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Place: Dublin, California; Howard Johnson's Motel

October 18-19, 1983 Date:

Particpate in NNWSI Waste Package Workshop Purpose:

Attendees: NRC: Don Alexander, Mike Bell, John Starmer, Everett Wick and Tom Jungling

DOE: Don Vieth

ORNL: Clyde Claiborne and Gary Jacobs

Aerospace: Ken Stephens

Claudio Pescatore and Peter Soo BNL:

LLL: Lyn Ballou (Project Manager), Dan McCright, Virginia Oversby, Larry Ramspott, and Jean Younker

State of Nevada: Carl Johnson

### Specific Comments

MEMORANDUM FOR:

FROM:

SUBJECT:

Waste Package Environment

A majority of the experimental work thus far conducted by NNWSI (Nevada Nuclear Waste Site Isolation) has been to characterize the interaction of groundwater with the tuff rock. The major objectives of this work were to determine the corrosion environment at the metallic barriers, the

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chemical composition of the fluid available for leaching and to obtain data which could be used to validate the computer codes which will be used for long-term data. Specimens of Bullfrog Tuff taken from the saturated zone of Yucca Mountain were tested with J-13 well water by varying temperature, rock to water ratio and contact time. Experiments were performed in Parr bombs and Dickson bombs at 90 and 150, and 250°C, respectively. The most significant variations in water chemistry with time were as follows:

- Silicon concentrations increased rapidly to and, in some instances, beyond the cristobalite (high temperature form of SiO<sub>2</sub>) saturation limit.
- <sup>°</sup> In the Parr bomb experiments the pH of the solution increased from 7.0 to 8.5 or 9.0, while the pH remained at 7.0 throughout the Dickson bomb experiments. The rise in pH in the former was attributed to the degassing of the teflon bombs.

Work is still in an early stage (longest runs are approximately four months in duration), but will continue in order to obtain long term data necessary to the validation of geochemical models, viz., EQ3/6, and to the understanding of reaction kinetics.

From the work of the U. S. Geological Survey, the most probable site for the repository in tuff is the densely welded Topopah Spring Member of Yucca Mountain, which is composed of an extensive network of vertical fractures. The proposed repository will lie at a depth between 300 and 400 meters, while the upper bounds of the water table approaches 480 meters. The calculated amount of water recharge into the rock varies from 1 to 10 mm/year. Using 8 mm/year of recharge LLNL calculated the maximum amount of water which could contact an individual canister during a year's time period, assuming that the total volume of water which falls within the repository boundaries divided by the number of canisters comes in contact with each of the canisters (a conservative estimate). The maximum amount of water contacting a canister is 40 liters/year for defense high-level waste (DHLW) and 208 l/yr for commercial high-level waste (CHLW).

To analyze the need for packing material in complying with 10 CFR 60, it was assumed that the dissolution of the waste glass will be controlled by

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the saturation of the silicon, therefore the maximum release of SiO<sub>2</sub> was calculated to be 7.6 gm/yr and 1.46 gm/yr for CHLW and DHLW, respectively. By relating the release of SiO<sub>2</sub> to the dissolution of the waste form a numerical value for the release of radionuclides was calculated to be 0.2 x 10<sup>-5</sup> parts/yr for DHL<u>W and <3.5 x 10<sup>-5</sup> parts/yr</u> for CHLW (borosilicate glass). By exposing/bare fuel pellets from spent fuel assemblies to a corresponding volume of water a release rate of 7 x 10<sup>-5</sup> parts/yr was obtained. Therefore it was concluded that the<sub>5</sub>glass waste forms could meet the controlled release requirement of 10<sup>-5</sup> parts/yr without packing material while only the spent fuel assemblies would retain the need for packing.

The two current candidate materials for packing are crushed tuff and crushed tuff containing 15% iron-bearing smectites. An arbitrary packing thickness of 15 cm was selected for evaluation purposes. Among the noted advantages for the use of packing were the reduction of corrosion by preventing localized salt drips, reducing water contact and providing porous flow which would aid in nuclide retardation by increasing the available surface area. The following factors were expressed as areas of concern if packing would be employed; thermal and chemical stability, fabricability and the effect on water chemistry and on temperature. Heavy emphasis was put on the concern that packing would decrease the thermal conductivity compared to that of the host rock and essentially act as an insulator, creating higher waste package temperatures than would normally exist.

