October 21, 2003

Joseph D. Ziegler, Director
Office of License Application and Strategy
U.S. Department of Energy
Office of Repository Development
P.O. Box 364629 M/S 523
North Las Vegas, NV 89036-8629

SUBJECT: PRE-LICENSING EVALUATION OF EVOLUTION OF THE NEAR-FIELD ENVIRONMENT (ENFE) KEY TECHNICAL ISSUE (KTI) AGREEMENTS 2.07 AND 2.08

Dear Mr. Ziegler:

The U.S. Nuclear Regulatory Commission (NRC) has completed its evaluation of the U.S. Department of Energy's (DOE's) September 27, 2002, submittal on KTI Agreements ENFE 2.07 and 2.08. NRC found that the information provided in the ENFE 2.08 agreement submittal satisfied the intent of the original agreement. Therefore, NRC considers ENFE 2.08 complete. However, NRC found that the information provided in support of ENFE 2.07 did not satisfy the intent of the original agreement. As a result, NRC needs additional information on ENFE 2.07. The additional information needed by NRC is described in the enclosed NRC evaluation of the agreement submittal.

With regard to ENFE Agreement 2.07, DOE agreed to identify specific coupling relationships that are included or excluded from the DOE TSPA model and to give the technical basis for their inclusion or exclusion. The DOE response focused on radionuclide transport processes in the engineered barrier system rather than on the coupling of processes and conditions that affect the chemical environment important to drip shield and waste package corrosion. Coupled processes and conditions conducive to radionuclide transport may be different from those conducive to corrosion. Consequently, the response to ENFE Agreement 2.07 did not cover all relevant aspects of the original agreement. DOE should provide NRC with the additional information needed to review all aspects of ENFE Agreement 2.07. The response should identify and evaluate the physical and chemical coupled processes and conditions that can affect corrosion or breaching of the drip shield and waste packages, and should document the technical bases for including or excluding these coupled processes and conditions in DOE's performance assessment models.

With regard to ENFE Agreement 2.08, DOE agreed to provide a stronger technical basis for the suppression of individual minerals predicted by equilibrium models. The DOE response presented an adequate summary of the criteria for determining if mineral suppression is appropriate, a list of minerals included or excluded from current engineered barrier system models, and the technical bases for suppressed and unsuppressed minerals. NRC found the response satisfies ENFE Agreement 2.08. DOE's response suggests that DOE is likely to have enough information on this issue for NRC to accept a licence application for review.

The NRC expects that results of studies conducted between now and the submittal of the license application will provide additional support needed to justify the suppression of some mineral reactions.

If you have any questions regarding this matter, please contact Gregory Hatchett at 301-415-3315 or by e-mail to GXH@nrc.gov.

Sincerely,

/RA/

Janet R. Schlueter, Chief
High-Level Waste Branch
Division of Waste Management
Office of Nuclear Material Safety
and Safeguards

Enclosure: NRC Review

cc: See attached distribution list

Letter or Memorandum to J. Ziegler from J. Schlueter dated: October 21, 2003
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The NRC expects that results of studies conducted between now and the submittal of the license application will provide additional support needed to justify the suppression of some mineral reactions.

If you have any questions regarding this matter, please contact Gregory Hatchett at 301-415-3315 or by e-mail to GXH@nrc.gov.

Sincerely,

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Janet R. Schlueter, Chief
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REVIEW BY THE OFFICE OF NUCLEAR MATERIAL SAFETY AND SAFEGUARDS OF THE DEPARTMENT OF ENERGY'S AGREEMENT RESPONSE RELATED TO THE PROPOSED GEOLOGIC REPOSITORY AT YUCCA MOUNTAIN, NEVADA "EVOLUTION OF THE NEAR-FIELD ENVIRONMENT" (ENFE) KEY TECHNICAL ISSUE AGREEMENTS 2.07 AND 2.08 [PROJECT NO. WM-0011]

1.0 INTRODUCTION

The U.S. Nuclear Regulatory Commission's (NRC) issue resolution goal during the pre-licensing period is to assure that the U.S. Department of Energy (DOE) has assembled enough information on a given issue for NRC to accept a license application for review. Resolution by the NRC staff during pre-licensing does not prevent anyone from raising any issue for NRC consideration during the licensing proceedings. Also, and just as important, resolution of an issue by NRC during pre-licensing does not prejudge the NRC staff evaluation of the issue during the licensing review. Issues are resolved by NRC staff during pre-licensing when the staff has no further questions or comments about how DOE is addressing an issue. Pertinent new information could raise new questions or comments on a previously resolved issue.

