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Development of Waste Packages for Tuff

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DEVELOPMENT OF WASTE PACKAGES FOR TUFF

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The objective of this program is to develop nuclear waste packages that meet NRC requirements for a licensed repository in NTS tuff. The program was transferred to LLNL in June 1981 after an initial period at Sandia and Los Alamos. Originally the task of this program was to integrate site data relevant to the waste package and provide input to others on site-specific needs for waste package designs and materials development. Responsibility for approving such designs was also part of the program. In addition, the program was given a mandate to do limited work on materials to develop and maintain expertise in this area.

In May 1982 DOE transferred total responsibility for design and materials development to the individual projects, to be more fully funded in FY83. Much of the work reported here, therefore, was done during the "integration" phase of our program. Because of the current philosophy for redundant engineered barriers, the goal has been to approach long-term containment of radionuclides (hundreds to 1000 years) followed by a longer term (thousands of years) of very low release rates. The approach has been to study candidate metals and select those having appropriately low corrosion rates for the containment requirement. For the low release rate period, the primary retention characteristics of the waste form (processed high level waste or spent fuel) is the primary barrier; a buffer/backfill with sorbing characteristics would be used only if necessary to meet release requirements. The rationale is that effective backfills typically act as thermal insulators (especially when dehydrated) compared to the host rock and raise temperatures of all components they surround, thus putting more stringent demands on these components.

A unique characteristic of the NTS repository site is the depth of the water table, allowing the option of emplacement above the water table. Recently the working assumption has been made that the repository will be located above the water table. Only if the two candidate unsaturated horizons prove unsuitable would the saturated zone come into primary consideration.

The original design effort and materials test environment were based on conservative assumptions, viz., oxidizing water deeply below the water table. These conditions require heavier metal barriers to resist the hydrostatic pressures than would be required for placement in the unsaturated zone.

Selected accomplishments for FY82 are outlined in Table 1. Discussion of some of these topics follows.

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Table 1
FY82 SELECTED ACCOMPLISHMENTS

- Selection, collection of rock, and characterization of suitable outcrops (for lab experiments)
- Rock-water interactions (Bullfrog Tuff)
- Corrosion tests of ferrous metals
- Thermal modeling of waste package in host rock
- Preliminary fabrication tests of alternate backfills (crushed tuff)
- Reviewed Westinghouse conceptual waste package designs for tuff and began modification for unsaturated zone.
- Waste Package Codes (BARIER and WAPPA) now running on our computer

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Studies of rock-water interactions are necessary: (1) to determine the probable fluid environment of the waste package and so permit us to set conditions for materials testing; and (2) backfills probably will contain some host rock, and we need to determine its stability and behavior in groundwater above ambient temperatures to steam temperatures (e.g., 100-250°C).

Initial tests at 150°C at LLNL have been completed using ground Bullfrog tuff from a suitably characterized outcrop and a reference water designated at J-13 (Table 2). In 4 + days steady state is approached, as confirmed by tests up to 48 days. The results show that Si increases, approaching its solubility limit (from 31 to 140 ppm). Modest decreases in Na^+ and decreases in Ca^{++} occur, with little change in K^+ at steady state. The pH increases from 7 to 8-8.5. Thus, the changes are such as to have little effect on the waste package components. Work is continuing to confirm the behavior on other tuffs in the unsaturated zone.

An 8-week screening test has been completed on ferrous metals at 80°C under a range of oxidizing conditions. The temperature was chosen as an anticipated worst case between increasing reaction rates and decreasing oxygen solubility with increasing temperature. The various gases were continuously bubbled through deionized water in the presence of ground tuff. As Table 3 shows, none of the metals corroded badly in nitrogen-saturated water. Mild steel and a low Cr-Mo alloy steel have very high corrosion rates under oxidizing conditions, while the 9 Cr - 1 Mo alloy shows very low corrosion rates. It is recognized that other studies are needed for this alloy, including the effects of stress, radiation, and weldment response to corrosion before it can be proposed as a canister-overpack.

A critical task is thermal and radionuclide migration modeling within a waste package. To date, thermal analyses of various waste package configurations have been done below the water table. Figure 1 shows the results of one such calculation. It appears that the use of bentonite would be questionable under these conditions because of its relatively low thermal conductivity when dehydrated (0.6-0.7 w/m°C) and thermal instability at high temperatures (>150°C under hydrothermal conditions and about 300°C+ under dry conditions).

Table 2

INTERACTION OF TUFF WITH GROUNDWATER

To determine change in groundwater after reaction with hot rock

150°C, 4 + days

Results to date, ppm

Ref. Water		After reaction
Si	31	140
Na	48	100
Ca	12	5
K	5	6-14
Al	0.01	0.2
pH	7.0	8.5

Table 3

CORROSION EXPERIMENT - FERROUS METALS

Conditions

Water + tuff

80°C, 8 weeks

	Corrosion rate, mm/yr		
Atmosphere	Mild carbon steel	Fe-2.5 Cr-1 Mo	Fe-9 Cr - 1 Mo
Air	1.1	0.5	0.002
O_2	0.7	0.7	0.007
N_2	0.002	0.001	$<10^{-4}$

Conclusions to date

Oxidizing conditions: severe on steel and low Cr alloy

Reducing conditions : little effect on all metals

9 Cr-1 Mo alloy shows excellent behavior

Backfills in the saturated zone typically utilize compressed bentonite, having low permeability and water-absorbing properties. For a backfill in the unsaturated zone, it may be desirable to use a coarsely granular backfill instead, avoiding attraction of water to the package by capillarity and encouraging drainage of small or intermittent water flows away from the waste package.

In FY83 materials work will be directed to waste packages in the unsaturated zone. Under these conditions the presence of steam and air during the dehydration period is anticipated, followed by possible liquid water cycles alternating with moisture-saturated air. Materials testing conditions will be selected to simulate this varied environment. A partial listing of tasks planned for FY83 is shown in Table 4. This work leads to the selection of the waste package conceptual design and provides a focus necessary for initiating preliminary design in FY84.

Table 4
WORK PLANNED FOR FY83 (PARTIAL LISTING)

1. Leaching experiments in glass
2. Initial studies on spent fuel leaching
3. Metal corrosion tests of ferrous metals and titanium alloys
4. Backfills -- properties and hydrothermal stability
5. Analysis of conceptual designs and selection of reference conceptual design
6. Evaluation and modification of Waste Package modeling codes.

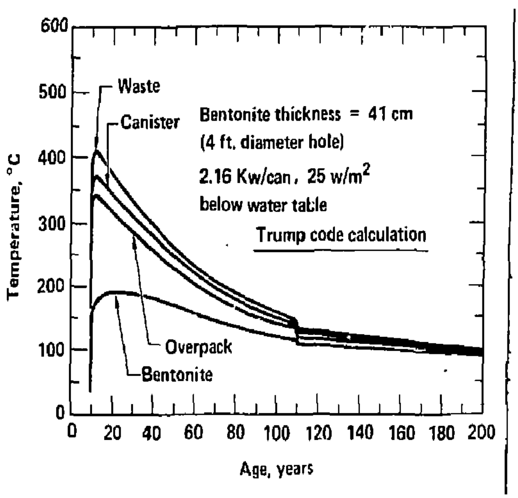

Fig. 1 - Typical Time - Temperature Calculation.

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