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THE CONSTRUCTIVE USE OF HEAT IN AN UNSATURATED TUFF REPOSITORY

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ABSTRACT

By designing the engineered barrier system in an unsaturated tuff repository to constructively use heat, the waste containers can be kept dry for hundreds of years. Without water, the aqueous processes that release and transport radionuclides do not operate. In the plans of most international programs, waste is cooled prior to disposal in granite or salt. For these rocks there are technical issues favoring reduced heat. Recently, it has been suggested that the U.S. Program adopt a strategy of cooling nuclear waste prior to disposal. This paper reviews technical issues associated with the role of heat in an unsaturated tuff repository and concludes that the overall effect of heat in such a setting appears to be beneficial to waste isolation.

INTRODUCTION

In 1982, tuff in the unsaturated zone was chosen in preference to three saturated units as the target horizon for a proposed repository at Yucca Mountain, Nevada. This selection recognized the numerous virtues of the unsaturated zone for waste disposal as summarized by the U.S. Geological Survey.[1,2,3] Among many reasons for the selection was the recognition that, in unsaturated rock, decay heat from high-level nuclear waste may be beneficial for a repository. Given the repository schedule and young age of waste then being considered, calculations showed that many waste containers might be kept above 100° C (and hence dry) for the entire "containment" period of 300 to 1000 years required in the NRC regulation 10 CFR 60.[4] Because water is required as a transport medium for all but gaseous radionuclides, more than 99.9 % of the curies in such dry containers would remain immobile even if the container were breached by some process other than liquid aqueous corrosion. Although the DOE Site Characterization Plan [5] does not explicitly allocate functional performance to the constructive use of heat, this concept underlies the conceptual designs for the waste packages and the repository.[6]

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Eriksson has recently advocated [7,8] that the US high-level waste program adopt an approach wherein a maximum temperature of the geologic setting of 90° C is maintained in the repository. The essence of Eriksson's argument is that "temperatures induced into the geologic setting reaching levels above the boiling point of water will seriously complicate and increase the uncertainty of predicting resulting processes and events." [7] After noting DOE published estimates that the surface of the rock adjacent to the waste canister may reach temperatures as high as 235° C, Eriksson states "A temperature of this magnitude will induce severe stresses and trigger complex coupled processes and events in a large volume of the geologic setting that cannot be credibly predicted today. Conversely, a lowering of the maximum temperature of the geologic setting below the boiling point of water would eliminate most of these uncertainties." [7] Eriksson and Pentz [8] state that "The most apparent benefit of reducing the temperature to this level is the simplification and possible elimination of coupled multi-phase flows and complex geochemical and hydrogeochemical reactions."

The U.S. Nuclear Waste Technical Review Board (NWTRB) has also raised the issue of reducing the thermal loading of a repository in order to reduce uncertainty. [9] Both Eriksson and the NWTRB base their suggestions in part on the common plans in the international nuclear community to cool waste prior to disposal. For media other than unsaturated rock, there are technical issues favoring reduced heat. In salt, brine migration, rapid closure of openings, and possible fracture of cap rocks have been raised. In saturated granite, the bentonite packing material is the major issue. The purpose of the present paper, however, is to review the role of heat only in an unsaturated tuff repository.

DISCUSSION

Two key assumptions appear to underlie the concept of limiting the temperature to below boiling or 90° C: (1) processes and events at higher temperatures have deleterious consequences, and (2) uncertainties in predicting processes and events are significantly greater for temperatures above the boiling point of water than below. In order to determine whether these assumptions apply at Yucca Mountain, one has to look at related information such as the volume of rock heated to above boiling, the processes and associated uncertainties, and finally at the engineered barrier itself.

Volume of rock heated to greater than 100° C

In the conceptual design for a Yucca Mountain Repository, the volume of rock heated to greater than 100° C is relatively small compared with the distance to the accessible environment (The boiling point of water at the Yucca Mountain repository level is about 97° C; [10] the value of 100 is used for convenience). It is less than about 30 m thick for various vertical hydrological fluxes and times out to 400 years post-emplacement.[11] Because the thermal peak at the emplacement hole wall is attained at about 20 to 40 years post-emplacement, this volume grows little beyond 100 years.

