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July 1—September 30, 1980

University of California

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Compiled by

A. E. Norris

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Compiled by

A. E. Norris

ABSTRACT

An H. B. Robinson-2 spent fuel wafer was dissolved for radiochemical and mass spectroscopic determinations of the source terms for elements that have been leached from similar spent fuel wafers. The values for the 22 measured nuclides are presented on an atoms/g UO_2 basis. Three papers have been accepted for presentation at nuclear waste management meetings next quarter. The conclusions of two of the papers have been given in previous quarterly reports. The third paper discusses the interpretation of leach rate data plots. Data interpretation appears to be most reliable when the incremental leaching rate is plotted against the mid-times of the leaching durations. When, instead, cumulative leach rates are plotted against cumulative leaching time, the leaching intervals should be in an invariant sequence.

I. PERSONNEL

This report covers some of the activities of the following individuals employed at the Los Alamos Scientific Laboratory:

G. K. Bayhurst	A. E. Norris
G. E. Bentley	A. E. Ogard
E. A. Bryant	C. J. Orth
C. J. Duffy	J. E. Sattizahn
G. F. Grisham	K. W. Thomas

II. PROGRAM OBJECTIVES

This program investigates aqueous reducing conditions for their relevance to the geologic disposal of radioactive wastes. One objective is that of producing aqueous reducing conditions at pH 8 with very low oxygen fugacities. The second objective is that of measuring the differences in concentration of reactor products leached from spent fuel in water under oxidizing and under reducing conditions.

III. PROGRESS REPORTED PREVIOUSLY

This program was funded by the Office of Waste Isolation on March 24, 1978. The first report of progress was the monthly progress report dated April 20, 1978. Quarterly reports have been issued since the first quarter of fiscal year 1979. Accomplishments in this program have included the publication of the results of a literature survey that collected data about the diffusion properties, chemical forms, and morphological characteristics of reactor products in UO_2 fuel elements; measurements of the solubility of UO_2 in water under reducing conditions; and the determination of concentrations of reactor products that were leached from spent fuel wafers with water under oxidizing and under reducing conditions. The results from the spent fuel leaching experiments did not yield data as unambiguous about the effects of oxidizing and reducing conditions as expected, in part because the actual reducing conditions in the leaching solutions were not well characterized. However, the data obtained indicate that solubility limits may play an important role in determining the concentrations of the lanthanide and actinide elements in the leachant solutions.

A major part of this program has been directed toward obtaining aqueous reducing conditions at pH 8 that result in measured UO_2 solubilities approximating those expected from thermodynamic calculations. The measured solubil-

ities of pure uraninite in deionized water have been used to infer that oxygen fugacities as low as $\sim 10^{-50}$ bars have been achieved.

IV. PROGRESS DURING THE CURRENT QUARTER

A. Source Term Measurements

One wafer of H. B. Robinson-2 spent fuel was dissolved to permit radiochemical measurements of the source terms for the leaching experiments that have been reported previously. The spent fuel is described in detail in the status report ONWI/Sub/79/E511-01200-11. The source term sample was the middle wafer cut from segment #32. The UO_2 was dissolved in aqua regia, but the Zircaloy-4 cladding remained intact under the conditions used. The mass of the UO_2 dissolved, 5.06 g, was determined by difference between the mass of the UO_2 plus cladding before dissolution and the mass of the cladding afterwards.

The results of the source term solution analyses are shown in Table I. The uncertainties associated with the experimental values shown in Table I are dominated by those arising from the hot cell procedure required for dissolving the spent fuel wafer. Each experimental value is estimated to have an overall uncertainty of $\pm 5\%$ or more. However, the counting errors for ^{243}Am and ^{242}Cm increase the uncertainties for these two nuclides to $\pm 20\%$.

B. Technical Presentations

Abstracts of papers were prepared for technical presentations at two nuclear waste management meetings. The papers are:

"Are Solubility Limits of Importance to Leaching?" by Allen Ogard, Glenn Bentley, Ernest Bryant, Clarence Duffy, Jane Grisham, Edward Norris, Carl Orth, and Kimberly Thomas, to be presented in Boston at the International Symposium on the Scientific Basis for Nuclear Waste Management during the November meeting of the Materials Research Society.

