LETTER REPORT

TITLE: "Review of the Report: Preliminary Bounds on the Expected Postclosure Performance of the Yucca Mountain Site, Southern Nevada, SAND84-1492 (December 1984), by S. Sinnock, Y. T. Lin, and J. P. Brannen"

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SUMMARY

The subject report from Sandia is a key document for the NRC evaluation of both the performance assessment strategy and the expected performance (radioactivity release) for the Yucca Mountain candidate repository. The report reaches favorable conclusions relative to the containment of radioactivity and compliance with the regulatory requirements for a repository at Yucca Mountain. The conclusions largely result from the favorable hydrologic model presented for the unsaturated zone at Yucca Mountain. It was concluded that groundwater flow aspects alone would assure that no waste could reach the water table underneath Yucca Mountain in 10,000 years or greater, thus the EPA radionuclide release requirements at the accessible environment would be automatically met. Favorable geochemical aspects such as sorption or matrix diffusion were relegated to secondary importance, while waste package performance was concluded to be relatively unimportant. A number of these conclusions may prove contentious, and we suggest that the NRC staff involved with the evaluation of Yucca Mountain information may wish to consider some of the assumptions and conclusions stated in the report.

1. DESCRIPTION OF THE REPORT

This report is a key document for the NRC evaluation of the current status of both the performance assessment strategy and the expected performance for a repository at Yucca Mountain. It was published (December 1984) by the NNWSI performance assessment team at Sandia, and we believe that it is considerably more up-to-date than the performance assessment information in the draft EIS for Yucca Mountain (DOE/RW-0012). The report is clearly written and is divided into a number of logical chapters. The report sequentially addresses: general assumptions, site conditions (hydrology, geochemistry, and rock characteristics), models the performance expected in relation to the regulatory requirements, and reaches a number of conclusions favorable to the licensability of a repository at Yucca Mountain. We have summarized some of these assumptions, conditions, and conclusions in the following sections of this Letter Report, along with our comments and recommendations.

2. GENERAL ASSUMPTIONS

The report carefully documents (Chapter 2, General Assumptions) the underlying assumptions utilized in the prediction of repository performance. The authors recognized that specific features of the repository have not been finalized at this time, and attempted to choose reasonable or conservative assumptions rather than actual information where necessary. Eleven general assumptions are detailed in the report; these are summarized in the following sections, along with our comments.

2.1 Repository Location

Assumption: The repository was assumed to be located in the lower part of the Topopah Spring Member of the Paintbush Tuff at Yucca Mountain.

Comment: The Topopah Spring Member appears to be the accepted or leading design location for the repository. This location is well above the current or historic water table but is still well below the surface of Yucca Mountain. The location is in a welded tuff member of appreciable size that may have acceptable mechanical properties. It seems likely that the Topopah Spring Member at Yucca Mountain may remain the candidate repository location.

2.2 Inventory of Spent Fuel

Assumption: The repository was assumed to contain 70,000 metric tons of heavy metal in 35,000 canisters of 10-year-old spent fuel which are simultaneously emplaced in the repository.

Comment: The waste form was assumed to be spent fuel in canisters; glass waste form was not considered in the report. Selection of spent fuel for the analysis seems realistic because no facility for reprocessing of commercial spent fuel, and the concomitant generation of glass waste form, exists or is planned in the United States. Some glass-form defense waste could be emplaced in the repository if it is not accepted by the WIPP facility, so extension of the analysis to include defense wastes could be desirable. The assumption that the fuel will all be 10 years old is conservative with respect to both heat load and fission product radionuclide inventory. When a repository becomes operational in the late 1990's, or later, much of the initial inventory of spent fuel will be more than 10 years old (out of reactor); some appreciable fraction of the fuel will be 20 to 30 years old. Thus, both the heat and fission product content of this older fuel will be less than for the assumed 10-year fuel. Some actinides such as americium and curium continue to grow into the fuel for a period after removal from the reactor, so the assumption of 10-year-age is not necessarily conservative for the inventory of these radionuclides. The assumption that the spent fuel inventory will be simultaneously emplaced in the repository also is conservative (and unrealistic). An operating period of several decades may be required to fill the repository, thus this time-before-closure, when added to the actual out-of-reactor time for the fuel elements, shows that the average age of the fuel elements would be considerably greater than 10 years, and the heat load and fission product inventory correspondingly less than that resulting from this conservative assumption. This conservatism may be of secondary importance, however, if no release from the waste package is assumed for 300 or 1000 years.

