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Dr. D. J. Brooks
Geotechnical Branch
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U.S. Nuclear Regulatory Commission
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Washington, D.C. 20555

Dear Dave:

Enclosed is a copy of Gary Jacobs trip report to attend the Penrose Conference on "Geochemistry of the Environment near a High-Level Nuclear Waste Repository," (MR-5.3), in Welches, Oregon, on September 9-14, 1984.

Sincerely,

Susan K. Whatley, Program Manager
Engineering Analysis and Planning
Chemical Technology Division

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WM-RES

WM Record File	WM Project
<u>B0287</u>	<u>10, 11, 16</u>
<u>ORNL</u>	Docket No. _____
	PDR <input checked="" type="checkbox"/>
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MEETING REPORT

AUTHOR: Gary K. Jacobs

LOCATION: Rippling River Resort, Welches, Oregon

DATE: September 9 - 14, 1984

PURPOSE: Penrose Conference on "Geochemistry of the Environment near a High-level Nuclear Waste Repository"

PROJECT TITLE: Technical Assistance in Geochemistry

PROJECT MANAGER: Susan K. Whatley

ACTIVITY NUMBER: ORNL #41 37 54 92 4 (189 #B0287)
NRC #51 19 03 01

AGENDA: See attachment 1 (changes in agenda are indicated)

PARTICIPANTS: See attachment 2 (participants who did not attend are so marked)

GENERAL

The stated purpose of the Penrose conference is given in attachment 3. This description was prepared by the conveners and distributed prior to the conference.

Penrose conferences offer the opportunity to gather experts in the geosciences to discuss important topics in a semi-formal atmosphere. No proceedings are published and no cameras or recording devices are allowed to be used during the discussions. In this way, the participants are encouraged to present their most recent ideas which may not be completely thought out or supported by hard data. This format promotes the rapid exchange of information and generally benefits all participants. However, one drawback of this approach is that some important ideas may be lost during the discussion. I have listed observations that I made during the presentations and discussions. I stress, however, that these are my own personal interpretations and may not be completely accurate in all accounts - it is possible that I may have misunderstood a comment during the discussions. My observations on each of the sessions (see attachment 1) follow:

Geochemical Environment of a Repository:

The speakers presented a standard version of the "party line" for each of the candidate sites. No new information was presented and the speakers made no attempt to indicate possible areas of controversy or significant uncertainty. The discussions were so general that little discussion from the other participants was received.

Water Migration in a Thermal Field:

Byerlee presented a summary of his work on the measurement of permeability changes in rock cores in response to thermal and pressure gradients. At elevated temperatures (approximately 150 to 300°C), the permeability of rock cores decreased dramatically in relatively short times (hours to days). The mechanisms contributing to this decrease of permeability were not known precisely, although it was thought that dissolution and precipitation of silica and/or clays were involved. The effect was most pronounced in rocks with low initial permeability. Discussion centered on the applicability of this mechanism and technique to repositories, because repositories will see gradients only for short periods of time and the gradients will generally be much smaller than those investigated by Byerlee. It was generally agreed that this effect of decreased permeability could be important to repositories, although the techniques of Byerlee may not be directly applicable.

Norton presented an excellent overview of the role of temperature, fluid, and rock characteristics on the generation of fractures and fluid flow in hard rocks. He illustrated how small increases in ambient temperatures can significantly increase the permeability of rocks through increased crack growth, thus altering the characteristics of fluid flow in the system. He emphasized that these effects may not be large for a repository, but that this has yet to be demonstrated and needs to be addressed. Discussion was limited and was interpreted to indicate general agreement with his approach and suggestions.