Not only is the volume of rock heated to greater than 100° C small, but also the volume heated as high as 150° C is still smaller. While the calculations cited by Eriksson [7] show maximum temperatures at the emplacement hole wall as high as 235° C, the reference he cites [10] also shows the maximum rock temperature one meter from the borehole wall peaking at 185° C ten to twenty years after emplacement and declining to 150° C by 100 years. Five meters from the borehole wall, the temperature in the reference design never exceeds 150° C. Even assuming uncertainty and deleterious processes at these temperatures, the rock volume affected is small relative to the scale of the geologic setting that will provide isolation.

Processes and uncertainties

Fluid flow: There are two fluid-flow systems that need to be modeled in order to understand the aqueous transport of radionuclides from an unsaturated repository: the natural system and the perturbed system. The natural system includes the large-scale infiltration of pulses of liquid water originating at the land surface and the potential for vapor-phase water transport and capillary condensation redistributing water through the vadose zone. The perturbed system is the response of the natural system to heat from the introduced waste. Both fluid-flow systems will have to be modeled regardless of the temperature limit of the repository. For both systems, the models used will have to handle two fluid phases: liquid water and water vapor. Thus keeping the temperature below 90° C does not result in the "possible elimination of coupled multi-phase flows and complex geochemical and hydrogeochemical reactions".[8]

The modeling of processes involved in drying the rock at the repository horizon due to heating above the boiling point of water is easier than modeling sub-boiling processes. First the drying involves one geologic unit, whereas modeling pulses of liquid water from the surface involves multiple units. Second, the scale of the drying is much smaller (vertical 60 m) than the 500-600 m from the surface to the water table. Third, some drying will occur for any temperature above ambient, and this will have to be modeled even if temperatures are held below 90° C. Fourth, experimentally validating the model is more feasible for the smaller scale, more rapid boiling process. Experiments demonstrating the feasibility of such validation have already been conducted.[12] Fifth, modeling the flow of water vapor is less sensitive to variations in fracture geometry and presence of fracture coatings than modeling the flow of pulses of liquid water.[13] The last is true even at the local scale in the repository, so that it is easier to model the drying of the rock around waste canisters than the flow of sub-boiling fluid in a thermal field.

To reiterate, the uncertainties of modeling fluid-flow in an unsaturated tuff repository are not reduced by maintaining a temperature below the boiling point of water.

Geochemistry and Mineralogy: The ambient rock temperature at Yucca Mountain at the proposed repository level is about 30° C. While it is possible that having no elevation of temperature at all may reduce uncertainty by removing an entire process from those that need to be modeled, the benefit of restricting temperature to about 90° C is not clear. Temperature accelerates the rate at which most reactions proceed in liquid water under hydrostatic pressure, following an Arrhenius relationship for which the boiling point of water has no special significance.

While water held under high capillary pressure may remain liquid to 140° C, [11] most water will boil at 100° C. Therefore, in rock at or above 100° C aqueous reactions will take place only with a small volume of residual capillary water, while rock at 90° C will react with a much larger volume of contained water. Even if the rock did alter in the presence of heat and liquid water, the alteration products would favor waste containment, being hydrous clays and zeolites that sorb radionuclides. Alteration in the presence of steam is most likely to take place along fracture surfaces, which is the exact place it would be most useful in inhibiting migration of radionuclides.

The chemical changes in the residual liquid during open system boiling need to be considered, because a more aggressive solution could be produced. Compared to the rock, the volume of water available to boil is small, less than ten volume percent. The time period during which boiling will occur is short for any discrete volume of rock. By contrast, evaporation of water at 90° C will occur slowly, producing a concentrated solution over long time periods. In either case, due to the chemical buffering available from the rock itself, the consequences are unlikely to be significant.

The same arguments that favor a dry environment for inhibiting reactions involving the container material or the waste form pertain to the rock itself. The rock where the waste would be emplaced consists predominately of non-hydrous silicate minerals. Even though thermodynamically metastable at ambient Yucca Mountain conditions, the kinetics of alteration to more stable mineral assemblages are very slow. These volcanic rocks have not altered their mineralogy despite hundreds or even thousands of years at temperatures between 100 and 235° C following their formation.

