bnl
au
BROOKHAVEN NATIONAL LABORATORY
ASSOCIATED UNIVERSITIES, INC.

Upton, Long Island, New York 11973

(516) 282-
FTS 666- 4094

Department of Nuclear Energy

August 24, 1984

WM-RES
WM Record File A-3167 BNL
WM Project 10, 11, 16
Docket No.
PDR ✓
LPDR B, N, S
Distribution: WICK
(Return to WM, 623-SS)

Mr. Everett A. Wick
High Level Waste Licensing Management Branch
Division of Waste Management
Mail Stop 623 SS
U. S. Nuclear Regulatory Commission
Washington, DC 20555

Dear Mr. Wick:

**BNL Review of Environmental Assessment (EA) Reports for Tuff and Basalt Repositories**

Enclosures 1 and 2 are in response to your letter dated August 16, 1984, in which you solicit BNL comments on issues to be addressed in the review of the subject DOE EA reports.

For the tuff EA review the issues outlined by Mr. T. Jungling are appropriate and we have added others to form an overall framework for barrier system evaluation.

In the case of the basalt EA report we suggest that a more limited review be carried out compared to that suggested by NRC since many of the issues identified will not greatly influence site selection procedures. We, therefore, suggest that some of the NRC-identified issues be grouped together and handled as a single item as outlined in Enclosure 2.

Please contact Dr. M. S. Davis or me if you require additional information.

Sincerely,

Peter Soo

Peter Soo, Associate Division Head
Nuclear Waste Management Division

PS:gfs
Enclosures
cc: M. S. Davis
W. Y. Kato
H. J. C. Kouts
D. G. Schweitzer
K. Chang, NRC
T. Johnson, NRC
T. Jungling, NRC
M. Tokar, NRC
Docket Control Center, NRC

WM DOCKET CONTROL CENTER
'84 AUG 27 P2:24

BROOKHAVEN NATIONAL LABORATORY

# MEMORANDUM

DATE: August 21, 1984

TO: File

FROM: P. Soo

SUBJECT: Review of Tuff Repository Draft Environmental Assessment (EA) Report (dated June 1, 1984)

A review of the subject EA report was carried out in order to correlate BNL findings with those of the NRC which are given in Appendix 1. Comments will be given below on the contents of Appendix 1 and additional issues which BNL deems to be important will also be given.

1. Geochemistry (EA Section 6.3.1.2)

NRC Comment No. 1

BNL agrees with NNWSI that the redox potential of the repository water may be affected by the presence of metallic materials. However, this effect could be small if the amount of oxygen present is such that the water is always saturated with this gas. Passive films on Zircaloy and stainless steel would greatly decrease the rate at which oxygen is removed from the water to the extent that these metallic materials will not control the redox potential. Thus, the implication by NNWSI that the groundwater could achieve lowered redox conditions is speculative and probably is quite unlikely. NNWSI should recognize that gamma radiolysis of groundwater will generate oxidizing species which will maintain an oxidizing environment.

NRC Comment No. 2

BNL agrees with NRC that some repository-water concentration effects are likely during the first few hundred years. DHLW and CHLW containers will cool to ≈100°C in 50 yr and 200 yr, respectively (McCright, 1984, Boston Waste Management Conference). Steam formation at these times will leave behind deposited salts from the water which, when contacted by liquid water at a later time, will likely give much higher levels of $Cl^-$ and $F^-$, etc., in the repository water. No experiments have been conducted to date to quantify this phenomenon. Simple tests with tuff and groundwater at 150°C (Knauss, 1984, Boston Conference Proceedings) do not consider the re-solution of deposited salts.

NRC Comment No. 3

See BNL comment immediately above.

NRC Comment No. 4

BNL does not agree with NNWSI that Zircaloy can effectively reduce the redox potential of the groundwater to the extent that radionuclide solubilities will be decreased. Quite likely, the Zircaloy will be quickly passivated and the rate of oxygen decrease in the water will be so slow at 100°C that the water will still be oxidizing. Hence, radionuclide solubilities will remain high.

