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Dr. Charles G. Interrante, Program Manager Metallurgy Division - Corrosion Section National Bureau of Standards U.S. Department of Commerce Gaithersburg, MD. 20899

Dear Dr. Interrante:

• :

We have reviewed the August 1988 NIST Monthly Letter Report for FIN A-4171, "Evaluation and Compilation of DOE Waste Package Test Data." Comments on the MLR are presented below in Attachment 1. Please revise the document reviews as noted herein and resubmit them in the next MLR.

Actions resulting from this letter are considered to be within the scope of FIN A-4171. No changes in costs or delivery of contracted products are authorized. Please notify me immediately if you feel this letter will result in additional costs or delay in delivery of contracted products.

Sincerely,

Charles H. Peterson Engineering Branch Division of High-Level Waste Management Office of Nuclear Material Safety and Safeguards

Enclosure: Att. 1

cc: w/Att. 1:

Dr. Neville Pugh, Director Metallurgy Division, NIST Dr. David Anderson, Group Leader Metallurgy Division, NIST

DISTRIBUTION WITH ATT. 1

	Central File PDR REBrowning, HLWM JJLinehan, HLPN DChery, HLGP		NMSS/RF LPDR BJYoungblood, HLWM RAWeller, HLEN MSilberberg, RES		HLEN/RF CNWRA	TLSS	TLSS JOBunting, HLEN KCChang, HLEN	
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# ATTACHMENT 1

# COMMENTS ON MONTHLY LETTER STATUS REPORT AUGUST 1988 (FIN A4171)

1. p 3, NNWSI Category 1 reports

Please terminate the reviews of UCRL-53795 and SAND85-7117 and substitute other reports that deal more directly with waste package materials. Under "Reviewer Comments", enter a note that detailed reviews were not completed.

2. p 4, Category 2 reports

Delete UCRL-96703, UCRL-96555, UCRL-53645, and UCRL-96318 from the waiting list. These documents may be entered into the database, but enter only the information for the block "Data Source".

3. p 6, Vitrified Waste Forms, Category 2

In view of comments below on the Wilson/Shaw report, we suggest you do not review any of the papers in MRS Proceedings unless you cannot get copies of the full reports on which they are based.

4. p 6, Task 2

We again request that the text in this section provide specific information on what is being "continued" from month to month.

5. p 6, Task 3

Regardless of how short the work month was, some time was charged to the project so the MLR should indicate what was accomplished in the activities mentioned. How many specimens are being prepared? What equipment is being built? How far along are these activities? How far along are the corrosion and pitting studies?

Although there is a general description of the three tasks in our files, there do not appear to be specific statements of the actual experimental plans. Do such plans exist?

## COMMENTS ON DOCUMENT REVIEWS

- 2 -

## General

Please revise the format to that agreed upon in June 1988. The categories and sequence are:

DATA SOURCE DATE REVIEWED PURPOSE/SCOPE KEYWORDS CONTENTS AMOUNT OF DATA TEST CONDITIONS UNCERTAINTIES IN DATA DEFICIENCIES/LIMITATIONS IN DATABASE CONCLUSIONS COMMENTS OF REVIEWER RELATED HLW REPORTS (OPTIONAL) APPLICABILITY OF DATA TO LICENSING AUTHOR'S ABSTRACT (OPTIONAL)

It is not necessary to make sentences out of the information to be entered. Just as the date of the review is simply entered as a date, so may the contents be described, e.g., as: 8 p: 5 figures, 5 tables, 6 references. The reason is to speed up scanning of the PC screen by the user as well as to reduce the effort needed to produce these document reviews.

Given the limited resources, it is far more preferrable to expend these on an in-depth critique of the substance of the report rather than taking time to list the titles of figures and tables. If these are listed, then some summary statement should be made as to what may be concluded from these figures and tables. Selectivity may be practiced.

## Specific Reviews

UCRL-94633: Experimental Study of the Dissolution of Spent Fuel at 85°C in Natural Ground Water. December 1986.

1. Delete redundant words. Under (a) Organization Producing Report, use:

Lawrence Livermore National Laboratory for the U.S. Department of Energy under Contract W-7405-Eng-48.

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- 3 -
- 2. In the citation use periods to separate main blocks of information.

Wilson, C. N. and Shaw, H. F. "Experimental Study of the Dissolution of Spent Fuel at 85°C in Natural Ground Water". UCRL-94633. December 1986.

- 3. The second document reviewed also listed the names of the reviewers. We favor the inclusion of these names.
- 4. Keywords should be inserted after Purpose/Scope.
- 5. Delete Type of Data. The material in Scope of Report might be included in Purpose/Scope above. However, it should be expanded to note that the tests were done with actual J-13 well water and on specimens prepared from actual spent fuel rods.
- 6. Delete Materials/Components as this information might be included in Test Conditions.
- 7. P9, Test Conditions

There were three series of tests, not several; there were several cycles of tests.

- 8. Delete Methods of Data Collection/Analysis
- 9. Delete redundant words: under Amount of Data, it is unnecessary to repeat that there are five tables and five figures.
- 10. Delete the lead sentence under Conclusions as unnecessary. Study of this document, however, indicates we should have two subsections:
  - (a) Author's Conclusions(b) Reviewer's Conclusions

Rather than forcing the user to shift back and forth from screen to screen, the reviewer's comments on each conclusion should be placed immediately after that conclusion. Format suggested is to begin a separate paragraph as follows:

#### CONCLUSIONS

1. Uranium concentrations...in the Series 2 tests.

Comment: Phraseology is vague. In place of "higher levels", the author should state the actual levels. However, more basically, this is not a conclusion but a result.