A few minerals contained in the tuff exhibit reversible changes in crystallographic structure at a certain temperature. Because these phase transitions produce a volume increase, this has been cited as a possible deleterious effect of heat. However, the significant transitions are not near 90° C. The alpha-beta cristobalite transition, which occurs at 225 ± 25° C and results in a volume increase of about 5%, is the only one within the possible range of repository rock temperature.[10] Because cristobalite is generally less than 30% of the rock volume and is distributed in small grains, there would likely be little effect even were this transition temperature reached in a few cm of rock around the borehole.

Hydrous clays and zeolites dry in the presence of heat and lose volume, some ultimately undergoing irreversible transformations. These reactions are inhibited by the presence of liquid water and water vapor, and are also compositionally dependent. The important considerations for this discussion are that:

(1) although significant in the underlying geologic unit, hydrous clays and zeolites are present only in very small amounts in the repository unit,
(2) there are measureable effects in both smectite clays and clinoptilolite zeolite as low as 50° C with no sharp discontinuity in the effect at the boiling point of water (For certain compositions, total reversible smectite collapse could occur at or lower than 90° C), and
(3) irreversible effects at the expected (~ 1 ATM) water vapor pressures tend to occur in excess of 150° C, a temperature that only a very small volume of rock will exceed.

<u>Rock Mechanics:</u> Heat-induced stresses are well below strength of the rock for designs in both granite and welded tuff. Concern over either borehole stability or drift stability have not been significant in conceptual design studies at Yucca Mountain. Matrix thermal expansion is first accommodated by closure of fractures. For the temperatures expected, the total volume change of the rock due to isobaric thermal expansion is less than one percent.

Only for the designs in basalt at Hanford, where the repository was very deep and the ambient temperatures and lithostatic stresses were already high, has rock stability been a concern in the U.S. Program. In the Hanford case, intact (unfractured) rock was essential as a barrier to underlying and overlying aquifers, whereas at Yucca Mountain the repository horizon is extensively fractured and induction of new fractures is not a concern.

The engineered barrier system

Elevated temperature in a saturated site involving liquid water under hydrostatic pressure would accelerate reactions involving many components of the engineered barrier system. A number of these aqueous reactions could be deleterious, including container corrosion and waste dissolution. In contrast, an unsaturated repository above the water table leads to a condition of elevated temperature in the presence of air at atmospheric pressure. The deleterious aqueous reactions noted for the saturated case do not occur.

While it is generally true that material degradation phenomena increase with temperature, there are exceptions. For example, the general corrosion and oxidation rates of candidate austenitic stainless steels for a Yucca Mountain container declined from 50 to 150° C.[14] In many aqueous solutions, pitting susceptibility of certain austenitic alloys reaches a maximum in the 50 to 90° C range and improves above that.[15] Protective oxide layers (which are stable in the Yucca Mountain oxidizing environment) on nickel-chromium-iron alloys and titanium alloys are thicker, more adherent, and denser at elevated (200° C) temperatures.[16]

An argument commonly made for limits on heat output of the waste is the ability to use packing material. Limits similar to the 90° C limit suggested by Eriksson originated for a Swedish repository in a saturated granite with bentonite packing.[17] Because of its low thermal conductivity, packing at Yucca Mountain could raise the peak cladding temperature of spent fuel above established limits.[18] Either older cooler waste or a reduction in the amount of waste in each container would be required. However, assuming these design adjustments are made, is packing really an advantage for a repository in unsaturated tuff?

Because it provides a solid matrix with a suction potential, packing <u>assures</u> that water arriving at the edge of the borehole wall eventually will contact containers. Packing also provides a reactive interface for corrosion and a medium for diffusion of waste radionuclides from the container to the rock. This is true for both saturated and unsaturated sites. With an air gap in an unsaturated site, it is <u>possible</u> that a single package could remain uncontacted by liquid water from the borehole wall for 10,000 years. Liquid water draining along a fracture could enter the borehole, drain down the wall and out the bottom of the hole without contacting the container.