It is not clear what NNWSI means when they say that the $UO_2$ fuel could be oxidized and lead to stress rupture of the cladding. For $UO_2$ to become oxidized, one presumably visualizes oxygen-bearing water penetrating the cladding to cause the oxidation. If the cladding is penetrated then further rupture of the cladding by fuel swelling is not important since the cladding has already failed.

2. Subsystem Preliminary Performance Assessments (EA Section 6.4.2.3)

NRC Comment No. 1

BNL agrees with NRC and NNWSI that the probable failure mode for the Type 304L stainless steel container is unlikely to be uniform corrosion. The estimated 3000-30,000 year uniform corrosion failure time may be realistic but it is felt that insufficient data are currently available to show this with any degree of assurance. The short-term test carried out by McCright (1984) lasted only 1000 hr and was for steam and liquid water conditions in the absence of irradiation and solute concentration effects. Longer term tests will be needed (5-10 yr) to determine if pitting, crevice corrosion, stress corrosion, or intergranular attack could occur. Also, no data have been generated on weld metal corrosion.

Stress-assisted attack, cited as a possibility by NRC, could be very important since stress-corrosion cracking has been observed by the Japanese in Type 304L stainless steel exposed to deionized water at 100°C in situations where crevices exist. A natural crevice would exist if the container is contacted by a crushed tuff packing or if the container is merely in contact with a borehole surface.

NRC Comment No. 2

BNL concurs with NRC that radionuclide solubilities used by NNWSI in their performance assessment studies are too preliminary to be taken as being valid. Deficiencies in the data base include:

a) Lack of information on the whole range of radionuclides in spent fuel, including the gaseous elements which may not be readily soluble and may be transported by convection. It is also not clear how the solubility data (Table 6.3.1.2-4) were derived in the calculations. What is the temperature and what are the uncertainties?

b) Lack of data on pH effects on solubility. Low pHs formed by gamma radiolysis of $O_2/N_2/H_2O$ mixtures could cause the generation of nitric acid which will enhance radionuclide solubility.

c) Radiocolloids could be present and give rise to enhanced release rates.

NRC Comment No. 3

BNL concurs with NRC that in all performance assessment calculations, the worst case containment times and release values should be emphasized. In addition, there should be increased emphasis on evaluating waste package component interaction effects. Whether these are beneficial or detrimental is not known at this time.

## 3. Additional Areas for Investigation

With respect to the engineered barrier system it is felt that a more quantitative assessment is needed before the system can be adequately characterized. In particular, NNWSI should be encouraged to:

a) Specify the preclosure environment with respect to steam/air conditions around the waste package. Gamma irradiation effects on nitric acid formation need to be determined so that corrosion and embrittlement of stainless steel can be meaningfully studied. Welds and sensitized steels must be evaluated since they are likely to possess inferior corrosion properties.

b) Stress analyses are required to determine if the approximately 1-cm thick container can fail mechanically during the containment period, taking into consideration corrosion losses in the metal.

c) The effects of solute concentration in the repository water due to steam formation and salt precipitation need to be assessed and quantified. Accelerated uniform corrosion, pitting and crevice corrosion in the concentrated water, in the presence of gamma irradiation, must be outlined.

d) Cyclic wet-dry conditions, such as those cited by McCright (1984), could also lead to concentration of $Cl^-$ at container surfaces and need to be addressed with respect to corrosion. Examples in the literature clearly show, for example, that intermittent wet-dry cycling can lead to stress-corrosion cracking in chloride-containing waters.

4. <u>Work to be Undertaken by BNL</u>

BNL work in support of the tuff repository EA review will focus on all of the items listed under subsystem performance assessment (Section 2). Emphasis on the geochemistry area will be limited to the extent that it is required to help define the barrier system environment. In addition, BNL proposes that the issues addressed in Section 3, above, be further investigated as part of the EA analysis and review.

The depth of the BNL work will involve a clear outlining of the specific issue, in terms of its importance to quantifying barrier system behavior. Supporting data from the open literature will be cited. It is estimated that approximately 2.0 scientific man-months of effort will be needed to complete the task. True overall cost will be about $20K including support and secretarial services.