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- 11. Additional Comments on the Original Document
  - a. p 2: The authors should state how the external surfaces of the test specimens were decontaminated.
  - b. p 3: Periodic removal of 10-30 mL samples of leachate and replacement with equal volumes of fresh J-13 water does not really simulate the flow-through in a tuff repository. First, we are dealing with flow in an unsaturated zone. Second, replacement with a 10% slug of water does not represent the more gradual change in ion contents obtained with a distributed replacement.
  - c. p 3: Were the interior surfaces of the apparatus checked for residual radiation after the final acid rinse?
  - d. p 3: The authors state that concentrations measured for filtered sample fractions were not significantly different from those measured in unfiltered samples. Thus, any particles remaining in the filtered solution were less than 0.4 microns. But they continue: "However, the laser-excited fluorescence method used is not sensitive to uranium contained in fine solid particles that may have been intially present in the unfiltered fractions." Is the "conclusion" still valid? In any case, the data should be presented so that the reader can make an independent judgement as to whether there was any "significant difference". No statement is made as to whether any colloids were present.
  - e. Figure 2 shows a line representing the concentration that would result if 1.0E-05 x the original inventory of uranium in the test sample were dissolved in the 250-mL test volume. It appears to be about 3 ug/mL. A sample calculation should be given. Is this figure less than the solubility limit? What is the sensitivity of the measurements? Why is there an early peak in concentration for uranium and not for any of the other nuclides (plutonium, technetium, carbon)?
  - f. The curve for bare fuel exposed at 25°C in silica vessels appears to level off after 220 days at about 1 ug U/mL vs the indicated limit of about 3 ug/mL. Thus, after the canister and the Zircaloy cladding have corroded away, the potential for an excessive release exists. This illustrates the importance of having a complete scenario of events: when does the temperature at the surface of the canisters drop below certainly 85°C, and preferably less?
  - g. The leach curves for undefected, hole defected, and slit defected clad fuel at 85°C are essentially the same. One would think this is worth drawing a conclusion about, e.g. minor defects in the cladding do

not signifcantly change the release rate, which is two orders of magnitude below the indicated 3 ug/mL limit.

- h. p 4: The authors state that iron began to appear in the 33-day samples from all the bare fuel tests. However, the table presented suggests that the first measurements made after 0 days were not made until 33 days. The statement appears imprecise: iron was detected at 33 days, and may have started to appear earlier. By the way, were any blanks run?
- p 4: The authors speculate that corrosion of 304SS could have affected the leaching process through consumption of oxygen in the sealed test vessel. There was an air space in the test vessels. If this were 100 mL, at standard conditions there would be about 30,000 ug of oxygen available. No estimate is given of how much oxygen might have been consumed by the postulated corrosion products, but since microgram quantities of corrosion products are involved, the speculation does not seem well-founded.
- j. p 4: The authors speculate that actinide species might have been reduced to less soluble, lower valence states. By what? The oxygen present would appear to be sufficient to account for the corrosion observed. Can metallic iron, chromium, or nickel reduce actinide oxides from a thermodynamic view?
- k. p 4: The authors do not comment on the possible effect of radiation.
- p 4: The authors note that the measured fractional uranium release in the Series 3 bare fuel tests was about 5 times greater at 85°C than at 25°C, with most of the difference being in the quantity of uranium in the terminal acid strip solution. This is a result. A conclusion is that the uranium deposited on the walls of the apparatus. Would not the same thing occur in the repository?
- m. p 6: The authors state that the dramatic drop in dissolved Tc-99 activity in the bare fuel test at 85°C after the 62-day sample is thought to be related to weld corrosion that appeared to have occurred in this test during this time. Only one data point is shown after 62 days. Another explanation is experimental error. How many replicates were used?
- n. p 6: The authors speculate that Tc was likely initially released as soluble  $TcO_{4}$  ion that was later either sorbed onto corrosion products

or was reduced to a less soluble species as more extensive oxidation of iron occurred in this test relative to the other tests. Again, thermodynamically, does iron reduce the pertechnate ion? What is the role of the excess oxygen present? o. p 6: Cs-137 is discussed but no data are given. No rationale is given for the result that all of the fission products appear to be preferentially released relative to the actinides.

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- p. p 6: The authors note that the amount of C-14 found was about the same for the bare fuel (0.49 uCi/g) as for the cladding (0.53 uCi/g). Some of the release (5-10% ?) is from the exterior of the cladding. If the latter is true, perhaps it will not be possible, at least for C-14, to take credit toward containment for the cladding. This should be noted in the conclusions.
- q. p 7: The authors state that at 25°C the chemistry of the well water remained stable, but that at 85°C 60 - 80% reductions in Ca, Mg, and Si were observed. Some comment should have been made as to the significance of these reductions to the corrosion process.
- r. A telephone conversation with one of the authors, Charles N. Wilson, revealed that a later report has been prepared and is expected to be released in perhaps three months. Some of the conlcusions will have been changed.
- PNL-5157: Chapter 3, Environmental Interactions. From "Final Report of the Defense High-Level Waste Leaching Mechanisms Program". August 1984.
- 1. Comments on format are similar to those for the preceding review.
- p 14, TEST CONDITIONS: Where a line of information carries over to the next line, indent the second line at least three spaces to facilitate reading. E.g.:

Contact (flush) time: 60 h, 28 days, 600 h, 1 month, 56 days, 3 months, 6 months, 12 months.

3. None of the text offered appears to be in the nature of comments, but rather a summary of certain results. We need to know whether the data provided, the results found, and the conclusions drawn are valid and defensible.

## COMMENTS ON THE FINANCIAL REPORT

The current balance appears satisfactory. Please provide the more detailed monthly statement as previously requested.