Experimental Considerations:

Barnes presented an excellent discussion of possible experimental approaches to the problem of near-field geochemistry. His discussion centered on characterizing the geochemical aspects of the near field and not on the behavior of radionuclides because of his limited background in radiation chemistry. He did stress however, that once the role of radiolysis in groundwater and radiation damage to solids was defined, these effects must be addressed in evaluating the geochemistry. Barnes' major thrust was that the sites must obtain sound kinetic and equilibrium data to predict the behavior of the repository systems. His emphasis resulted from two premises: (1) reactions must be accelerated in the laboratory and extrapolated over long periods of time, and (2) a 1:1 scale model of a repository is impossible to construct and monitor. Therefore, a fundamental understanding of the behavior of the repository system is necessary to accelerate and extrapolate short-term laboratory results to repository-scale space and time. Without basic kinetic and equilibrium

data, such predictions and extrapolations will be highly uncertain. For example, a site which anticipates taking credit for the presence of smectites must show through careful kinetic determinations that the conversion of smectite to illite in the presence of small amounts of K and Al is slow enough so as to not compromise the performance of the system. The only way results from short-term laboratory tests on this reaction can be extrapolated with reasonable assurance is to determine the reaction mechanism.

Seyfried discussed his efforts in trying to experimentally model basalt-seawater interactions representative of spreading centers and seafloor hydrothermal systems. His discussion illustrated the usefulness of the Dickson-type autoclave in elucidating reactions in this complex system at elevated temperatures and pressures. He did stress, however, that he agreed with Barnes that the complex system experiments will not provide all the answers and that results from such experiments should be used to identify key reactions requiring more quantitative assessments in simple systems in order to adequately describe the behavior of any complex system.

Holloway presented a summary of his work for BWIP looking at the systems basalt-water-supercalcine and basalt-water-borosilicate glass. As with the presentation of Seyfried, Holloway showed how significant amounts of information can be derived from these "dirty bucket" experiments in a relatively short amount of time. As with Seyfried, however, he stressed the importance of establishing a basic understanding of the processes involved in order to make confident predictions and extrapolations. Another point made by Holloway is that the solids need to be evaluated as a function of time just as the solutions are in most hydrothermal tests, because the phase assemblage observed at the end of the test may not be the same as the assemblage present during the initial portions of the test. Information such as this would aid in interpreting the results from the solution analyses. The audience was in general agreement with this suggestion.

Potter discussed the role of flow-through autoclaves in hydrothermal testing. The basic premise in this approach is that, by increasing the flow rate, reactions may be accelerated and more stable phase assemblages may be produced in shorter periods of time than in conventional autoclave tests, e.g., Dickson-type and Barnes-type rocking autoclaves. This technique is still in its early stages of use and there are still some questions to be ironed out. For example, the flow-through autoclave is essentially an open and undefined system - making thermodynamic assessments of the results difficult. In addition, because the sample of solution from the outlet to be analyzed is a bulk solution averaged over time, less information can be derived from the solution than in conventional tests. However, the advantages gained by the acceleration of the growth of stable phase assemblages may offset this problem. In addition to these questions, it was brought up that for a repository, it may be more appropriate to deal with metastable assemblages rather than accelerate the formation of stable assemblages, because metastable assemblages are more likely to form during the early periods of repository history.

The behavior of these metastable phases may be essential to the performance of the repository if radionuclides are incorporated into their structure. This area of flow-through testing has tremendous potential and may represent an important area in future experimental techniques for repository testing.

Runnells laid to rest the issue of the measurement of Eh in dilute low temperature groundwaters. He used material from his Science article (Science, v. 255, p. 925, 1984) to illustrate how essentially all redox couples are in disequilibrium - thus negating the basic premise of equilibrium in the concept of Eh measurement. It was noted from the floor that in natural hydrothermal systems (150 to 300°C) equilibrium among redox couples is generally ubiquitous. Therefore, the concept of Eh and possibly the measurement of it may be valid for portions of repository history. However, it is apparent that careful determinations of the role of kinetics in the equilibration of redox couples as a function of temperature may be necessary. Runnells also pointed out that in evaluating the behavior of a redox sensitive radionuclide in a groundwater, one must determine which redox reactions are active and not just consider some overall bulk redox potential of the solution. It is likely that specific reactions will govern the behavior of the radionuclide and these must be identified to be able to predict and extrapolate radionuclide behavior. Runnells briefly discussed some new work he is starting in the area of scanning voltammetry. This is not a new area, but has not been typically applied to geochemical systems. This is a method of determining the reversibility and kinetics of redox couples. Runnells has begun studying some common systems and hopes to identify some ranges of conditions where the redox couples are reversible. Some possible extensions he discussed included: establishing the active redox reactions in a system of radionuclide + redox couples; evaluate the redox behavior of a rock + water system; look at the importance of bacteria to the reversibility of several redox couples; determine the kinetics of redox reactions as a function of temperature. This area of scanning voltammetry may be able to shed some light on the behavior of some of the more complex and unpredictable radionuclides which are redox sensitive.