2.3 Repository Size

Assumption: The total area encompassing the waste will be 6.06 x 10^{-6} m^2 (~1500 acres) and the initial thermal power output will be 12 to 13 W/m^2 (~50 kW/acre).

Comment: As discussed above, the average age of the spent fuel (out-of-reactor time plus repository-operation time) will be greater than 10 years, thus, use of a calculated heat load for 10-year fuel is conservative. We are not able to comment on the design size of the repository or the value for the calculated heat load.

2.4 Waste Package Performance

Assumption: The waste package was assumed to remain intact (no release of radioactivity) for either 360 or 1060 years; a 60-year operational life for the repository plus the NRC waste package containment time regulation of 300 to 1000 years. The waste inventory of radionuclides was then calculated for 300 and 1000 years after closure.

Comment: The selection of 300 and 1000 year times appears reasonable, considering the regulatory requirements. The analysis in the bulk of the report also considers scenarios for failure of the waste package at shorter times. Consideration of possible-but-not-expected events is commendable, since this allows analysis of the radioactivity releases which could occur if the waste package fails to meet the regulation. Since prediction of waste package containment integrity for periods of up to 1000 years may be difficult to validate, it seems desirable to explore repository performance for such "what if" scenarios.

2.5 Release Mechanism

Assumption: Groundwater transport was assumed to be the only release pathway from emplaced fuel to the environment. It was assumed that spent fuel immediately dissolves into groundwater that contacts the spent fuel, and that the groundwater then contains dissolved uranium at the appropriate saturated solution concentration. It was further assumed that all other radionuclides in the spent fuel dissolve congruently with the uranium, (i.e., the ratio of any radionuclide to uranium in the fuel leachate solution is the same as in the spent fuel).

Comment: The assumption that groundwater transport is the only probable release pathway seems reasonable and is generally accepted for repository performance modeling. The assumption that fuel immediately dissolves to saturate ingressing groundwater is conservative. The UO_2 matrix of the fuel pellets is at close to theoretical density and UO_2 is very insoluble in groundwater in the absence of oxidants. Oxidation of solid UO_2 to soluble U(VI) species is assumed to occur during dissolution, and the rate of these reactions (oxidation and dissolution) may be relatively slow. As experimental information on spent fuel dissolution in Yucca Mountain groundwater is developed by ongoing DOE work at HEDL and actual leach rate values can be substituted for the instantaneous dissolution assumption, it seems possible that this instant-solubility assumption may prove to be very conservative. The assumption that all other radionuclides dissolve congruently with uranium and that these radionuclides maintain the same ratio to uranium in the leachate as in the spent fuel is inaccurate for many radionuclides and nonconservative for some radionuclides. Segregation of fission products occurs during reactor operation and volatile elements concentrate in the the fuel-fuel pin gap and/or at UO_2 grain boundaries where they are more available for rapid release during leaching than elements in the UO_2 matrix; cesium, iodine, technetium, fission gasses such as xenon, etc., are in this category. A number of fission products can reasonably be expected to leach more rapidly than the bulk uranium, thus, the assumption used in the modeling may lead to significant underprediction of releases for some fission product radionuclides. An ameliorating factor is that some of these radionuclides will decay during the 1000-year containment period and be of limited importance in the calculation of total radioactivity releases. The assumption of congruent release may be valid for actinides such as americium and plutonium; these are generally believed to be in solid solution in the UO_2 matrix. The solubility or saturated solution concentration for radionuclides other than uranium was not considered in the assessment. Some elements are less soluble than uranium, thus, ignoring the individual solubility values would be conservative for these elements, but unnecessarily so, since solubility information for many radionuclides is readily available from the geochemical programs used to calculate the uranium solubility in the report. For elements that are more soluble than uranium, however, this treatment may be nonconservative. Unfortunately, some elements, such as cesium, iodine, or technetium, may be both segregated in the fuel matrix and rapidly leached, as well as more soluble in groundwater than uranium. In these cases, the assumptions used in the analysis to calculate the leachate concentration of these radionuclides could be significantly nonconservative and result in important underpredictions of radioactivity releases.

2.6 Uranium Solubility

Assumption: The uranium solubility was assumed to be equivalent to that calculated for current geochemical conditions, and to remain constant over time.

