Quarterly Progress Report

Project Title: Nuclear Waste Criticality Analysis

Principal Investigator: William G. Culbreth

Covering the Period: 1 July 1995 - 30 September 1995

Date: October 19, 1995

Summary:

The control of criticality in spent nuclear fuel is necessary during all phases of fuel management during storage, transportation, and permanent disposal. The work completed to date is described below. The tasks indicated in the original proposal include:

- Task 1 -- Seek coverage by an approved quality control program.
- Task 2 -- Review documents related to criticality.
- Task 3 -- Attend criticality meetings and workshops.
- Task 4 -- Maintain an expertise in criticality.

The current work is covered by the University of Nevada System QA plan, however, coverage under a more thorough plan will be sought during the next quarter to ensure that the results of any criticality studies that we conduct can be used during NRC licensing.

Work Completed to Date:

1. Review of documents.

Based on task #2, the YMPSCO requested a review of contractor reports related to criticality. Two documents have been reviewed including:

- a. MPC Subsystem Nuclear Criticality Safety Strategy: An Overview
- b. Criticality Safety Strategy for Spent Nuclear Fuel Packages: An Overview

Copies of the reviews are included in appendix A. Other reports are currently in review.

2. Submission of Abstracts for Workshop

During August, the Office of Environmental Management of the U.S. Department of Energy scheduled a workshop dealing with the stabilization and immobilization of plutonium contaminated wastes titled the: "Plutonium Vitrification/Immobilization Workshop". We submitted two abstracts relating to criticality in vitrified MOX fuel and the relationship of MPC studies to plutonium wastes. The abstracts were accepted for presentation at the workshop, however, the workshop was rescheduled for December 12-15 in Washington, D.C. Copies of

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the abstracts are attached as appendix B.

3. Student Involvement

Two students and an postdoctoral research associate have been employed on the project. They have been active in the criticality research by conducting literature reviews and have assisted in computational studies related to the project. Louise Steeps is a senior in a civil and environmental engineering student. She has collected a number of articles related to the Oklo phenomenon and is enrolled in an independent study course in the "Criticality Analysis of the Oklo Natural Reactors". Dr. James Ventresca, a computational fluid mechanician, has been assisting in the development of a computer program to model the behavior of the Oklo site. Cliff Lawson, an undergraduate student in civil engineering, is assisting with the studies.

4. The Oklo Phenomenon

The natural nuclear reactors discovered at Oklo, French Gabon, Africa in the early 1972 provide a unique resource for the study of the immobilization of fission products. There have been a number of articles published regarding the concentration of fission products in and around the site of subterranean uranium "lenses" where fission occurred over 2 billion years ago and continued to operate for approximately 150,000 years. Oklo has been used as a model for Yucca Mountain where the immobilization of similar fission products will be necessary for 10,000 years. An abbreviated summary of some of the literature that we have obtained related to the Oklo phenomenon is included in appendix C.

One view of the Oklo site not often discussed concerns the conditions that caused nuclear criticality. The Oklo reactors continued to operate until the effective enrichment of the uranium in the "lenses" decreased to approximately 1.5%. Water filtering through the layers of surrounding sedimentary deposits provided the moderator for the fission reaction. Neutron poisons such as boron, samarium, and cadmium were not found in large quantities in the Oklo deposits. These conditions were ideal to cause criticality in natural uranium over 2 billion years ago.

It should be noted that the average effective enrichment of much of the spent fuel to be buried at Yucca Mountain is on the order of 1.5% and, in the presence of water, may also sustain a chain reaction. The knowledge gained through the Oklo natural reactors should be applied to Yucca Mountain to discover if a similar chain reaction can be sustained in the spent fuel and under what conditions.

Kuroda [1-3 in Appendix C] predicted the possibility of a natural uranium-fueled reactor in 1956 in order to explain the discrepancies in the concentration of certain isotopes of lead at certain geologic sites in Johanngeorgenstadt pitchblende formed 2.1 billion years ago. Dr. Kuroda retired from the University of Arkansas and now lives in Las Vegas. Dr. Culbreth and Louise Steeps visited with Dr. Kuroda to discuss the Oklo phenomenon and his impressions of criticality at Oklo. Dr. Kuroda's past work indicates that the Oklo reactors may have operated in an unsteady manner with a period of 2 to 3 hours. He believes that its periodicity may have exhibited behavior similar to a geyser where fission heat vaporized water ejecting it violently

from the region surrounding the reactor lens. The absence of water as a moderator shut down the fission process until liquid water reentered the reactor site beginning the process again. A criticality event similar to Oklo would be expected to disperse water-soluble fission products far from the site of the uranium lens. Dr. Kuroda's work also indicated that the Oklo reactor operated between the temperatures of 183°C and 452°C.