By letter dated September 27, 2002, DOE submitted a report titled "Agreements ENFE 2.07 and 2.08" (Adams Accession No. ML022800254) to satisfy the informational needs of the two key technical issue agreements pertaining to ENFE Subissue 2, "Effects of Coupled Thermal-Hydrologic-Chemical Processes on the Waste Package Chemical Environment." The subissue and the two agreements are considered important to repository performance because they deal with chemical and physical conditions that may affect drip shield and waste package corrosion. NRC preliminary analysis suggests that ENFE Agreement 2.07 has high risk significance and ENFE Agreement 2.08 has low risk significance (Travers to Chairman Diaz et al. 2003, (ADAMS Accession Nos. ML031320684, ML031320709, ML031320701, and ML031550762). In this memorandum, the staff described its basis for risk-ranking the key technical issue (KTI) agreements. Generally, high risk significance during post closure is associated with features, events, and processes that could affect a large number of waste packages or significantly affect the release from the waste package or affect the transport of radionuclides through the geosphere. Agreements that are necessary to provide the baseline information on the site were considered to have low risk significance relative to repository performance.

The DOE response to ENFE Agreements 2.07 and 2.08 provided some information concerning the specific coupling relationships that are included in or excluded from the total system performance assessment (TSPA) modeling, and included the technical basis for the suppression of minerals in chemical equilibrium modeling. In the response, DOE states that it has satisfied the NRC's information needs regarding both agreements and that both agreements should be considered closed.

ENCLOSURE

2.0 WORDING OF THE AGREEMENT(S)

ENFE KTI Agreements 2.07 and 2.08 are identified in a NRC staff letter dated January 26, 2001, which summarized the Evolution of the Near-Field Environment Technical Exchange and Management Meeting held on January 9 through 12 (ADAMS Accession Nos. ML010300165 and ML010290320). The wording of the agreements is as follows:

"ENFE.2.07: The DOE will identify specific coupling relationships that are included and excluded from Total System Performance Assessment, including Onsager couples, and give the technical basis for inclusion and exclusion. This information will be documented in a revision to the Engineered Barrier System Degradation, Flow, and Transport Process Model Report (TDR–EBS–MD–000006), expected to be available by September 2001."

"ENFE.2.08: DOE will provide additional technical basis for suppression of individual minerals predicted by equilibrium models, in a revision to the Engineered Barrier System: Physical and Chemical Environment Model Analysis Model Report (ANL–EBS–MD–000033)."

The technical information provided by DOE in response to these agreements and NRC's evaluation is given below.

3.0 TECHNICAL INFORMATION PROVIDED IN THE DOE AGREEMENT RESPONSE

ENFE Agreement 2.07 was developed to provide a preliminary indication that DOE has considered all important coupled processes when predicting the chemical and physical environments important to drip shield and waste package corrosion. ENFE Agreement 2.08 was developed to provide a preliminary indication that DOE understood the mineralogical system at Yucca Mountain well enough to adequately model expected changes to that system.

The DOE report provided information to address ENFE Agreements 2.07 and 2.08, but not in the form prescribed by the original ENFE 2.07 and 2.08 agreements. ENFE Agreement 2.07 stated that DOE would document the requested information in a "revision to the Engineered Barrier System Degradation, Flow, and Transport Process Model Report (TDR–EBS–MD–000006), expected to be available by September 2001." Likewise, the original ENFE 2.08 agreement stated that DOE would provide the requested information in "a revision to the Engineered Barrier System: Physical and Chemical Environment Model Analysis Model Report (ANL–EBS–MD–000033), expected to be available in fiscal year 2002." However, DOE programmatic changes resulted in the rescheduling of these items.

3.1 ENFE Agreement 2.07

The DOE response identified and discussed specific information related to coupling relationships that are included in or excluded from the TSPA model (including Onsager couplings). Coupled processes in this case are processes which depend on two or more physical and chemical variables simultaneously interacting to produce a result. The coupled processes discussed in DOE's response involved transport of chemicals, specifically radionuclides, which can affect dose calculations. DOE identifies the locations of liquid diffusive and advective transport processes in the Engineered Barrier System (EBS). DOE also, mentions seepage, chemical interactions with ground support materials and gas, microbial

activity, evaporation, salt precipitation, corrosion, condensation, imbibition, diffusion, advection, dissolution and colloidal stability (dispersion/flocculation), and colloidal transport. The response does not delve into these processes in detail. Instead DOE evaluates the affect of the off-diagonal Onsager couples on radionuclide transport. Using a mixture of preliminary site-specific and generic data, DOE concludes that the off-diagonal couplings are unimportant to radionuclide transport.