Comment: The ambient geochemical conditions used in the calculation in the report are a temperature of 25°C, a pH of 7 to 8, and an Eh of 700 mV. The repository temperature may be somewhat higher than 25°C at the time of uranium release, and the groundwater parameters (ph and Eh) could be somewhat different than ambient as a result of geothermal and/or radiolytic groundwater/tuff reactions during the period of containment. Use of current conditions probably represents a default situation, since better data on geochemical parameters over repository time seem not to be available yet. Further, thermodynamic data to support elevated temperature calculations is not available. This assumption, use of calculated uranium solubility, is not in concordance with the NRC Technical Position on Solubility; that position calls for the use of empirically measured solubility values because the thermodynamic data base and calculation methodology may be inadequate to describe the complex elevated temperature behavior of important radionuclides. It seems possible that this assumption may have to be altered in future Yucca Mountain performance assessment analyses. It is not possible to estimate if the present assumption may over or underpredict the uranium-saturated solution concentration under repository conditions.

2.7 Amount of Groundwater

Assumption: The amount of groundwater available to transport radionuclides will be only a fraction of the total volume of water moving through the repository. This fraction will depend upon hydrologic factors (infiltration, flux at repository level, etc.) and repository engineering design factors (area of the repository, spacing and location of waste canisters, etc.).

Comment: This assumption seems reasonable. Various values for the factors affecting the amount of groundwater are taken in the analysis. Because groundwater transport is the only release mechanism treated, the selection of the values used becomes very important for the results of the analysis and warrant attention by the NRC Hydrology Section.

2.8 Release Pathway

Assumption: The release pathway from the repository to the accessible environment is assumed to be vertically downward to the water table, and then horizontal along the water table to a point either 2 or 10 km distant.

Comment: This assumption does not consider that the potable water beneath Yucca Mountain could be designated as a potable aquifer. Also, the assumption treats only a simplified release pathway and does not allow for lateral flow in any of the tuff members above the water table. This assumption may have to be modified as the hydrology is better understood at Yucca Mountain.

2.9 Porous Flow Assumption

Assumption: The water velocity away from the repository is assumed to be equal to the flux divided by the effective porosity of the tuff members through which flow occurs.

Comment: This is an important hydrologic assumption for the performance assessment because it has a major impact on the calculated radioactivity release rate. Groundwater flow is assumed to occur via porous flow, and only minor attention is given in the assessment to the possibility of fracture flow in one or more of the unsaturated zone tuff members. This porous-flow assumption and the idealized treatment of water flow in the unsaturated zone should be given careful scrutiny by the NRC Hydrology Section.

2.10 Radionuclide Retardation Factor

Assumption: The rate of transport of radionuclides in groundwater away from the repository will be equal to the groundwater velocity divided by a retardation factor. The retardation factor combines the effects of chemical reactions, such as radionuclide sorption and mineral precipitation, and of physical effects such as diffusion.

Comment: Use of a single retardation factor to represent the effects of geochemical processes as dissimilar as sorption, precipitation, and matrix diffusion is not conventional and may represent a significant over-simplification in the performance assessment. Further, the retardation factor is assumed to remain constant with time; for some of the reactions involved, this could be a nonconservative assumption. It would be desirable to treat each of the geochemical processes which may result in radionuclide retardation separately so that changes over time can be realistically modeled. Also, the calculation of a retardation factor from laboratory sorption experiments assumes that radionuclide migration can be described as an ion-exchange phenomena, that the sorption reactions are rapid and reversible, that multiple radionuclide species and forms do not exist, and that the laboratory methodology and materials selected model repository materials and conditions. Some of these underlying assumptions may not be well met by the published sorption information for Yucca Mountain (see ORNL Letter Report LR-287-7). In general, the treatment of radionuclide retardation used in the performance assessment seems oversimplified and likely to be significantly inaccurate and possibly nonconservative for a number of radionuclides. This may be particularly true for actinide elements where multiple species and forms are known to exist under repository conditions and some reactions are slow or irreversible. Information on possible colloidal transport of radionuclides may also be particularly uncertain. It seems possible that this radionuclide retardation treatment may have to be substantially altered in future Yucca Mountain performance assessment reports.

2.11 Radionuclide Decay

Assumption: The decay of radionuclides over time and the accumulation of daughter products is calculated using a five-member decay chain calculation.

Comment: Because of the long times involved in modeling repository performance, it is highly desirable to include decay and growth of daughter radionuclides over time in the modeling calculations. We are not able to comment on the calculational model selected for this report.