The work of Dr. Kuroda and others indicates that should critical conditions occur within Yucca Mountain wastes at any time in the future, the periodic behavior of the Oklo reactors may indicate how a subsequent "criticality event" would occur and how fission products might be dispersed about the repository horizon.

At this point, three important observations arise from the Oklo reactors that should be studied for their relevance to the Yucca Mountain site:

- Fission products were effectively immobilized for a long period of time.
- Fission occurred in natural uranium with an enrichment of approximately 1.5%. Does this indicate the possibility of criticality in Yucca Mountain wastes?
- If fission occurs at Yucca Mountain at any time in the future, will the resulting criticality events exhibit a behavior similar to that of the Oklo reactor?

5. Oklo Simulation Program

Due to the importance of the Oklo reactor data as it relates to an underground repository at Yucca Mountain, we have started the development of a simulation program to predict the behavior of the Oklo reactor. A draft outline of the code is included in appendix D. While the outline will likely be modified during further development, we intend to use modules from KENO-V and ORIGEN for criticality and radionuclide decay and fission product buildup. We are in the process of developing subroutines to describe the properties of water in both liquid and vapor phases.

6. Scale Software

Our chief neutron criticality code is the Oak Ridge code, SCALE 4.1. This PC-based code is NRC approved for nuclear reactor criticality analyses. Oak Ridge (RISC) has developed a newer version, SCALE 4.3 that should be available by the end of October. We will acquire the new version as soon as it is available.

Appendix A

Reviews of Criticality Reports

- 1. Review of "MPC Subsystem Nuclear Criticality Safety Strategy: An Overview"**
- 2. Review of "Criticality Safety Strategy for Spent Nuclear Fuel Packages: An Overview"**

Title: Review of the Article Titled: "MPC Subsystem Nuclear Criticality Safety Strategy: An Overview"

By: William G. Culbreth, Ph.D.
University of Nevada, Las Vegas

Date: May 18, 1995

1. Overview

The reviewed article describes the intended DOE strategy to ensure nuclear criticality safety during the storage, transportation, and disposal of high-level nuclear waste. An important aspect of criticality safety outlined in the article is the intention to seek burnup credit with the NRC through a probabilistic assessment of criticality risk. To accomplish this, tests are proposed to increase the accuracy of cross-section databases for key neutron absorbers, criticality experiments to verify the computer codes used to analyze criticality, and to assess axial distribution of burnup in fuel assemblies. Burnup credit will be sought based on fissile depletion, the presence of key actinide absorbers, and key fission product absorbers.

2. Critique

The methodology proposed to seek burnup credit is sound and addresses the key issues that must be verified before seeking such credit. The proposed plan and established techniques are adequate for ensuring criticality safety during storage, transportation, and during pre-closure operations. More data will be needed before adequate criticality control can be assured during post-closure and waste isolation periods, however.

As outlined in the article, long-term control of criticality will be assessed through the use of probabilistic risk models. These models will take into account the time that it takes for corrosion and subsequent failure of the waste package, the basket material, the cladding, and the spent fuel. Accurate data on corrosion rates are necessary if the beneficial impacts of fission product neutron absorbers and added neutron poisons are to be acceptable in maintaining the effective neutron multiplication factor at safe levels over the life of the repository. Corrosion rate data and the leaching rates of fission products from spent fuel and of proposed neutron absorbing basket materials are not well known. The data currently available has a high degree of uncertainty. The temperature, humidity, and groundwater chemistry in the proposed repository will significantly affect the transport of poisons and fissiles from the MPC over the 10,000 year period of waste isolation. Accurate corrosion rate data for spent fuel, additive poisons, and the MPC wall materials are necessary if the PRA (probabilistic risk assessment) method is to be used to ensure

subcritical conditions over the period of waste isolation. All corrosion data acquired in the future should include an uncertainty analysis.