PS:gfs

## NNWSI EA Review Questions and Comments

### Geochemistry (6.3.1.2)

1) It is stated that the oxidation state of the water contacting the waste forms may be lowered by interaction with the stainless steel canister as well as the Zircaloy cladding. In the process the passive film on the stainless steel will be further stabilized. Would the redox potential and the passivity of the stainless steel be significantly affected by this interaction?

2) Oxygen, bicarbonate and dissolved organic carbon are identified as the groundwater constituents which must remain unchanged or change only slightly during the life of the repository as a result of their effect on speciation, sorption and the solubility of the waste elements. However, changes in other ionic species may effect performance, e.g., changes in chlorine or fluorine concentrations could effect waste package performance if they were to occur in the next few hundred years. Are such changes possible in a short geologic time period?

3) It is stated that, "There are no known conditions in the pre- or post-emplacement environment water chemistry expected to compromise the performance of the metal barrier in the repository setting." Apparently, they have not considered the possibility of ionic species becoming concentrated as a result of decay-heat groundwater evaporation, which could lead to increased problems from localized corrosion.

4) Two potentially adverse conditions relevant to spent fuel have been identified as a result of the oxidizing environment.

   a) Increased solubility of the radionuclides and

   b) The possibility that the $UO_2$ may become oxidized and led to stress rupture of the cladding.

   They may deserve further consideration or comment by the NRC.

### Subsystem Preliminary Performance Assessments (6.4.2.3)

1) Uniform corrosion was assumed as the mode of failure for a 304 L stainless steel container. Furthermore the uniform corrosion rate data corresponds to a package lifetime ranging from 3000-30,000 years, i.e., the time necessary to completely corrode the entire canister wall thickness. The referenced life time was chosen as 10,000 years.

   a) The choice of 10,000 years is obviously not the most conservative, moreover the choice of uniform corrosion is probably non-conservative.

# BROOKHAVEN NATIONAL LABORATORY

## MEMORANDUM

DATE: August 24, 1984

TO: File

FROM: M. S. Davis

SUBJECT: Plan for Review of the Basalt Repository Environmental Assessment Report

A review of the Environmental Assessment for a basalt repository will be carried out with the primary objective of assessing issues which relate specifically to the interaction of the near-field environment with the waste package. The specific areas which will be emphasized in the review are outlined below and are related to several of the detailed issues outlined by the NRC in the letter of 8/16/84 (Appendix 1).

<u>Near-Field Environment</u>

Evaluation of the anticipated performance of the waste package and the engineered barrier system will include, to the extent practicable, all near-field variables and the impact of the waste package on these environmental variables. The important objective of this assessment is to determine if interaction of the environment with the waste package has been adequately addressed. This part of the assessment is related to issues 9, 10, 18 and 26 of Appendix 1.

<u>Containment Barrier</u>

Anticipated failure mechanisms of the primary containment barrier, carbon steel container, will be assessed for the adequacy with which they have been addressed by the EA. This evaluation will include an assessment of both uniform, localized corrosion and, to the extent possible, the interaction with the overpack (bentonite/basalt backfill). This assessment is related to issues 5, 7, 8 (17?) of Appendix 1.

McCright states that the limiting use of austenitic stainless steel is rarely uniform corrosion but rather some form of localized corrosion (1).

b) Also is it reasonable to expect the canister to provide containment for the full lifetime predicted by the experimentally observed corrosion rate? Could failure occur by a combination of mechanical stresses on a partially corroded canister at a time significantly shorter than the predicted lifetime?

For the purpose of Environmental Assessment it may not be necessary to define a precise waste package failure mechanism or life time, albeit a realistic, conservative estimate should be used which reflects the current uncertainty especially when the waste package estimates impact the results of other calculations which are essential to the Environment Assessment, e.g., radionuclide travel times.

