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LETTER REPORT

TITLE: Review of "Effect of Host-Rock Dissolution and Precipitation on Permeability in a Nuclear Waste Repository in Tuff," by J. F. Braithwaite and F. B. Nimick, Sandia Report SAND84-0192, September, 1984.

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REVIEW

An important aspect of the performance assessment of a waste-isolation system at Yucca Mountain is the extent of change in permeability due to thermally-induced host-rock dissolution and precipitation processes and whether such changes constitute a potentially adverse condition, even though the minerals composing the tuff at the Yucca Mountain site are thought to have limited solubilities in groundwater. This report concerns a theoretical study which attempted to establish bounds on the permeability changes which could occur due to such alteration processes. The assumptions chosen were expected to yield conservatively high estimates of changes in matrix and fracture porosity. These predictions then were used to calculate new matrix and fracture permeabilities. The three types of information deemed essential to this estimation process were: identification of the controlling mineralogy, environmental effects on amorphous silica solubility, and the temperature distribution.

The authors observed that the Topopah Spring Member of the Paintbrush Tuff, where the proposed repository would be located, is composed primarily of alkali and plagioclase feldspars, together with several forms of silica, including quartz, cristobalite, and tridymite. The analysis of the dissolution and precipitation of minerals was considerably simplified by assuming that the groundwater always maintained saturation with respect to amorphous silica. Thus, the exchange of material between the groundwater and the rock was attributed to changes in the solubility of amorphous silica with changes in temperature. The analysis employed three bounding assumptions: (1) cristobalite is the solid phase contributing most of the silica to the groundwater; (2) silica concentrations in groundwater will be determined by equilibrium saturation with amorphous silica; and (3) amorphous silica is the solid phase which precipitates from cooling groundwater. These assumptions were thought to produce conservative results, because the authors found that laboratory validation, where available, suggested that these assumptions led to maximum potential for reducing permeability.

Environmental effects on amorphous silica solubility appeared to be relatively minor. Several studies quoted in this report have shown that moderate quantities of cations such as sodium, potassium, calcium, and magnesium either have no effect or decrease the solubility. Silica solubility is not affected significantly by changes in pH over a pH range of at least 4 to 9. Studies on the effect of temperature on solubility have shown that silica concentrations in water increase with increasing temperature, regardless of the form of silica investigated. The effect of pressure on solubility is not clear, as literature references suggest either that the effect is negligible or that there is a slight increase in solubility with pressure.

Of the two repository waste forms being considered, it was concluded that spent fuel would contain more heat-producing radionuclides, which would be expected to lead to higher temperatures in the repository and a greater amount of dissolution and precipitation. Thus, for the purposes of these calculations, spent fuel was chosen as the waste form which would lead to a conservative bound on thermally-induced dissolution and precipitation. The fuel was assumed to be deployed uniformly throughout the repository as a flat disk, at a depth of 390 m below the surface. Two areal power densities were considered: the current reference value of 57 kW/acre (14 W/m^2), and a higher value of 90 kW/acre (22 W/m^2). Temperature profiles to depths of 600 m were considered, as that is the approximate depth of the water table, at which depth permeability changes become less important. Decay times to 10,000 years were considered, as specified in 10 CFR 191.

The calculational model assumed that the groundwater was always at equilibrium with amorphous silica. The computer program calculated the temperature distribution as a function of time and position, the amorphous silica solubility, the incremental amount of silica dissolved or precipitated, and the cumulative change in porosity. The effects of water flow rate, initial rock porosity, power density, and time on the change in porosity were evaluated. The 600 m of rock along the vertical centerline of the repository was subdivided into six stratigraphic units for the purpose of modeling hydraulic behavior.

Some important results of this study are:

1. Cumulative porosity changes derived from the model correlate qualitatively with the calculated temperature profiles.

2. Only very small changes in cumulative porosity are predicted to occur.

3. Net cumulative porosity decreases do not occur in the calculations unless water is assumed to vaporize.

4. Changes in water flow from 0.1 to 4 mm/year have only a small effect on cumulative porosity changes.

5. The question of whether host-rock dissolution and precipitation could produce significant decreases in permeability was investigated for two regimes of water flow:

   a. If porous flow dominates, as now seems the case for percolation rates less than 0.5 mm/year, no observable change in porosity is expected.

   b. If fracture flow dominates, with water fluxes greater than 0.5 mm/year, large changes in fracture permeabilities can occur, but the bulk permeability due to fractures is much greater than needed to transmit the amount of water required. Thus, there should be no significant effect on the total hydrologic flow patterns through the mountain.

EVALUATION

This report describes a preliminary examination of the effect of heating and subsequent cooling of groundwater as it percolates vertically down through the host rock of a repository in the tuff of Yucca Mountain, where the heat of the radioactive waste could induce host-rock dissolution and precipitation, which, in turn, could alter the permeability of the tuff. It is believed that the transmissivity of the tuff layers is sufficient to allow water to flow through the tuff without saturation. Dissolution of rock would only enhance the transmissivity; however, mineral precipitation would decrease the permeability and, in an extreme case, might cause the rock to saturate.

The model employed and the underlying assumptions appear adequate for setting bounds on the problem, although a number of simplifying assumptions were made which might not be adequate for a final performance assessment. As might be expected for a system characterized by such low water flow rates, the model assumes equilibrium between the host rock and the dissolved minerals. Kinetics may be more important than realized, although the authors have otherwise attempted to bound the problem in as conservative a manner as current knowledge will permit. The data from recent studies on water flow at the Yucca Mountain site appear to be adequate for setting bounds on this problem with the current model. However, for more precise modeling, the groundwater flow characteristics will need to be determined more accurately than at present.

A key assumption in the model is the identification of the controlling mineralogy in the dissolution-precipitation process. The model is simplified by making the reasonable assumption that the groundwater is always saturated with respect to amorphous silica, and that the rock and aqueous solution are at equilibrium. This assumption is conservative only if the choice of controlling mineralogy is correct. On page 41 of the report the authors give as a result of their calculations a simple restatement of their premise that the groundwater solution is at equilibrium with amorphous silica. This reviewer takes the position that the calculations produce results consistent with the subject assumption as a starting point, but such consistency is not proof that the starting assumption was correct. Finally, without further validation of the results of the calculations to some degree, we cannot be certain that other dissolution-precipitation processes are not also significant.

Because there is not yet a final design for a nuclear waste repository at Yucca Mountain, the authors' choice of repository geometry and areal power density may be open to question. However, it seems to this reviewer that their choice of parameters is sufficiently generic for the purpos of setting bounds on the problem at hand.

Validation of a model such as this presents formidable difficulties, but, at the very least, it appears that more detailed laboratory and field studies are needed to verify the controlling mineralogy, and to improve our knowledge of water flow characteristics, tuff porosities, and kinetic effects. This model as it stands is a useful tool for preliminary examination of the effects of thermally-induced dissolution and subsequent precipitation of host-rock minerals and shows that such effects should not be expected to affect significantly the total hydrologic flow patterns through a repository in tuff.