The article implies that criticality control is only required over the 10,000 year period cited for waste isolation. Spent fuel may be capable of sustaining criticality long after 10,000 years. Who decides this ethical issue?

3. Specific Issues

- a. Page 8, 4th and last paragraphs: U-233, a fissile material, is not placed on the list of actinides to be considered in the criticality analysis. The last paragraph a justification for eliminating this isotope from the analysis. A further reason that it may be ignored is due to its small concentration in spent fuel. For 10-year old fuel (3.11% initial enrichment, 30 GWD/MTU burnup), the concentration of U-233 is only 0.00252 grams per metric ton in heavy metal. After 10,000 years, this has increased to 4.7 grams. This only amounts to 0.05% of available uranium fissile.
- b. Page 9, paragraph 1: Will the NRC express any concerns about possible radial variations in burnup within a single fuel assembly? Of the 269 fuel pins within a typical PWR fuel assembly (Westinghouse, 17 X 17 pin), many are adjacent to the 20 control pins within the assembly. These pins are in regions of suppressed neutron flux and will suffer less fissile depletion than those further from the control rods.
- c. Page 12, paragraph 3: For disposal, this paragraph gives the impression that fissile depletion helps to decrease the value of k_{eff} in spent fuel over a one million year period. In reality, this is very dependent upon initial enrichment and burnup. It should be noted that the concentration of fissile material remains very constant over 10^6 years as indicated in figure 1. While decay removes Pu-239 and Pu-241 from the inventory of fissile material, the amount of U-235 will increase due to the decay of heavier radioisotopes. This results in a fairly constant concentration of fissile for at least 10^6 years.

The second point raised in the paragraph indicates that k_{eff} decreases over time ($> 20,000$ years). This is true only for low to moderate initial enrichments and burnups. Figure 2 shows the effect of time on 4.7% initial enrichment, 45 GWD/MTU spent fuel. Although the values of k_{eff} are low, they increase after 1,000 years. As seen in the figure, fission product poisons also play an important role in decreasing k_{eff} .

- d. Page 15, paragraph 1: Current DOE assumptions indicate

that no filler material will be added to the MPC and no backfill will be placed in the drifts. This paragraph should indicate these assumptions to the NRC, unless they have been changed.

- e. Page 17, paragraph 2: A set of CERES experiments are proposed to ensure the efficacy of neutron absorption cross-sections for important fission product neutron absorbers. Of those of major interest (Eu, Gd, Sm), only a subset of the available isotopes found in spent fuel are proposed for testing. Figures 3 through 5 show the relative concentration of these isotopes present in 3.11% initially enriched, 30 GWD/MTU spent PWR fuel. The 1,000 year inventory is shown. Isotopes preceded by a star (*) are those that will be tested in the CERES program, according to the report. Accurate cross-section data is needed for all of these isotopes unless:

- 1) Accurate absorption cross-section data is already available.
- 2) The indicated isotope has such a short half-life that testing is not possible.

Samarium Concentrations by Isotope
3.11%, 30 GWD/MTU, 1000 years

Figure 3

Europium Concentration by Isotope
3.11%, 30 GWD/MTU, 1000 years

Figure 4

Gadolinium Concentration by Isotope
3.11%, 30 GWD/MTU, 1000 years

Figure 5

**University of Nevada, Las Vegas
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May 24, 1995

To: Paige Zielinski, DOE, YMPO
From: Bill Culbreth

Subj: Review of "Criticality Safety Strategy for Spent Nuclear Fuel Packages:
An Overview", dtd. May 23, 1995.