"Eh and Fission Product Solubilities: Two Factors in the Leaching of UO_2 ," by A. E. Ogard and C. J. Duffy, to be presented in Gatlinburg at the Oak Ridge National Laboratory's Conference on the Leachability of Radioactive Solids, December 9 through 12.

"The Misused and Misleading IAEA Leach Test," by A. E. Ogard and E. A. Bryant, to be presented at the same conference as the preceding paper.

The information in the first two papers has been included in the previous quarterly reports. The information in the third paper is summarized here.

TABLE I
ISOTOPIC CONTENT OF H. B. ROBINSON-2 SPENT FUEL WAFER

<u>Atoms/Gram Oxide Corrected to day 72.450, 1979</u>	
<u>Nuclide</u>	<u>Experimental</u>
⁹⁰ Sr	2.73×10^{18}
¹⁰⁶ Ru	$>1.71 \times 10^{16}^*$
¹²⁵ Sb	7.45×10^{15}
¹³⁴ Cs	7.61×10^{16}
¹³⁷ Cs	3.75×10^{18}
¹⁴⁴ Ce	1.41×10^{16}
¹⁵⁴ Eu	3.92×10^{16}
¹⁵⁵ Eu	1.28×10^{16}
²³⁴ U	3.24×10^{17}
²³⁵ U	1.34×10^{19}
²³⁶ U	7.68×10^{18}
²³⁸ U	2.15×10^{21}
²³⁷ Np	4.09×10^{19}
²³⁸ Pu	3.25×10^{17}
²³⁹ Pu	1.08×10^{19}
²⁴⁰ Pu	5.23×10^{18}
²⁴¹ Pu	2.18×10^{18}
²⁴² Pu	1.29×10^{18}
²⁴¹ Am	6.55×10^{17}
²⁴³ Am	2.2×10^{17}
²⁴² Cm	1.8×10^{13}
²⁴⁴ Cm	5.10×10^{16}

*Uncertainty in the ruthenium chemical yield permitted determination only of a lower limit.

The first point made in the paper "The Misused and Misleading IAEA Leach Test" concerns the time intervals against which incremental leaching rates should be plotted. The conventional method used by most American investigators of spent fuel leaching appears to be that of plotting the fraction of a given element leached as a function of the cumulative time of leaching. This

method is that recommended in the 1978 draft ISO standard for Long-Term Leach Testing of Radioactive Waste Solidification Products. However, the proposed International Atomic Energy Agency standards for leach testing [At. Energy Rev. 9, pp. 195-207 (1971)] specify that the incremental leaching rate is to be plotted against a function of time that approximates the mid-time of the leachant renewal period. These two different methods of plotting leach rate data can lead to different interpretations of the results, particularly when the duration of the leaching intervals change. An analysis of hypothetical leach rates indicated that data plotted as a function of the mean time of the leachant renewal period, in the manner of the IAEA proposal, is likely to be more useful for interpretation of the results than the other method.

The second point in the paper being summarized is similar to the first, but concerns the interpretation of cumulative leaching data plotted against cumulative time. This display is one that is recommended in the IAEA proposal. The resulting plots can be misleading when leaching data from different experiments are compared, unless the experiments are done in an identical manner with an invariant sequence of leachant renewal intervals.

These two points do not appear to have been appreciated by all those who are performing leach tests.

C. Conclusions

The experimental source term measurements were completed this quarter. These measurements permit the recalculation of spent fuel leaching data without reliance on calculated source term values.

The interpretation of data from spent fuel leaching experiments has been found to depend, at least in part, on the manner in which the data are plotted. Artifacts in the plots can occur, particularly when the data are derived from experiments in which the leaching intervals vary. Data interpretation appears to be most reliable when the incremental leaching rate is plotted against the mid-times of the leaching durations. When cumulative leach rates are plotted against cumulative leaching time, the leaching intervals should be an invariant sequence.

V. PROBLEMS

The difficulty that has been experienced in preparing quarterly reports on time should be alleviated next quarter, when this program is combined with the

Oklo - Natural Fission Reactor Program. One quarterly report henceforth will cover the activities of these two programs under the title Natural Repository Analogue Program.

VI. ACTIVITIES PLANNED FOR NEXT QUARTER

The three papers whose titles are listed in Section IV will be presented at the indicated nuclear waste management meetings. Work will commence on a topical report to be completed by April, 1981, concerning the immobilization of uraninite in ore deposits.