3. GEOCHEMICAL INFORMATION

The geochemical information utilized in the performance assessment is described in the report Section 3.2, Geochemistry. Solubility and retardation information is given, and the underlying assumptions and the calculations used to develop the values employed in the modeling calculations are reported. The information is summarized below, along with our comments.

3.1 Uranium Solubility

Information: The uranium solubility in the repository over time was assumed to be similar to the current ambient temperature solubility. The groundwater composition and parameters were assumed to be similar to those for saturated-zone groundwater: a dilute Na^+, HCO_3^- solution at pH 7 to 8 and Eh of 700 mV. The uranium solubility was then calculated using EQ3 and MINTEQ both in the presence and absence of plutonium complexation of CO_3^{2-}. The uranium solubility was calculated to be $<10^{-4}$ mol/L, and possibly to be as low as 10^{-6} or 10^{-7} mol/L.

Comment: This calculational approach to develop solubility information is not in agreement with the NRC Technical Position on Solubility which calls for the use of measured solubility values. Even for uranium, the thermodynamic data base may be insufficient to support defensible calculations. This inadequacy is particularly true for elevated temperatures, and the lack of thermodynamic data probably led to the assumption of repository solubility over time being equivalent to that calculated for current ambient conditions. The treatment assumed that the ingressing groundwater composition and parameters would be equivalent to current saturated zone groundwater. No attempt was made to model geochemical or radiolytic groundwater/tuff/waste package reactions which might occur over time at elevated temperatures. The approach taken for obtaining values of uranium solubility may be oversimplified and may not be defensible. It is possible that the NRC may not find the treatment of uranium solubility given in this performance assessment report to be adequate.

3.2 Uranium and Other Radionuclides Dissolution Rate

Information: The uranium in spent fuel waste is assumed to dissolve instantly to reach the saturated solution concentration. All other radionuclides in the waste are assumed to be limited by the uranium dissolution rate and to dissolve at the same rate.

Comment: The assumption of instant uranium solubility must be conservative. The dissolution rate calculated, therefore, also will be conservative if the values for the groundwater flux and saturated solution concentration are accurate or conservative. While the report concludes that the assumption of congruent dissolution of other radionuclides is "probably conservative," we feel that it likely is inaccurate for a number of radionuclides and possibly nonconservative for some important radionuclides (see Section 2.5 of this Letter Report). It is possible that the NRC may find this greatly over-simplified description of fuel dissolution behavior to be inadequate.

3.3 Retardation Factors

Information: It is assumed that all radionuclide retardation processes can be described by a single retardation factor value which is invariant with time. It is further assumed that this retardation factor can be calculated from sorption ratios (distribution coefficients) measured in batch contact laboratory experiments with crushed tuff. Sorption retardation under fracture flow conditions are also approximated using a simplified expression which relates fracture sorption to the fracture surface area. Radionuclide diffusion into the rock matrix is also discussed.

Comment: In Table 5 of the report, representative sorption ratios are given for eight elements for each of six tuff members; large differences can be seen for the values for a given element in different tuffs. Use of these sorption ratios to calculate retardation factors assumes that porous flow predominates, that multiple radionuclide species or forms do not exist, and that the sorption processes are rapid and reversible. These basic requirements are not well satisfied in the Yucca Mountain tuff/groundwater systems. We have previously criticized the methodology and underlying assumptions involved in this batch contact work; see our Letter Report LR-287-7. The estimation of retardation under fracture flow situations must be considered highly speculative, since little information on fracture surface area now exists for Yucca Mountain. Only limited emphasis is given in the analysis to fracture flow or diffusion since porous flow is expected to describe groundwater movement in tuff at Yucca Mountain. It is possible that the NRC may find that the treatment of retardation in the performance assessment analysis may be too simplified and potentially nonconservative to be defensible.

4. EVALUATION OF THE REPORT CONCLUSIONS

The conclusions of the Yucca Mountain repository performance assessment are contained in just over one page of text which comprises report Chapter 5, Conclusions. The conclusions are summarized below, along with our comments and recommendations.

4.1 Groundwater Flux

Conclusion: The report concludes that paramount among the favorable features for a repository at Yucca Mountain is the small amount of water available to dissolve waste after it has been emplaced.

Comment: This conclusion is based on the assumed low infiltration rate, combined with porous flow in the unsaturated zone. Emphasis is given in the analysis to the favorable waste isolation aspects resulting from the slow movement of a small volume of groundwater.

