:

Set	Items	Description
1	1	WASTE(W)PACKAGE?
2	0	CANISTER?
3	264	CORROSION
4	29	LEACHING
5	267	GLASS
6	8	VITRIFICATION
7	553	S3-S6/OR
8	5	HIGH(W)LEVEL(W)WASTE?
9	25	RADIOACTIVE(W)WASTE?
10	9	NUCLEAR(W)WASTE?
11	0	(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
15sep 12:29EST PR 11/5/1-20 (no items to PRINT)

Total items to be printed: 0

31

SDI103, UD 8917, SER. DD017

File(s) searched:

File 103:DOE ENERGY - 83-89/SEP(ISS17)

Sets selected:

Set	Items	Description
1	10	WASTE(W)PACKAGE?
2	25	CANISTER?
3	178	CORROSION (1974 DEC)
4	51	LEACHING (1974 DEC)
5	107	GLASS (1974 DEC)
6	35	VITRIFICATION (1974 DEC)
7	327	S3-S6/OR
8	155	HIGH(W)LEVEL(W)WASTE?
9	458	RADIOACTIVE(W)WASTE?
10	68	NUCLEAR(W)WASTE?
11	11	(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
17sep 18:56EST PR 11/5/1-25 (items 1-11)

Total items to be printed: 11