2) Waste form releases

a) In Wilson and Oversby's (2) spent fuel cladding containment tests only uranium, plutonium and cesium releases were measured. Radionuclides with higher solubilities such as technetium, carbon, and iodine may be expected to show greater releases than those which were measured.

b) This leads to the question of whether congruent leaching, controlled by the $UO_2$ matrix, is a valid assumption or will releases by long lived, highly soluble elements contribute significantly. Could such radionuclides possess release rates which are greater than that for Pu from bare fuel pellets ($2x10^{-5}$ per yr), which NNWSI has identified as their upper limit?

3) Fractional release rate

a) As with other calculations the most conservative values are not used but rather average or "reference case" values. Should not worst case calculations at least be attempted and not just in this instance but throughout the performance assessment?

b) Are the hydrology and geochemistry groups satisfied with the values of water flux and uranium solubility limits.

<u>References</u>

(1) McCright, R. D. et al., <u>Selection of Candidate Canister Materials for High-Level Nuclear Waste Containment in a Tuff Repository</u>, UCRL-89988, 1983.

(2) Wilson, C. N. and V. M. Oversby, <u>Spent Fuel Cladding Containment Credit</u>, HEDL-SA-3017, UCRL-89869, 1984.

Release Rate

An evaluation of the information provided in the EA on the performance of package materials with respect to their ability to control the release from the EBS to $10^{-5}$/yr will be carried out. This evaluation will concentrate primarily on the waste form, with emphasis on spent fuel, and the ability of the bentonite/basalt backfill to contribute to controlled release. This evaluation is related to issues 2, 13, 14, 19, 20 and 21 of Appendix 1.

The anticipated level of effort to complete this review is 2.5 staff months or $25K.

MSD:gfs

CHANG/REVIEW/84/07/23/0



AUG 16 1984

## EA REVIEW ON WASTE PACKAGE

FUNDAMENTAL QUESTIONS:

1. Does the EA include a description of the site specific emplacement environment to enable DOE to generate a clear definition of a waste package program for waste package research, development, design and performance analysis considered appropriate for the site? (10 CFR 60.11a(8) and 60.135(a))

   This description must be adequate for defining an up-coming waste package program to address interaction of the waste package with the repository environment taking into consideration the following factors: solubility, oxidation/reduction reactions, corrosion, hydriding, gas generation, thermal effects, mechanical strength, mechanical stress, radiolysis, radiation damage, radionuclide retardation, leaching, fire and explosion hazards, thermal loads and synergistic interactions.

2. Does the EA include a description of the preliminary waste package design(s) being considered for the specific site(s) so that (a) performance assessment studies may be pursued with meaningful estimates of waste package containment life and controlled release of radionuclides from the waste package after loss of containment (10 CFR 60.140) and (b) waste package monitoring programs may be planned to focus on the internal condition of the waste package (10 CFR 60.143)?

3. Is there anything about the waste package design(s) and performance that would lead us to conclude that a site is unsuitable for further characterization?

   Does the EA provide adequate data for a qualitative evaluation of the site on the specific issues concerning the waste package (appendix C of NUREG-0960, see enclosures )

   Is there any data about the site on subjects related to these specific issues which indicate that unless the specific issues are resolved, the site is unsuitable for further characterization?

| | | | | | | |
|---|---|---|---|---|---|---|
| | | | | | | |
| | | | | | | |
| 84/08/15 | | | | | | |

BWIP's Discussion of Waste Package and Subjects Related to Waste Package

BWIP's Waste Package Design

The draft BWIP EA does not give a description of the waste package design. A waste package conceptual design is however, mentioned in Section 5.1.2.2.7.

Section 5.1.4 of the EA is titled "Waste Package Optimization Study." This section is however still in preparation and may or may not provide more detailed information than that in the present draft EA.

Performance Analysis

It is clear from reading Section 6.4.2.2.2 of the EA that for the purpose of calculating radionuclide transport in performance assessment, BWIP's analysis "assumed that the waste package design can meet the regulatory requirements of containment time." Therefore, there may not be any discussion on waste package performance analysis in the final BWIP EA.

Emplacement Environment Pertinent to Waste Package Design and Performance

This is quite adequately addressed for EA's purpose under sections related to geology, geochemistry, and hydrology in Chapters 5 and 6, especially, Section 6.3.