1. Page 4, paragraph 2: This paragraph refers to a set of documents to be submitted to the NRC with the initial topical report to be issued in May 1995. Did this document go out already?
2. Page 4, paragraph 3: A current application to the NRC for acceptance of transportation casks with "four, three, or two PWR assemblies for the initial enrichments up to 2.9, 3.5, and 4.5 [wt% U-235]" does not include burnup credit, yet the very next sentence says that once the NRC accepts the burnup topical reports, "OCRWM will seek burnup credit ... allowing 4 PWR assemblies with initial enrichment values of up to 4.5 [wt% U-235]." The two sentences appear redundant.
3. Page 8, paragraph 2: Small mistakes include: Am-242m (I don't believe that the "m" should be there), Cm 244, (add hyphen). Also, the sentence that starts "The results of ongoing analysis ...", should be analyses.
4. Page 13, figure: Transportation (10 - 100 year period) could be added to the figure for completeness.
5. Page 15, paragraph 2: The list of fission product neutron absorbers is limited (as I indicated in the earlier review).
6. Page 15, paragraph 3: There is a discussion about the MONK computer code and the last sentence makes it appear that the CERES experiments will be used to validate MONK which will in turn be used to validate SCALE. This chain of validation introduces uncertainty into the accuracy of the calibration of SCALE to the CERES experiments. SCALE should directly be calibrated to the CERES data without the extra "pass" through MONK.

Appendix B

Abstracts Submitted to the Plutonium Vitrification/Immobilization Workshop

**Sponsored by the U.S. Department of Energy
August 24, 1995 (Postponed)**

Abstracts removed at

Appendix C

Summary of Articles Related to the Oklo Phenomenon

1. Kuroda, P. K., The Origin of the Chemical Elements and the Oklo Phenomenon, Springer-Verlag, 1982.
2. Kuroda, P. K., "The Pre-Fermi Natural Reactor," *Journal of Radioanalytical and Nuclear Chemistry*, vol 142, no 1, pp. 101-112, 1990.
3. Kuroda, P. K., "The Pre-Fermi Natural Reactor: A Note on the Periodic Mode of Operation," *Journal of Radioanalytical and Nuclear Chemistry*, vol 142, no 1, pp. 113-118, 1990.
4. Kuroda, P. K., "Behavior of Radionuclides Around Oklo Reactor," presented at the Third International Symposium on Advanced Nuclear Energy Research - Global Environment and Nuclear Energy, 1989.
5. Gauthier-Lafaye, F., and Weber, F., "Natural Fission Reactors at Oklo," *Economic Geology*, vol 84, pp. 2286-2295, 1989.
6. Menet, C., Menager, M.-T., and Petit, J.-C., "Migration of Radioelements Around the New Nuclear Reactors at Oklo: Analogies with a High-Level Waste Repository," *Radiochimica Acta*, 58/59, pp. 395-400, 1992.
7. Loss, R. D., Rosman, K. J. R., et al, "Fission-Product Retentivity in Peripheral Rocks at the Oklo Natural Fission Reactors, Gabon," *Chemical Geology*, 76, pp. 71-84, 1989.
8. Nagy, B., Gauthier-Lafaye, F., et al, "Organic Matter and Containment of Uranium and Fissionogenic Isotopes at the Oklo Natural Reactors," *Nature*, vol 354, pp. 472-480, 12 December 1991.
9. Hidaka, H., Konishi, T., and Masuda, A., "Reconstruction of Cumulative Fission Yield Curve and Geochemical Behaviors of Fissionogenic Nuclides in the Oklo Natural Reactors," *Geochemical Journal*, vol 26, pp. 227-239, 1992.
10. Curtis, D., Benjamin, T. and Gancarz, A., "Geochemical Controls on the Retention of Fission Products at the Oklo Natural Fission Reactors," *Natural Analogues in Radioactive Waste Disposal*, The Commission of the European Communities, 1987.

Appendix D

Overview of the Proposed Analysis of Nuclear Criticality at the Oklo Sites

Plan for a Computer Program to Model the Behavior of the Oklo Natural Reactors

**William Culbreth
University of Nevada
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Abstract

The Oklo location in French Gabon, Africa was the site of a natural nuclear reactor some 2 billion years ago. An accumulation of extremely rich uranium ore occurred in strata approximately 1 meter thick within the Oklo site and the combination of water intrusion and a natural enrichment of approximately 1.5% provided the necessary conditions to sustain a continuous fission chain reaction. It has been estimated that fission took place for several hundred thousand years producing low levels of power and a host of radioactive fission products. Considerable work has been done to investigate the subsequent migration of fission products from the site of the reactors. Technetium, tellurium, plutonium, europium, and other substances not commonly found in nature do occur near the site of the reactor. The site has had considerable interest from the perspective of nuclear waste management because it demonstrated that fission products did not migrate far from the site of the reactor, even over the intervening period of 2 billion years.

