UCRL- 90044
PREPRINT

CONF-831217--1

UCRL--90044

DE84 003894

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Civilian Radioactive Waste Management
Information Meeting
Washington, D. C.
December 12-15, 1983

November 1983

Lawrence Livermore National Laboratory

WASTE PACKAGE FOR A REPOSITORY LOCATED IN TUFF

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INTRODUCTION

The Nevada Nuclear Waste Storage Investigations (NNWSI) project is evaluating a potential repository system to be located on or adjacent to the Department of Energy Test Site in Nye County, Nevada. The particular site which is being investigated is a thick sequence of volcanic tuff beds which form Yucca Mountain on the southwestern portion of the Test Site and adjoining Federal land. A distinguishing feature of this site is a very deep water table which permits consideration of a densely welded devitirified tuff horizon, the Topopah Spring member, which is located above the water table. Among the advantages of siting a repository in the unsaturated zone are very limited availability of water due to the low influx rate in this arid region; low temperatures at which liquid water can be present; and absence of significant hydrostatic stress on the waste packages.

Within the NNWSI project, the Lawrence Livermore National Laboratory (LLNL) is assigned responsibility for the development and qualification of designs for waste packages suitable for emplacement in a high level waste repository at Yucca Mountain. The topics which must be addressed to establish the viability of waste package designs can be logically structured within four broad categories, recognizing that each interacts with the others. These include the package environment, the materials to be utilized, the package design, and analysis and test of its performance. These areas provide the basis for the

*Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract number W-7405-ENG-48.

organization of the LLNL waste package effort, and this discussion will address each of them. The major accomplishments during the past year and the planned activities for the coming year will be described.

PACKAGE ENVIRONMENT

A thorough understanding of the time and temperature dependent conditions to be anticipated in the environment immediately adjacent to the waste packages is essential. The important environmental parameters to be evaluated include the geochemical properties of the tuff and vadose water, the variation in thermal and thermomechanical properties of the tuff, and the response of the interstitial water to the thermal energy deposited in the very-near-field by the decay energy of the waste forms.

During the past year, we have conducted a number of tuff-water interaction tests utilizing both crushed and intact Topopah Spring tuff samples in contact with water from a nearby well (J-13) which is completed in the Topopah Spring member where it occurs below the water table. These rock samples have been obtained from outcrops, cores from near surface exposures, and from deep borings at several locations near the candidate repository block. The test matrices have included a range of temperatures, rock surface area to water volume ratios, and test durations. The details of these test results are reported elsewhere (Knauss et al 1983). In general, the tests indicate that changes in water chemistry in the vicinity of the waste packages will be minimal; no significant alteration of the primary mineral phases of the rock are to be expected; and the near-neutral pH of the system should be maintained.

During the next year some of these tests will be continued and extended to include metallic components and corrosion products. Geochemical modeling of the rock-water systems will continue in order to provide a basis for extending these test results to the very long times of interest in waste package performance prediction.

A preliminary test, using an instrumented intact core sample, has been initiated to evaluate the dehydration and resaturation of the very-near-field rock in response to the thermal cycle imposed by the decay heat. Migration of the moisture front has been monitored. The test is being extended to include a core sample which contains a natural fracture. The candidate repository horizon is expected to be extensively fractured and therefore this test is more representative of the anticipated conditions. During the next year we plan to continue this important activity and scale the laboratory test up to permit measurements which are less influenced by local boundary effects.

MATERIALS DEVELOPMENT AND TESTING

The materials to be employed in high-level waste packages are required to function in a predictable manner for time periods much longer than are normally expected in more conventional engineered structures. These periods - hundreds to thousands of years - are not amenable to real time testing and therefore require development of short-term test data and relationships which will permit extrapolation to very long times. Three major components form the waste package engineered barrier subsystem of a repository: the waste forms, which will be expected to provide the primary control of radionuclide release rates in the isolation (post-containment) phase; the metallic containment barrier(s), which will be relied upon to provide the "substantially complete" (NRC 1983) containment of the waste form during the first several hundred years; and any other materials, such as packing materials (backfill), emplacement hole liners, etc. which may be required by the overall package and repository design.

Waste Forms

The two high level waste forms which are expected to be disposed of in a geologic repository are unreprocessed spent fuel and reprocessing wastes bound in a borosilicate glass matrix. Tests directed toward qualifying the release rates of both these waste forms were initiated during the past year, and will continue for several more years.

Fuel from commercial power reactors is encapsulated in zirconium-alloy cladding which, if intact, can provide a very corrosion-resistant barrier to release of radionuclides. Projections of the fraction of the spent fuel inventory which will contain significant cladding defects (Woodley 1983) indicate that this potential barrier may be very effective. Defects, such as pinholes or slits, will occur in some fuel rods, however. Tests to quantify the release rate of nuclides from defected fuel have been initiated at the Westinghouse-Hanford Co. laboratories. Artificial defects, both laser-generated punctures and sawed slits, have been induced in previously intact spent fuel rod sections and release rates compared with both intact rods and with bare unclad fuel materials. Preliminary test results indicate that reductions of several orders of magnitude in release rates can be expected from fuel with defected cladding compared with bare fuel pellets.

