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MASTER

Hanford Engineering Development Laboratory

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OBJECTIVE

The Spent Fuel Characterization Program at HEDL is divided into two main areas of work: The Waste Form Assessment and Spent Fuel Characterization and Deterioration Studies. The objective of the Waste Form Assessment is to evaluate five candidate waste forms for post-emplacment performance with emphasis on their ability to retard the release of radionuclides to the geology. The Spent Fuel Characterization and Deterioration study will identify and analyze those characteristics of spent fuel significantly affecting leaching and dissolution and those processes which may be active to degrade such fuel characteristics during the period from emplacement in the repository until breach of the waste package. This work focuses on the performance of the spent fuel waste form and its long-term resistance to release of radionuclides. Correlations to predict these properties as a function of time will be developed and evaluated based on theoretical considerations and available data.

The work and the correlations developed will ultimately be used to help establish appropriate repository and waste package design limits and in long-term performance assessments of spent fuel package design and candidate repositories.

PROGRESS REPORTED PREVIOUSLY

Work on the Waste Form Assessment was initiated at HEDL late in FY-1980 and the Spent Fuel Characterization and Deterioration Program started at the beginning of FY-1981. Other HEDL work on Spent Fuel/Package Performance was started in FY-1978. Recent accomplishments include the following:

The Waste Form Assessment task is evaluating the relative in-repository performance of five waste forms, especially their ability to retard the release of radionuclides. The waste forms are under evaluation for prebreach and postbreach performance relative to support against lithostatic pressure, long-term stability for radionuclide retention, minimization of cladding degradation, breach of the canister by possible gas pressurization, heat transfer, and criticality. Performance analysis of each of the waste form/stabilizer combinations for four temperature ranges was also initiated.

Spent fuel characterization and degradation studies have identified surface area, surface structure, grain boundary conditions, fission product positions, fuel phases, and valence states as important fuel characteristics which might significantly influence the dissolution and leaching rates of radionuclides. Chemical changes in the fuel, volatile fission product migration, diffusion of gas to grain boundaries, radiation damage due to decay of fission products, and fragmentation due to thermal or mechanical effects were identified as mechanisms which could possibly change spent fuel characteristics during repository residence. HEDL also identified fuel manufacturing and reactor operating conditions affecting the spent fuel characteristics and degradation mechanisms described above.

An analysis of prepressurization gas and fission gas release from spent fuel during geologic disposal was completed. Gas pressurization presents no harmful effects to the waste form, package or repository. Fuel assembly structural and fracture mechanics analyses of spent fuel under disposal cycle dynamic loading conditions were completed. Results show

ACTIVITIES DURING THE REPORTING PERIOD

Five spent fuel waste forms were analyzed for relative in-repository performance. The ultimate goal of the waste form/stabilizer combination is to prevent radionuclide release for as long a time as possible and then to limit the release rate to a low value. In order to evaluate the waste form/stabilizer combinations, their performance in terms of possible functional requirements was assessed. These included:

- 1) Waste Form/Stabilizer Support Against Lithostatic Pressure
- 2) Long-Term Stability for Radionuclide Retention
- 3) Minimization of Cladding Degradation
- 4) Prevention of Canister/Repository Breach Due to Pressurization
- 5) Stabilizer Heat Transfer
- 6) The Stabilizer as an Independent Barrier to Radionuclide Migration
- 7) Prevention of Criticality

Three stabilizers were considered: inert gas, particulate and solid. Only generic classes of stabilizers were evaluated since it was not within the scope of this work to study the merits of individual stabilizer candidates. For some of the functional requirements, a clear cut division between satisfactory and unsatisfactory performance could not be found. As such the rankings of many categories could only be interpreted as an attempt at qualification of a particular waste form's ability to meet the possible function. The waste forms were rated overall using a sequential ordering of the functional requirements. Conclusions from the study are as follows:

- 1) The stabilizer plays a minor role as an independent barrier unless its corrosion resistance is nearly the same or better than Zircaloy.
- 2) Criticality can be a concern if a single assembly is placed in a canister with a gas stabilizer, but only if some unlikely event occurs which rearranges the fissile material into an optimum critical shape.
- 3) Three critical fuel temperatures exist: 425°C - above which the cladding may be compromised; 600°C - where gas release from the fuel may become significant; and 660°C - where significant gas release will occur over the thermal period.
- 4) As long as the cladding stays intact during the thermal period, the intact waste forms perform equally well in limiting radionuclide migration. If the cladding is compromised during the thermal period, fuel-stabilizer-canister interactions may occur which degrade the fuel significantly. This interaction must be studied if fuel temperatures are set above 425°C since cladding integrity may be compromised.
- 5) Waste forms with particulate stabilizers will not appreciably help the canister resist lithostatic pressure.
- 6) Relative rankings of the waste forms is rather independent of the repository temperature although the absolute performance may vary with temperature drastically.
- 7) Ranking of waste forms is fairly independent of the relative weighting of the functional requirements.

Based on these conclusions, the relative performance of the waste forms dictates that the candidates be ordered as follows:

- 1) The best waste form/stabilizer combination is the intact assembly, with or without end bells, vented or unvented, with a solid stabilizer.
- 2) A suitable alternative is the combination of bundled close-packed rods with a solid stabilizer around the outside of the bundle to resist lithostatic pressure.
- 3) The other possible waste forms are of lower ranking with the worst waste form/stabilizer combination being the intact assembly with a gas stabilizer or the chopped fuel.

ACTIVITIES DURING THE REPORTING PERIOD (Continued)

Long-term degradation of fuel residing in a repository was studied as part of the Spent Fuel Degradation task. It was concluded that in intact fuel volatile fission product migration, fragmentation and radiation damage would not take place to any extent that could damage the fuel. Gas diffusion would also have little or no effect on the fuel, but it may influence the canister design. Studies using 10-year old fuel indicate that chemical changes should not be a problem either, but it is recommended that the change in valence state with decay of $Cs \rightarrow Ba$ be investigated further. The study concluded that cladding integrity should be preserved since fuel exposed to the canister environment may undergo major thermodynamic changes which cannot be analyzed with the current state of knowledge.

PROGRESS REPORTED PREVIOUSLY (Continued)

negligible damage to spent fuel assemblies because the resulting stresses are low under normal expected transportation and disposal cycle conditions. Fatigue crack propagation analyses demonstrated that large cladding flaws are stable under dynamic response loading conditions. Analysis of pretest destructive examination data for five rods from a Climax - Spent Fuel Test assembly was completed.

FUTURE ACTIVITIES

Comments from ONWI and the other contractors working on the Waste Form Assessment will be incorporated with a final draft report to be issued to ONWI before April 30, 1981. An evaluation of whole rod testing conducted during 1980 will be completed. New mid-year work, approved by ONWI in March 1980, will be initiated. This includes surface area modeling, analyses of the effect of stagnant water in the fuel-cladding gap on the fuel condition, and a comparison of analytic techniques to characterize grain boundaries. The spent fuel characterization task will be changed to consider and evaluate nondestructive techniques for determining characteristics of spent fuel.