Waste Form

The waste form presentation concentrated on PNL 76-68 borosilicate glass as the reference CHLW material due to the large data base available but also included an examination of zircaloy cladding for spent fuel. It was suggested that if the controlled release criteria of 10 CFR 60 was met, then the EPA requirement would also be met for all isotopes except six (2-Am, 3-Pu and Np). Therefore, since LLNL believes the controlled release criteria will be met for the glass waste forms, as previously stated, only the release and transport of these six isotopes need be considered. Phase I testing consisted of exposing the glass waste forms to environments saturated with J-13 well water plus a combination of tuff and 304L stainless steel. The results indicated that the rock contributed silica to the water and thus implied that the actual leach rate of the glass would decrease.

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Phase II tests are currently being conducted to determine the effect of unsaturated conditions on the waste form. To simulate these conditions, a waste form sample will be exposed to one drop of water every 3.5 days then the liquid will be collected and analyzed for elemental release and waste form alterations.

The amount of protection afforded by zircaloy cladding on spent fuel rods was examined by intentionally introducing defects in the cladding and then monitoring the release rates of various radionuclides. Preliminary results show that some inhibition to nuclide migration is likely to be provided by the cladding.

Metal Barrier

For the metallic barrier the current emphasis is on 304L (0.02% C max) stainless steel with secondary consideration given to three other austenitic alloys, 316L and 321 stainless steel and Incoloy 825. The low carbon content was stressed as a remedy to the formation of grain boundary chromium carbides which can sensitize the steel and result in intergranular stress corrosion cracking. However, 304L SS is still susceptible to localized corrosion (pitting and crevice) and transgranular cracking in the presence of chloride ions. Ongoing testing which includes immersion, crevice and electrochemical tests, has not shown evidence of susceptibility to localized or stress corrosion cracking. Livermore further claims that the small quantity of water which will be present at the Yucca Mountain site will greatly reduce the possiblity of serious corrosion. Serious concerns were raised as to the reliability of predicting long-term performance of 304L SS based on short term experiments. It was emphasized by the NRC that long term and accelerated testing should be conducted.

Waste Package Design and Analysis

The conceptual reference design consists of a vertically emplaced, 1 cm thick canister constructed of 304L stainless steel with packing only for spent fuel assemblies. Although the possibility of packing for the glass waste forms has not been completed eliminated, LLNL, for reasons previously discussed, does not perceive the need for it. The selections of 304L and the secondary materials previously mentioned were made by designating equal importance to four factors (corrosion resistance, mechanical properties, weldability and cost) and then analyzing 17

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different alloys before selecting the four currently under consideration. The rationale for assigning equal weight to each factor was questioned by P. Soo (BNL/NRC) on the basis that the corrosion resistance will ultimately determine the life of the package while the other factors can be modified if necessary.

The computer code TACO2D is being employed to provide the thermal analysis of the conceptual design. The structural integrity will be analyzed by subjecting the canister to a series of transportation test albeit actual shipping plans are to use a transportation cask.

A shortcoming of the design is that although it was geared towards complying with 10 CFR 60 it admittedly neglected to consider the EPA standards, which include unanticipated events.

Performance Assessment

The evaluation of variability in the geological siting will be concerned with: the changes that might result in the rock from testing and drilling, how can data be extrapolated to the large scale in situ situation and what are the time dependences of stress, percent saturation, pore pressure, temperature and infiltration.

A Waste Package Performance Assessment Code (WAPPA) is to be used to analyze the time to the first release of radionuclides and the subsequent release rates. The code is currently on line at Livermore but is undergoing modifications on the process models. One specific modification currently being made is to adapt the code to perform in an unsaturated environment instead of the saturated type for which the code was originally written. As the process models are modified they will be validated by running them against an individual specific code, e.g., the thermal process model has been run and compared to TACO2D and the outputs did show agreement.

The proposed reliability/uncertainty analysis consisted of a deterministic-probabilistic approach that uses a deterministic degradation model with bar-graph distributions of the individual input parameters.

Testing in the Exploratory Shaft

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A brief discussion was presented on the planned tests at the exploratory shaft. Three areas of testing are planned to improve estimates of water infiltration: the effect of moisture content, the imposition of a high recharge event and testing of packing material and the engineered capillary design. Geochemical models as well as heat and mass transport codes will be used to characterize the necessary conditions.

> ORIGINAL SIGNED DI Thomas L. Jungling Engineering Branch Division of Waste Management

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## CLOSEOUT COMMENTS OF NRC AND DOE ON THE NNWSI WASTE PACKAGE WORKSHOP

### Dublin, California October 18-19, 1983

The following points are raised by NRC.