These spent fuel tests, augmented by testing to quantify cladding corrosion rates, will be continued.

Tests on borosilicate glass waste forms were also initiated during the past year. Materials which approximate both commercial and defense program waste forms are being tested. The initial parametric tests on CHLW glass employed the PNL 76-68 glass formulation with simulated fission products and uranium. The parameters investigated included waste form surface area to water volume, temperature, and the effects of tuff and stainless steel on the system. The initial results, when scaled to a full waste package imply an annual release rate of less than 10^{-5} (Oversby 1984).

Initial tests with DHLW glass are being conducted by the Savannah River Laboratory using J-13 water and tuff reaction vessels furnished by LLNL. The tests are a modified MCC-1 type configuration. Preliminary results from these tests also are encouraging and imply that this waste form, if emplaced in a tuff repository, will perform in a manner consistent with the regulatory requirements.

It must be emphasized that all of the waste form testing now in progress has been initiated recently and the initial results are far from conclusive. However, it should also be remembered that all of these tests are being conducted under conditions where the amount of water available for dissolving the waste form is large compared to the anticipated conditions in a repository located in the unsaturated zone at Yucca Mountain.

A test configuration which more nearly approximates the anticipated conditions is under development at Argonne National Laboratory. This test development was initiated earlier this year and it is expected that the initial tests, which will be primarily for the purpose of establishing test techniques and reproducibility of the data, will get underway during the coming year.

Metallic Barriers

The primary containment of the waste forms is provided by a metallic barrier, variously referred to as a canister, overpack, or container. This barrier is expected to perform for a period of up to 1000 years and provide "substantially complete" containment of the waste form. In addition, this container is expected to protect the repository from contamination during the operational period, including waste handling activities associated with emplacement and possible retrieval for a period of up to fifty years after emplacement.

After reviewing the preliminary package environment information, a list of 17 candidate metals which could potentially meet the requirements was developed. This list was screened with a semi-quantitative ranking scheme to reduce the list to four metals for further consideration (Russell et al 1983). These are all iron-base to nickel-base alloys with an austenitic structure. The reference alloy which was selected is AISI 304L stainless steel. The alternatives, which were selected based on increased resistance to specific localized corrosion mechanisms, include the molybdenum or titanium stabilized alloys 316L, 321, and Incoloy 825.

An extensive testing program has been initiated to evaluate the performance of these alloys in the appropriate tuff-conditioned water and saturated steam environments. The uniform corrosion rate measured for 304L is less than 0.2 micrometers/year at temperatures in the range of 50-150^{o}C. However, these materials are known to be susceptible to localized and stress-assisted attack and are therefore being evaluated in more hostile environments and stress conditions, including tests in the presence of an intense ionizing radiation field.

A significant simplification of the packages for the glass waste forms could be achieved if the pour canisters could be used directly, i.e., without an overpack. Two factors, residual stress due to differential thermal expansion and time-temperature sensitization, are of concern in this regard. We are investigating these with both the testing program and supporting calculational activities.

Electrochemical polarization tests of the candidate materials complement the corrosion coupon testing. This technique provides a method for evaluating the corrosion potential of solutions with higher ionic concentrations than those found in J-13 water and for testing in simulated radiolysis conditions by addition of species such as hydrogen peroxide and nitrate ion to the test solutions.

Work during the next year will continue to focus on localized and stress-assisted corrosion mechanisms to develop the data base from which the final selection of materials and fabrication processes will be specified, and on which the models for extrapolation to long term corrosion behavior will be based.

Packing Materials

A variety of functions have been described for packing materials (formerly called backfill) which might be emplaced around waste package containers. These include reducing access of water to the container or waste form, modification of groundwater chemistry, limiting stress on or cushioning the package, and retarding the movement of nuclides released from the waste form. The materials which are capable of performing these functions have the common drawback that their thermal conductivity is lower than the host rock and therefore the temperatures of all components which they surround are raised. This adverse characteristic may require reduced waste loading in each package and therefore more packages with attendant increased costs and complexity.

In the unsaturated zone tuff environment, preliminary assessments indicate that a packing material should not be necessary around the borosilicate glass waste forms. In the case of spent fuel the need for packing material will depend on the effectiveness of the fuel cladding in reducing the nuclide release rate, and the dominant water flow mechanism in the near-field rock mass.

We have therefore initiated a low level investigation of the feasibility of fabricating preformed crushed tuff materials which might be preemplaced in disposal holes. Small samples have been pressed using both crushed tuff alone and with a low percentage addition of smectite clay as a binder material. The thermal conductivity of these samples has been measured and is about 40% that of dehydrated host rock.

During the next year, additional samples will be fabricated and tested for permeability at appropriate temperatures and thermal gradients.