The Oklo site is also interesting from the perspective of nuclear criticality in an underground repository. The effective enrichment of the high-level nuclear wastes considered for placement in Yucca Mountain are also about 1.5%. The decay of actinides over time will increase the concentration of U-235 in the fuel while maintaining a fairly consistent concentration of fissile to fertile for several million years. The question is, over a long period of time, how will the wastes in Yucca Mountain behave? The results from Oklo demonstrate that the isotopes of uranium and plutonium are effectively bound near the site of each reactor by adsorption in the surrounding rock. In the presence of water, fission occurred until enough of the U-235 decayed to bring the effective enrichment of the uranium ore to less than 1%. In Yucca Mountain, the enrichment of uranium will not decrease to these levels for another XX billion years.

After the spent fuel waste packages have corroded and distribution of their radioactive contents have begun spreading through the host rock of the repository, lighter radionuclides will diffuse away from the repository horizon into the volcanic tuff. Adsorption in the backfill and surrounding tuff will greatly retard the motion of uranium and plutonium. This phenomenon has been observed at Oklo and is seen as advantageous since it decreases the flow of radioactive material to the accessible environment. Unfortunately, it also may lead to the formation of actinide-rich "lenses" within the repository with little or no neutron poisons present. If water intrudes into these lenses, a critical event may be possible. What impact would this have? Venneri and Bowman¹ site the possibility of a nuclear explosion. While their analysis requires a specific set of unusual events to lead to this phenomenon, it may be possible to precipitate a steam explosion and eject radioactive material from the repository. A more likely event is

analogous to the behavior of the natural reactors at Oklo. In this scenario, the fissionable uranium will create small amounts of heat in the presence of water due to fission. As the temperature of the lens increases, water will be vaporized and driven from the core of the lens. The subsequent absence of water, a necessary neutron moderator, will decrease the neutron flux within the lens. It is expected that a steady-state fission reaction would result where the heat produced by fission that subsequently vaporizes water is just enough to maintain an equilibrium condition. The power levels would be very low (tens of watts). This steady-state operation may seem fairly benign, but it will create new fission products and these serve as a new source term for radioactive material in the repository.

At this time, there is not a clear model of the Oklo natural reactors. The total neutron fluence has been deduced from the inventory of fission products and their daughters in the geologic medium surrounding the reactor sites. The fission product inventory and the current disposition of uranium in the reactors provide an opportunity to create a model of the reactor behavior and to verify the model by comparing the current disposition of fission products to that observed at Oklo.

In the proposed project, a numerical model of the Oklo reactor site will be developed. A draft outline of the initial program is included below. The program is being written in ANSI-C.

A. Program Modules:

1. Define the reactor:
 - a. Distribution of fertile and fissile uranium.
 - b. Initial distribution of water.
 - c. Definition of bulk material.
2. Diffusion model:
 - a. Using a 1-D model, calculate the distribution of water as a function of time. This includes the conversion of liquid water into vapor.
 - b. Ignore the influence of air.
 - c. Assume that the phase change occurs, but water does not migrate due to heat.
3. Energy model:
 - a. Compute the transfer of energy by conduction, (convection).
4. Criticality model:
 - a. At each point in time, compute the value of k_{eff} , the subsequent power output as a function of position (1-D).
5. Radionuclide inventory:
 - a. Compute the depletion of fissile and the accumulation of fission products as a function of time and burnup.
 - b. Include the effects of fission product decay.
6. Report (display) the results.

B. Computational Procedure:

1. Start with an initial temperature, fissile inventory, and water content.
2. Compute keff, the neutron flux, and the power output as a function of time.
3. Calculate the temperature distribution within the reactor.
4. Use the temperature distribution to calculate phase changes in the reactor water.
5. Calculate the subsequent diffusion of water.
6. Calculate the production of fission products over the time step.
 - 6.a Calculate the diffusion of fission products and their decay with time. (next phase)
7. Calculate the depletion of fissile over the time step.
8. Report:
 - a. Fissile distribution.
 - b. Temperature distribution.
 - c. Pressure distribution (water).
 - d. Density distribution (water).
 - e. Fission product distribution.
 - f. Neutron flux distribution.
 - g. Quality distribution.
9. Repeat 1-8 for subsequent time steps.

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