3.2 ENFE Agreement 2.08

The DOE response presented additional technical bases for suppression of individual minerals predicted by equilibrium models. The DOE response describes the criteria for determining if mineral suppression is appropriate, a list of minerals included and excluded from current engineered barrier system models, and technical bases for suppressed and unsuppressed minerals. Specifically, the response presented the technical basis for the suppression of 12 mineral phases and the rationale (based on literature sources) for the inclusion, to date, of 26 minerals in the model output from calculations using Version 8.0 of EQ3/6 and a new Pitzer database. DOE noted that these technical bases will be finalized and incorporated into appropriate model reports and that if additional suppressions or inclusions are necessary in future model calculations, the technical basis for including or excluding each of these additional minerals will be documented.

4.0 NRC EVALUATION AND COMMENT

ENFE Agreements 2.07 and 2.08 are pertinent to understanding important facets of ENFE Subissue 2, “Effects of Coupled Thermal-Hydrologic-Chemical Processes on the Waste Package Chemical Environment,” and have been reviewed by NRC staff in that context.

4.1 ENFE Agreement 2.07

In the letter dated September 27, 2002, DOE mentions that the contents of its ENFE 2.07 response were discussed on September 11 and September 26,2002, with NRC, who indicated that ENFE 2.07 should also address the impacts of coupled processes on waste package corrosion. DOE asserts that the impacts of coupled processes, such as corrosion on the waste package, should not be included in the response to ENFE Agreement 2.07. Instead, DOE maintains that the scope of ENFE 2.07 should be restricted to the effects of coupled processes on the waste package chemical environment, as originally intended. NRC staff agrees with DOE that the scope of the response to ENFE Agreement 2.07 should be focused on the effects of coupled processes on the waste package chemical environment. However, the material scientists use this chemical/physical environment in tests and models to measure and estimate mechanisms and rates of waste package corrosion.

NRC’s intent in this agreement was to have DOE identify and consider potential processes and chemical conditions, including relatively exotic ones, that may exist over a range of scales (from crevice size to drift size) in a chemical system (the repository) that is out of equilibrium due to the introduction of heat and engineered materials, and provide a technical basis for including in or excluding the processes and chemical conditions from TSPA. For example, with the apparent lifetimes of waste canisters extending for over 10,000 years, NRC wanted to be reasonably assured that no credible near-field process or chemical condition omitted from

testing or modeling could lead to significantly shorter waste canister lifetimes. However, DOE's response provided no information on chemical couples that were included or excluded. This information is needed to determine the range of credible chemical environments that are important to drip shield and waste package corrosion.

NRC found that DOE's response to agreement ENFE 2.07 focused too narrowly on transport processes in the engineered barrier system. Section 3 of the DOE letter report, "Information to Satisfy Key Technical Issue Agreements" states that "the coupled processes to consider in the Total System Performance Assessment involve transport of chemicals, specifically radionuclides which can affect dose calculations." Detailed analyses of radionuclide transport processes are most appropriately treated in the context of ENFE Subissue 4, "The Effects of Coupled Thermal-Hydrologic Chemical Processes on Radionuclide Transport in the Near Field." DOE has agreed to provide related analyses of coupled thermal-hydrological-chemical effects on radionuclide transport properties within the context of agreement ENFE 4.03.

While it is appropriate for DOE to consider changes in the near-field chemical environment due to chemical transport, DOE should not focus solely on the effects of these changes on radionuclide transport. These changes can occur before waste package failure and thus do not necessarily involve radionuclides. Further, the report lists many processes expected to occur in the near-field environment, and it is unclear to NRC how some of those processes are coupled. For example, condensation is mentioned as a process that can occur, under appropriate conditions, beneath the drip shield and on the waste canister. Current evidence in the enhanced characterization of the repository block suggests that condensation may play a role in the redistribution of water in the repository. The NRC found that this evidence leads to more uncertainties and that the following questions need to be considered:

- How much water can condense on a waste package?
- How does the amount of condensed water compare with the amount of seepage water?
- Will the repository be unstable with regard to temperature distributions?