Chapter 6 is written in standardized sections under:

1. Qualifying Condition
2. Evaluation Process
3. Favorable Condition
4. Potentially Adverse Condition
5. Conclusions of Qualifying Conditions

Suggested Approach to NRC Review of Draft BWIP EA

1. Evaluate rationale and data presented in Chapter 6 of the EA and comment on whether they are acceptable to NRC or not. The extent and areas of concern in this review is detailed in the NRC review plan and further discussed for the waste package by the "Fundamental Questions" of page 1.

2. Identify specific issues not covered in Appendix C of NUREG-0960 which NRC considers must be addressed in this EA and discuss the information BWIP must provide to settle the issues.

3. Prepare written comments on the EA to address Waste Package related subjects in a format agreed to by the BWIP review team.

Enclosure 1

Waste Package Issues Stated in NUREG 0960, March 1983

A. When and how does water contact the waste package?

B. When and how does water contact the waste form?

C. When, how and at what rate are radionuclides released from the waste package?

D. What are the conditions which affect criticality?

To address these issues, a series of specific issues has been developed in Appendix C of NUREG-0960, as follows:

1. What are the possible mechanisms by which water will penetrate packing materials around containers?

2. To what extent over time will groundwater flow, temperature, or other effects change the ability of packing materials to control flow through those materials? What chemical and physical changes are possible? What are the chemical and physical properties?

3. What are the hydrothermal conditions with time at the surfaces of the waste form and containers and within packing materials which influence property changes and radionuclide release?

4. What are the possible mechanical failure modes for the container?

5. What are the chemical and physical property changes in container materials and what are the resultant properties?

6. What are the mechanical loads on containers vs. time? How do the packing materials affect the loading?

7. What are the possible corrosion failure modes for the containers?

8. What is the effect of packing materials on the corrosion mechanisms for the container?

9. How do Eh, pH, and $PO_2$ change with time in the vicinity of the container in the packaging?

10. What is the radiolytic generation of hydrogen, oxygen, and other species due to gamma radiation in the vicinity of the containter?

11. What is the dependence of the oxygen removal rate from packing materials upon temperature, pressure, radiolysis, packing materials, physical characteristics, grounwater flow rates, and composition and time?

12. How do microbes effect conditions affecting corrosion modes? What effect do microbes have on the conditions affecting transport?

13. What is the solubility of radionuclides vs. time in the vicinity of the waste form and packing materials? How are radionuclides released from the waste form?

14. What properties of the waste form change with time and alter the ability of the waste form to contribute to the overall performance of the repository system or impact the performance of other barrier materials and properties of the site?

15. What is the effect of water residence time on release of radionuclides from the waste form?

16. What are the ranges of residence times of a unit volume of water in contact with a unit area waste form and when do the residence times occur? For spent fuel how do hulls change the effective residence time?

17. How do the packing (spent fuel hulls if applicable), canister, and container materials and/or their alteration products interact with the waste form to cause its alteration and/or effect release radionuclides?

18. How does the Eh, pH, and $PO_2$ change with time in the vicinity of the surface of the waste form? (relates to 9)

19. What is the production of particles and colloids (by or near the waste form) which can hold or transport radionuclides or effect waste form degradation?

20. For spent fuel what are the failure mechanisms for hulls and what is their failure rate?

21. What are the transport and retardation processes and how do they effect the flux of radionuclides with time in packing materials.

22. How do the species which incorporate radionuclides change with time in the waste package? (This includes particles, colloids and solubles.)

23. Can actinides be concentrated to increase heating in the packing materials or create a potential for criticality?

24. How do radionuclides migrate through failed containers and how does this change with time? (relates to 5)

25. What are the convective flows in the waste package vs. time?

26. Does alpha radiation in the waste package materials affect chemistry and hence transport and species identification?

27. What are the conditions which affect criticality?

Enclosure 2

Additional Issues for EA Review

1. What are the effects of dissolved gas (especially methane) on the release rate of radionuclides from the waste package?