#### **Observations**

- LLL, in considering environments that could affect the waste, did not show that they have considered the effects of salts (silica, calcite) that could be deposited when groundwater is vaporized. These deposited salts could plug the pores of the rock and prevent steam, and later water, from escaping from the emplacement hole. These deposited salts could also be redissolved and result in a higher dissolved salt content in water contacting the waste package.
- 2. LLL bases its waste form leaching or dissolution estimates on the assumption that leach rates are controlled by maximum silica solubility in the groundwater. This assumption is not yet accepted by the scientific community. For example, colloidal silica should be considered.
- 3. The sensitivity of the waste package to water flow and temperature should be considered. For example, the distance of the repository from the water table varies (a 6 to 8 degree slope was mentioned) and this could result in different waste packages experiencing different water flow rates and temperatures.
- 4. NRC encourages continuation of the spent fuel cladding studies and suggests that actual failed fuel rods be used.
- 5. The Draft EPA standard 40 CFR 191 is tentative and may be changed. If it is changed, NRC must reevaluate 10 CFR 60 to see if it must be changed.
- 6. Graphs should contain error bands. For example, data used to show differences may actually overlap if error bands are considered. Also, experiments should be replicated for each test condition.
- 7. The horizontal borehole design for spent fuel uses a 15 centimeter annulus of packing material that is encapsulated. Thus, an annular gap between the waste container and the packing material will be necessary to install the waste package. The benefit of the packing

material, therefore, is not obvious because the groundwater will flow through the annular gap rather than through the backfill.

8. The use of equilibrium codes in obviously non-equilibrium situations is not clear. It is not clear how kinetics are treated. For example, what are the effects of flow rate variation? Another example is that the observed compatibility of J-13 well water and Topapah Springs water may be just lack of time for reaction.

- 9. Parameter values should be estimated reaslistically, even when "conservative" values are used. Otherwise, there is no way to judge how conservative the estimate is. The estimated parameter values should also include reaslistic upper and lower bounds.
- 10. The selection of candidate container materials from only two generic classes of metals is very risky. It is especially risky to select <u>three</u> austenitic stainless steels because they are susceptible to stress corrosion cracking in the presence of chloride ion and oxygen.
- 11. The number of stainless steels tested should be minimized and the number of metals from other generic classes (e.g., titanium or zirconium-based alloys, carbon steels, and low alloy steels) should be increased.
- 12. The ranges of repository conditions should be defined so that meaningful corrosion tests can be designed. The conditions whose ranges need to be defined include temperature, water chemistry, pH, gamma dose rate, stresses on the canister, aqueous phase (water or steam) and oxygen level.
- 13. A range of tests should be selected which encompass anticipated environmental conditions; tests under accelerating conditions should also be carried out. The data bases obtained from these tests should be modeled to obtain constitutive equations for observed failure modes. Accelerating parameters may include higher temperature, higher stress level, and groundwater containing higher concentrations of impurities and increased levels of oxygen.
- 14. Multiple heats of each candidate canister metal should be tested to ensure that heat-to-heat variations are quantified and factored into the derived constitutive equation.
- 15. Very long-term tests (10-15 years) should be performed under prototypic conditions to investigate stress corrosion cracking, pitting, and uniform corrosion rates.

- 16. Weld qualification must include demonstration of acceptable mechanical properties and corrosion resistance.
- 17. Experimental effort can be minimized by selecting materials with as few documented failure modes as possible.

### Items for Further Discussion

- 1. Susceptibility of container materials to stress corrosion cracking.
- 2. Reliability of the waste package, including demonstration of the reliability of the method itself.

### Information Requested

- 1. Where can iron-bearing smectite for the packing material be obtained?
- 2. What information will be gained from the near-field hydrothermal tests and how does the information relate to the information needs of the project?
- 3. NRC is interested in all supporting analyses for the conclusions presented in the meeting.

The following points are raised by DOE.

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### Observations

- 1. The meeting provided the waste package development staff (LLNL) supporting the NNWSI project good insight into the questions important in a regulatory arena and the requirements necessary to address issues of importance.
- 2. It provided good insight into the logic of the NRC staff and its contractors and how they might review information important to licensing.
- 3. The meeting was conducted in a professional and open manner which facilitated an effective and constructive exchange of information.