A concern about heat specific to a Yucca Mountain repository is its potential for increasing the rate at which any UO_2 in spent fuel that is exposed to air will oxidize to U_3O_8, thereby disaggregating the fuel.[19] For this reason, containers are filled with an inert gas and sealed before being emplaced in the repository. In this case, heating to above the boiling point of water can keep the container dry (hence intact and filled with inert gas) during the exact time the spent fuel is most reactive (due to heat). By the time that the container temperature falls below boiling and aqueous corrosion can occur, the reactivity of the UO_2 is significantly lower and the short-lived highly soluble and gaseous radionuclides have decayed.

Although current engineered barrier system designs would lead to a small volume of rock heated above boiling, having a larger volume would not necessarily be deleterious. Danko[20] has proposed techniques for distributing heat in a Yucca Mountain repository that would lead to a larger volume heated above 100° C, but in which the heat is more evenly distributed and maximum rock temperatures reduced. Eriksson in the paper suggesting a temperature limit below 90° C discusses a design concept (Minimum Disturbance to the Geologic Setting or MD) wherein heat could be redistributed and removed in a repository instead of storing waste on the surface to cool.[7] Such concepts that allow the rock to dry while not greatly exceeding 100° C offer the potential benefit of combining constructive use of heat to dry the rock while moderating processes that accelerate with increasing temperature.

CONCLUSIONS

In order to evaluate whether possible deleterious effects of heat outweigh the beneficial effects, the following questions need to be answered:

1. Is the volume of rock heated above 100° C significant?
2. Are the processes in this heated zone deleterious to waste isolation?
3. Are there significantly greater uncertainties in this zone compared with surrounding areas of lower temperature?

<u>Volume</u>: The volume of rock heated to greater than the boiling point of water is about 30 meters thick. At Yucca Mountain the boiling point isotherm is unlikely to extend into overlying

and underlying rocks that have a higher percentage of hydrous minerals affected by temperatures as low as 100° C. The volume heated above 150° C is small, extending only a few meters around each waste package. Even if such temperatures were deleterious to the rock's capability to isolate waste, these volumes are small compared with the overall scale of the mined geologic disposal system.

Processes: The fluid flow processes at temperatures above the boiling point of water dry the containers and adjacent rock and are thus favorable to waste isolation. The geochemical and mineralogical processes at elevated temperatures may lead to a slight decrease in sorptive capability near the waste that is reversible upon resaturation and cooling and which can occur even below 90° C. The heat-induced stress may close fractures and induce loss of permeability immediately adjacent to the waste packages--an effect which is also continuous with respect to temperature and therefore present below 90° C. Neither the loss of sorptive capability nor permeability in the very near-field has been shown to be deterimental to waste isolation. Therefore the processes that take place in the rock at temperatures greater than 90° C are either favorable or benign to waste isolation.

Uncertainty: Because most of the effects of temperature on geochemical processes are continuous rather than discontinuous with respect to the boiling point of water, there is no discrete increase in uncertainty at 100° C. While it is true that a great deal more information exists for many materials and processes at room temperature than at elevated temperatures, the difference between 90 and 110° C is not nearly as pronounced. Furthermore, because of the acceleration of reaction rates with increasing temperature, the certainty of information becomes greater at higher temperature for many reactions because they are more easily measured experimentally. The uncertainty of predicting liquid pulses in the unperturbed environment is much greater than the uncertainty of predicting the location and effects of a drying front. Because tracking of a drying front is discrete, more and better data can be obtained than for a sub-boiling condition in rock.

Overall, the effect of heat in an unsaturated tuff repository appears to be beneficial to waste isolation.