Additionally, could the dust/aerosol collected from the current-day exploratory studies facilities be compositionally different from dust in a heated repository? The evaporation of groundwater may increase the proportion of salts in the dust/aerosol. These salts could settle on the waste package and absorb moisture even though the drip shield is intact. Moreover, processes in the near field occur in a high gamma flux field. DOE states that due consideration was given to this aspect of the environment. The report concludes that the near-field environment remains unchanged. The report provides no details to support this conclusion, but it is the NRC's understanding that in the past DOE has noted that a high gamma flux field can produce hydrogen peroxide in an aqueous environment. Consequently, some corrosion experiments were performed in which hydrogen peroxide was added, to no effect. However, the staff is uncertain of the effects that may occur if deliquescent minerals coat the waste package when there is a high gamma field flux. In addition, Green et al. (1987) calculate that Compton scattering of electrons could produce an electric field around a waste canister. The corrosion of the waste package is electrochemical in nature. The DOE response discounts electrochemical processes that only affect radionuclide transport.

Finally, given its complexity, the staff is unclear how DOE will bound the chemical compositions and physical conditions that might be predicted in the repository. For example, the situation

where water drips on a sloped heated surface (e.g. drip shield or waste package) resulting in precipitation of various solid phases along the water droplet's flow path, ending where the droplet evaporates to dryness, needs to be considered. Work by John Walton and Drew Hall presented at the Advisory Committee on Nuclear Waste meeting on March 26, 2003, described physical separation process during evaporation as common in arid environments. Furthermore, Drever (1988) describes the added complexity of cyclic wetting and drying. The relative rates of the various processes, the spatial and temporal distribution of those processes and the range of masses of materials that might be involved in those processes, all contribute to the complexity of characterizing the near-field.

4.1.1 Need for Additional Information

The NRC's intent in this agreement was to obtain reasonable assurance that no near-field processes or chemistries omitted from testing or modeling could lead to significantly shorter waste canister lifetimes. DOE's response to ENFE Agreement 2.07 lacks an analysis of the role of chemical reactions in drip shield and waste package corrosion. While it is appropriate for DOE to consider changes in chemical environment due to chemical transport, a variety of solid phase-gas-water interactions might occur in concert with changing thermal-hydrological conditions and alter the waste package chemical environment. NRC expects DOE to provide technical bases for electrochemical and microbial processes, rates of reactions, mass balance, probability considerations, and temporal and spatial distributions of processes and conditions on scales important to performance. In addition, in addressing chemical couples that were included or excluded from the determination of important near-field chemical environments, DOE should discuss the effects of coupled near-field processes and conditions on waste package and drip shield corrosion.

Additionally, NRC notes that a May 13, 2003, presentation to the Nuclear Waste Technical Review Board by Mark Peters of Los Alamos National Laboratory described the character of the in-drift environment. Among other topics, the presentation covered the three abstracted temperature regions of the drift environment, the chemical evolution of the drift environment, the chemical divide theory, observed water chemistries, in-drift water chemistry modeling and validation, and the investigation of deliquescence during dryout temperature. This presentation provided clarification of DOE's method of characterizing the near-field environment. The environment is modeled using TOUGHREACT and EQ3/6. The modeling produced a range of "bounding" in-drift water compositions, which DOE consolidated into 11 bins. DOE should consider addressing ENFE 2.07 in the context of providing the technical basis for the consolidation or establishment of the 11 bins. This approach would need to address the couples considered, the range of chemistries considered, the rationale for including or excluding couples and chemistries, and the limitations of any codes used to develop the bins.

4.2 ENFE Agreement 2.08

DOE's response to agreement ENFE 2.08 states that the DOE provided additional technical bases for suppression of individual minerals predicted by equilibrium models. The DOE response describes the criteria for determining if mineral suppression is appropriate, a list of minerals included and excluded from current engineered barrier system models, and technical bases for suppressed and unsuppressed minerals.

Section 3.3.2 of the DOE response to the ENFE 2.08 agreement states that the report presents "a discussion of the appropriate process that governs the selection and suppression of mineral phases in geochemical models." The NRC agrees that the report satisfies the ENFE 2.08 agreement to provide the requested information about mineral suppression from equilibrium models. NRC found that the report did not present a comprehensive discussion of the appropriate process for selecting mineral phases in geochemical models. However, as suggested in Section 3.3.2 of the report, additional criteria are likely needed to develop robust conceptual models. The relative significance of uncertainties in the thermodynamic properties of individual minerals, for example, could dominate predictions of the evolution of the waste package chemical environment, and should be considered in the development of conceptual models. At this time NRC has no further questions regarding mineral suppression from equilibrium models.