PACKAGE DESIGN

As has been implied in the previous discussions, a number of uncertainties are present with respect to the performance of package components. These include spent fuel cladding and glass waste form pour canisters as barriers, and packing material effectiveness. The water influx rates, dominant flow mechanisms, and very-near-field response of water to the thermal cycle are not well known. In addition, the Repository Design group at Sandia National Laboratory is continuing to evaluate the relative merits of long multi-package horizontal borehole versus single package vertical emplacement geometries.

Given these uncertainties and alternatives, we have undertaken to develop and analyze a set of design concepts during the past year in order to focus the emphasis of the balance of the development activities and provide a range of package alternatives for use in repository conceptual design studies (Gregg and O'Neal 1983).

The conceptual package design set includes alternatives to accommodate either vertical or horizontal emplacement geometry; either non-overpacked or overpacked glass pour canisters; and spent fuel canisters emplaced either with or without a preformed tuff packing material. Thus a total of eight design concepts are currently being considered, with several dimensional variations to accommodate differences in waste form such as CHLW or DHLW glasses, and PWR or BWR spent fuel, either as intact assemblies, boxed preconsolidated rods, or rods consolidated at the repository.

Extensive thermal analyses have been performed on all of these design options to insure that the packages conform to peak temperature limitations imposed by the waste forms. Structural analyses have been done to check for adequate safety margins on normal handling loads as well as selected off-normal conditions such as fire and drop loads. For the spent fuel designs, criticality calculations have also been made for selected credible geometries. Economic considerations have been evaluated for the selected design concepts.

During the next year all of the design and analysis activities described will be documented in a Conceptual Design Report. No major additional package design activity is planned until some of the uncertainties enumerated earlier are resolved.

PERFORMANCE ANALYSIS AND TESTING

The primary objective of performance analysis is to provide a quantitative prediction of long term waste package performance. Two measures, which follow from the containment and isolation functions of the waste package subsystem, must be calculated to quantify this performance. They are the radionuclide release initiation time and the subsequent release rate. These calculations are necessary to provide feedback to design optimization studies; and define a source term for overall repository system performance assessment. Our approach to this analysis is to adapt a 1-D waste package subsystem model to track the response of individual package components to potential degradation mechanisms as a function of time. Simple analytic equations and an extensive, empirical data base are used to represent several degradation mechanisms, including: thermal, mechanical, radiation, corrosion and release. Inputs to the analysis require a definition of: 1) the emplacement environment, from site geotechnical investigations, 2) the waste package design, from material and design selection studies, and 3) the material interaction properties, from the results of waste form-metal-host rock-groundwater geochemical modeling and experimental efforts. From this information the subsystem model first calculates the physical state (i.e., temperature, stress, radiation flux and fluid flow) for each component. The results of geochemical analyses, modeled externally, predict the metal corrosion and/or waste form release rates appropriate to these conditions. The subsystem model must then integrate the results of these physical and geochemical processes with the waste package geometry to calculate barrier integrity (i.e., containment) and radionuclide release profiles (i.e., isolation) as a function of time.

Based on a preliminary review, it appears that the WAPPA code (INTERA 1983) can be modified to provide performance analyses of a waste package emplaced in unsaturated tuff. In order to identify the limitations of the existing code and to plan the necessary modifications, we have initiated a detailed evaluation of each degradation process model. This evaluation consists of review of the theoretical model, verification of the algorithm coding, and assembly of a data base. We have completed a review of the theoretical basis for each process model and have begun the verification and data assembly steps. During this coming year we plan to complete these steps for the thermal and mechanical models and assemble initial data bases for metal corrosion and waste form release rates. Modification efforts will be directed toward the development of a system level model for transport in the unsaturated zone.

The second component of the performance analysis and testing activity is directed toward planning a major test of a set of prototype waste packages in the candidate repository horizon as a part of the exploratory shaft in-situ testing. This test will be configured to provide site-specific data on the dehydration-resaturation phenomena for use in analyses of package performance. In addition, extensive instrumentation will be deployed to monitor the other important package environment parameters to compare with laboratory-scale measurements made previously on core samples.

During the past year the major effort in this area has been development of a conceptual test plan and participation in a project-wide working group to draft a comprehensive plan for exploratory shaft testing.

In the next year details of the test will continue to be developed so that final test geometries can be selected and instrumentation procurement can be initiated in 1985.

SUMMARY

The development of waste packages for emplacement in a tuff repository has been proceeding during the past year on a broad front. Experimental work has been focused on determination of important package environment parameters and testing the response of waste forms and package materials to the anticipated environment. Conceptual designs have been selected with alternatives to accommodate present uncertainties in the environment and material performance. Computational capabilities are being adapted to provide analyses of anticipated package performance and plans are being developed for in-situ testing. The waste package activities have been integrated into the overall NNWSI project to assure timely completion consistent with the statutory and regulatory requirements leading to repository site selection around the end of the decade.

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