REFERENCES

1. I. J. WINOGRAD, "Radioactive waste storage in the arid zone", Eos, Transactions of the American Geophysical Union, v. 55, p 884-894, (1974)

2. I. J. WINOGRAD, "Radioactive waste disposal in thick unsaturated zones,"Science, v. 212, p. 1457 (1981)

3. E. H. ROSEBOOM, JR, "Disposal of high-level nuclear waste above the water table in arid regions,"U.S.Geological Survey Circular 903. (1983)

4. W. C. O'NEAL AND OTHERS, "Preclosure Analysis of Conceptual Waste Package Designs for a Nuclear Waste Repository in Tuff", UCRL-53595, Lawrence Livermore National Laboratory (1984)

5. DEPARTMENT OF ENERGY, "Site Characterization Plan, Yucca Mountain Site, Nevada Research and Development Area, Nevada," DOE/RW-0199, Chapter 8 (1988)

6. M.O. CLONINGER, D. SHORT, D. STAHL, "Waste Package for Yucca Mountain Repository: Strategy for Regulatory Compliance", Waste Management '89 Proceedings, Vol 1, p 537 (1989)

7. LEIF G. ERIKSSON, "Is it time to reduce the heat on the concept of geologic disposal of high-level radioactive waste?", American Underground-Space Association News, Vol 5, No.1, p 18, (April 1990)

8. LEIF G. ERIKSSON, DAVID L. PENTZ, "Natural System Issues in the OCRWM Program" in High Level Radioactive Waste Management: Proceedings of the International Topical Meeting, Las Vegas, Vol 1, pp 10-19, (April 1990)

9. NUCLEAR WASTE TECHNICAL REVIEW BOARD, "First Report to the U.S. Congress and the U.S. Secretary of Energy", U.S. Govt Printing Office, (March 1990)

10. DEPARTMENT OF ENERGY, "Site Characterization Plan", Chapter 7, pp. 7-40 to 42

11. JOHN J. NITAO, "Numerical modeling of the thermal and hydrological environment around a nuclear waste package using the equivalent continuum approximation: horizontal emplacement", UCID-21444, Lawrence Livermore National Laboratory (May 1988)

12. ABELARDO L. RAMIREZ AND OTHERS, "Prototype Heater Test of the Environment Around a Simulated Waste Package", High Level Radioactive Waste Management: Proceedings of the International Topical Meeting, Las Vegas, Vol 2, p 870, (1990)

13. THOMAS A. BUSCHECK, JOHN J. NITAO, "Modeling Hydrothermal Flow in Variably Saturated, Fractured, Welded Tuff During the Prototype Engineered Barrier System Field Test of the Yucca Mountain Project", presented at Lawrence Berkeley Laboratory TOUGH Workshop, September 1990, to be published as a Lawrence Livermore National Laboratory UCID (1990)

14. R. D. McCRIGHT, W. G. HALSEY, R. A. VAN KONYNENBURG, "Progress Report on the Results of Testing Advanced Conceptual Design Metal Barrier Materials Under Relevant Environmental Conditions for a Tuff Repository", UCID-21044, Lawrence Livermore National Laboratory (1987)

15. J. C. FARMER, R. A. VAN KONYNENBURG, R. D. McCRIGHT, D.B. BULLEN, "Survey of Degradation Modes of Candidate Materials for High-level Radioactive Waste Disposal Containers: Localized Corrosion and Stress Corrosion Cracking of Austenitic Alloys", UCID-21362 Vol.3, Lawrence Livermore National Laboratory (1988)

16. G.E. GDOWSKI, R. D. McCRIGHT, "Degradation Mode Surveys of High Performance Candidate Container Materials", Proceedings of International High Level Radioactive Waste Management Conference, Las Vegas, American Nuclear Society (1991)

17. Kärn-Bränsle Säkerhet (KBS), "Safe Handling and Storage of High-Level Radioactive Waste", KBS I ,Swedish Nuclear Fuel Supply Co, (1979)

18. G. L. JOHNSON, "Thermal performance of a buried nuclear waste storage container storing a hybrid mix of PWR and BWR spent fuel rods", UCID-21414, Lawrence Livermore National Laboratory (1988)

19. R.E. WOODLEY, R. E. EINZIGER, H.C. BUCHANAN, "Measurement of the Oxidation of Spent Fuel Between 140° C and 225° C" Nucl. Technology, 85, 74, (1989)

20. G. DANKO, P. MOUSSET-JONES, "The analysis of horizontal cooling enhancement for nuclear waste container emplacement", Proceedings Intl. High Level Radioactive Waste Management Conf., Las Vegas, American Nuclear Society (1990)

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