5.0 SUMMARY

In summary, ENFE Agreements 2.07 and 2.08 relate to understanding important facets of ENFE Subissue 2, "Effects of Coupled Thermal-Hydrologic-Chemical Processes on the Waste Package Chemical Environment," and have been reviewed by NRC in that context. This subissue is very important with regard to whether the model abstractions for the near-field environment and the abstractions for engineered barrier integrity used in performance assessment are adequately supported and whether the results are adequate for evaluating the inclusion or exclusion of related features, events, and processes. NRC reviewed the DOE KTI agreement responses in the report to determine whether any aspects of the agreements were excluded from the response. In addition, NRC, with assistance from the Center for Nuclear Waste Regulation, performed an independent assessment to determine whether the information provided would support submission of a potential license application for a geologic repository.

With regard to ENFE Agreement 2.07, DOE agreed to identify specific coupling relationships that are included or excluded from the DOE TSPA model and to give the technical basis for their inclusion or exclusion. DOE response focused on radionuclide transport processes in the engineered barrier system, rather than on couplings needed to define the chemical environment of the drip shield and waste package. DOE's response lacks an analysis of the role of chemical reactions in determining the waste package chemical environment. Consequently, the status of ENFE Agreement 2.07 is considered "needing additional information." To complete ENFE Agreement 2.07, DOE should expand its response to identify the chemical couplings used to define the chemical conditions that will likely contact the drip shield and waste packages. DOE should also document the technical bases for including or excluding any coupled process that might alter the chemical environment and adversely influence repository performance.

With regard to ENFE Agreement 2.08, DOE agreed to provide a stronger technical basis for the suppression of individual minerals predicted by equilibrium models. DOE response presented an adequate summary of the criteria used to determine if mineral suppression is appropriate, a list of minerals included or excluded from current engineered barrier system models, and the technical bases for suppressed and unsuppressed minerals. While NRC considers that the report satisfies ENFE Agreement 2.08, information from ongoing tests and analyses, including comparisons of results to conceptual models and model validation test cases, will be needed to support a license application. Nevertheless, the status of ENFE Agreement 2.08 is considered

"complete," and NRC has no further questions regarding mineral suppression from equilibrium models at this time.

6.0 STATUS OF THE AGREEMENTS

6.1 ENFE 2.07

Based upon the above review, NRC does not agree with DOE that the information provided satisfies the intent of the agreement. Therefore, NRC considers that agreement ENFE 2.07 needs additional information. The additional information sought by NRC is described in Section 4.1.1 of this evaluation.

6.2 ENFE 2.08

Notwithstanding new information that could raise new questions or comments concerning ENFE Agreement 2.08, based upon the above review, NRC agrees with DOE that the information provided satisfies the intent of the agreement. Therefore, the NRC considers that ENFE Agreement 2.08 is complete, and NRC has no further questions regarding mineral suppression from equilibrium models at this time.

7.0 REFERENCES

Drever, J.I., *The Geochemistry of Natural Waters*, Prentice Hall, Englewood Cliffs, New Jersey 07632, 1988.

Green, R.T., W.L. Fillippone, and D.D. Evans, "Effect of Electric Fields on Vapor Transport Near a High-Level Waste Canister," presentation at the International Symposium on Coupled Processes Affecting the Performance of a Nuclear Waste Repository in *Coupled Processes Associated with Nuclear Waste Repositories*, Academic Press, Inc., 1987.

Reamer, C.W., NRC, "U.S. Nuclear Regulatory Commission/U.S.Department of Energy Technical Exchange and Management Meeting on Evolution of the Near-Field Environment January 9-11, 2001," letter (January 26, 2001) to S. Brocoum, DOE, Washington, DC, NRC 2001 (ADAMS Accession Nos. ML010300165 and ML010290320).

Travers, W.D., "Final Staff Response to March 19, 2003 Requirements Memorandum on the Waste Arena Briefing-M030303A," Memorandum to Chairman Diaz, Commissioner Dicus, Commissioner McGaffigan, and Commissioner Merrifield, June 5, 2003 (ADAMS Accession Nos. ML031320684, ML031320709, ML031320701, and ML031550762).

Ziegler, J.D., DOE, "Transmittal of Report Addressing Key Technical Issue (KTI) Items Evolution of the Near-Field Environment (ENFE) 2.07 And 2.08" letter to J. Schlueter, NRC, September 27, 2002 (ADAMS Accession No. ML022800254).