Recommendation: This is the single most important aspect of the performance assessment, and the consequence of utilizing slow movement of a small volume of water dominate the assessment calculations and results. This hydrologic issue should be carefully analyzed by the NRC Hydrology Section. If this groundwater model should prove to be inadequate or nonconservative, then the entire performance assessment treatment of radioactivity releases to the environment is correspondingly inaccurate or nonconservative. Conversely, if this model is validated or substantiated after further scrutiny, then the Yucca Mountain site may indeed be uniquely favorable as a repository location.

4.2 Groundwater Flow Mechanism in the Unsaturated Zone

Conclusion: Openings in the rock in the unsaturated zone tend to block the flow of groundwater (this is the converse of the situation in the saturated zone, where openings are conduits). This flow mechanism ameliorates concerns about repository-induced or natural changes that might fracture the repository rock.

Comment: This conclusion is based on the suction and storage capacity created by the unsaturated pores in the unsaturated zone tuffs, and the assumption that free liquid water cannot coexist with such rock properties. This condition is believed to preclude the flow of liquid water in fractures until, or unless, sufficient infiltration should occur to saturate or nearly saturate the rocks. Thus, as long as the rocks remain relatively unsaturated, all groundwater movement must be porous flow.

Recommendation: This is another important hydrologic issue that may be favorable for the location of a repository at Yucca Mountain. The NRC Hydrology Section should carefully examine this conclusion. Much of the performance assessment analysis rests on the validity of this conclusion.

4.3 Times for Waste Leachate to Reach the Water Table

Conclusion: Even if liquid water reaches the waste, the low flow rate and high rock porosity will limit flow velocities so that no waste leachate will reach the water table for 10,000's or 100,000's of years.

Comment: The report actually concludes that it is unlikely that water can contact and dissolve the waste, but for this calculation, it is postulated that a low flux of groundwater does enter the repository and leach emplaced waste. These long times for waste leachate to even reach the water table, not the accessible environment at 2 or 10 km distance along the water table, are obviously concluded to be very favorable for a repository at Yucca Mountain.

Recommendation: This conclusion results from the low groundwater flux and porous flow assumptions described above, plus the expected favorable retardation due to radionuclide sorption by zeolites. The NRC Geochemistry Section may wish to evaluate the radionuclide sorption treatment given in the report. If sorption is not as favorable as estimated, then the migration times obviously will not be as great as calculated.

4.4 Satisfaction of EPA 10,000-Year Release Requirements

Conclusion: No wastes could move the several hundred meters from the repository to the water table in 10,000 years. Thus, the EPA requirements are automatically satisfied since no release can occur during the first 10,000 years. It is further specifically stated that water travel time through the unsaturated zone alone is sufficient to provide the necessary isolation. Geochemical retardation is not considered essential to ensure compliance with regulatory requirements, but it is stated that geochemical processes do add confidence in the ability of the site to perform satisfactorily. It is also concluded that the behavior of the waste package is relatively unimportant.

Comment: This is a very forceful set of conclusions that are based on the favorable hydrologic model presented for Yucca Mountain.

Recommendation: These strong statements are based on hydrologic issues that should be carefully scrutinized by the NRC Hydrology Section. The relegation of waste package performance to a minor role and the conclusion that sorption is not essential may prove to be contentious.

4.5 Possible but Unlikely Event

Conclusion: If infiltration and groundwater flux exceed the carrying capacity of the rock matrix, then rapid fracture flow could occur through the unsaturated zone. However, fracture flow is not expected to jeopardize complete waste isolation for 10,000 years since matrix diffusion would provide adequate retardation. If fracture flow could somehow occur in the absence of matrix diffusion, then the waste package performance and geochemical retardation would be more important in providing isolation.

Comment: The favorable performance expected even under infiltration conditions that overwhelm the expected unsaturated zone favorable hydrology are based on assumptions about the behavior of groundwater in the tuff that may be only poorly supported by experimental validation.

Recommendation: The presence, or absence, of data to support the contention of matrix diffusion under near-saturated conditions should be considered. The NRC Geochemistry Section might want to address the issues raised in this brief, and possibly superficial, treatment of a possible but unexpected event.

4.6 Qualifying Statement

Conclusion: The results presented in the report must be considered provisional until the level of understanding of site characterization at Yucca Mountain is improved.

Comment: The report correctly states that more detailed information will be needed to improve the performance assessment analysis.

Recommendation: The NRC might wish to consider what performance assessment strategy approach could be required to achieve a valid and defensible analysis that would satisfy regulatory requirements.