## Topics for Future Discussion

- 1. NRC's view point of what constitutes the physical evidence for a waste package containment being "substantially complete."
- 2. Discussion of the treatment of uncertainty analysis with respect to likely events and unlikely events. Attention should be given to testing requirements for selecting unlikely events.
- 3. Discussion of characterization of spent fuel can we use an ensemble average or must we characterize the content of every package?
- 4. Discussion of the changes to 10 CFR 60 as they relate to the waste package in the unsaturated zone.

Information Requested

- 1. References by Peter Soo regarding the provisions of stress corrosion cracking of austenitic stainless steel.
- 2. Information on the level of detail that NRC believes is prudent in a detailed materials test plan.
- 3. Information regarding the level of design detail for the waste package that NRC believes should be included in the SCP.
- 4. Identification of references for test methods that can be used to accelerate testing of materials of a waste package.

Donald L. Vieth (Date) Director Waste Management Project Office Michael J. Bell (Date) Deputy Director Division of Waste Management

# AGENDA

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# NNWSI WASTE PACKAGE WORKSHOP WITH NRC

## October 18-19, 1983 Howard Johnson's Hotel Dublin, CA

# Tuesday, October 18

0900-0930 Introductory comments - DOE/NRC/LLNL
0930-1200 I. Waste Package Environment in the Repository a. Ground water chemistry as affected by thermal and radiation fields o water chemistry after rock-water interactions experiments at 90°C and 150°C including experiments planned for FY84 in gamma field b. Hydrothermal effects in very near field
o rock response to near field conditions Younker (15) c. Current USGS estimates of ground water flux and flow mechanisms at Yucca Mountain
Oversby (10) o annual volume of water contacting waste package d. Packing Material
Oversby (15) o preliminary assessment/discussion of need for packing material for release control
Younker (15) o potential functions under unsaturated emplacement conditions Oversby (20) o preliminary candidates for packing material in tuff environment: importance of thermal conductivity of packing material
1200-1300 Lunch
1300-1500 II. Waste Form
Oversby (60) a. Characteristics of waste forms: assumed reference cases b. General approach to waste form testing: plans for acquiring necessary data
<ul> <li>(c. Saturated waste form testing</li> <li>(d. Unsaturated waste form testing (Argonne National Laboratory)</li> <li>(30)</li> <li>(a) Unsaturated waste form testing (Argonne National Laboratory)</li> <li>(b) Unsaturated waste form testing (Argonne National Laboratory)</li> <li>(c) Unsaturated waste form testing (Argonne National Laboratory)</li> <li>(c) Unsaturated waste form testing program (HEDL)</li> <li>(c) Characteristics of spent fuel population</li> <li>(c) Characteristics of spent fuel population</li> <li>(c) Characteristics of role of cladding in containment and release control</li> </ul>
<pre>1500-1700 III. Metal Barrier: Testing and Evaluation (30) a. Selection of candidate materials for container b. Rationale and supporting data for material selection (30) o restrictions/specifications for container fabrication and welding (15) o potential and probable corrosion modes to be investigated (45) o test conditions to acquire supporting data and plans for</pre>
long-term testing

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Wednesday, Octobe	er 19
0800-1000 IV. (30)	<pre>laste Package Design and Analysis    a. Design requirements/historical development of waste package       designs</pre>
0'Neal (15) b. (45) c. (20) d. (10) e.	Current reference conceptual designs for unsaturated zone Analysis of current reference designs o thermal: very near field temperatures as a function of time and waste package design o structural o criticality o economic Alternative conceptual designs: single or multiple metal barrier Future plans
1000-1200 V. P Bell (30) a. Younker (30) b.	erformance Assessment/Uncertainty Analysis NRC comments on Reasonable Assurance Anticipated environmental conditions in an unsaturated emplacement environment o variability in characteristics of host rock o potential effects of variability on performance of waste
Revelli (30) c. Sutcliffe (30) d.	Deterministic analysis of time to first release and long-term release rates o potential use of WAPPA o WAPPA subsystem model reviews to determine applicability to unsaturated emplacement conditions o model development plans Uncertainty Analysis o reliability/uncertainty: approaches for predicting waste package performance o preferred approach for waste package system
1200-1300 Lunch	
1300-1400 VI. Younker (60) a. c.	Planned Tests in Exploratory Shaft at Yucca Mountain Description of tests Purpose of specific experiments: methods/procedures/parameters Predictive modeling of results
1400-1500 NRC ca	ucus
1500-1630 Feedba	ck and wrap-up discussion

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Phone Numbers for Messages

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415-829-2144