Ulmer discussed the current state of development of electrodes for the measurement of pH and Eh at elevated temperatures and pressures. Progress is being made, but no reliable "stock" item is available - it is likely that these measurements will continue to be "research" efforts for some time. The audience was in agreement that this work is important and needs to be continued so that a working electrode can be developed.

Grandstaff discussed techniques for estimating Ph and Eh at elevated temperatures and pressures. Basically, a thermodynamic approach can be successful in systems where the temperature is high enough to warrant the assumption of equilibrium, e.g., 150 - 300°C. Grandstaff also compared the results from basalt/water hydrothermal tests to the geothermal systems located in the basalts of Iceland. The agreement in solution chemistry, pH, and Eh was remarkable. The audience was in general agreement with the techniques and results discussed by Grandstaff.

Field Trip:

A guide book for the trip is available and if anyone is interested in looking at it, please contact me. The field trip was general in nature and few details of hydrothermal alteration were observed or discussed.

Theoretical Considerations:

Giggenbach presented a summary on the use of activity diagrams as applied to porphyry copper deposits. The emphasis was on the large amount of useful information which can be obtained through an equilibrium thermodynamic treatment of systems at elevated temperature. Discussion centered on the possibility that this approach is not valid for repositories where metastable states may persist for significant portions of repository history (which is generally much shorter than the life of most natural hydrothermal systems). Giggenbach was emphatic that the equilibrium treatment is a good starting point and should not be discarded so lightly at this early stage. The typical response from repository site representatives was to ask for an example specifically related to their system - they seemed to miss the point that Giggenbach was merely illustrating a technique which might be adopted by the sites, not to present results for the sites.

Dibble summarized his theoretical relationships contained in previous articles (Geochim. Cosmochim. Acta, v. 45, p. 79, 1981 and Clays and Clay Min., v. 29, p. 323, 1981). The theory of kinetic reaction models was applied to the results from flow-through tests discussed previously by Potter. In general terms, the theory and experimental results appear to be consistent. However, as discussed previously, there are many complexities involved in flow-through testing which have not been quantified adequately to allow definitive conclusions to be reached. It is clear, though, that this area of testing and theory will make an impact in future experimental programs.

Seward described the complexation, transport, and deposition of metals in hydrothermal systems of New Zealand. The purpose of the discussion was to illustrate one approach in attempting to understand the mechanisms involved in ore deposition. Seward pointed out that a combination of geologic characterization, thermodynamic calculations, and careful laboratory experiments is an accepted and well-established approach to solving problems in hydrothermal systems. He emphasized that this is the direction that the sites should be taking, although he recognized the importance of kinetic processes when dealing with a system likely to contain significant quantities of metastable phases.

Natural Analogues:

The three presentations on natural analogues (Capuano, Elders, and Cummings) discussed three separate occurrences of active hydrothermal systems. None of the talks were specifically geared toward repositories, but rather were examples of how to approach a problem. Discussion was lively during this session. The two applications of natural analogues

thought to be most useful to repository development were: (1) validation of geochemical, thermal, and transport computer models, and (2) gaining information on analogues for radionuclides in a natural geologic setting. It was generally agreed that natural analogues must be chosen carefully for their applicability and any conclusions drawn from such studies would be "soft", except in the area of model validation. Of course, validating a model for several natural analogues does not guarantee that the model is valid for a specific repository system. A plea from the floor was also made to ask the following question prior to jumping into any natural analogue study: What question related to repository performance will be answered by the study? Unless a clear goal and the relevance to repository behavior is established ahead of time, the usefulness of the natural analogue study may be limited.

Wrap-up Session:

This session was disappointing. No summary of the previous sessions was given to help focus discussions. It was my impression that because no recommendations or summary of the conference was to be released, many people did not actively participate because they felt these discussions would have little impact on the sites, DOE, or NRC. However, there did seem to be an overriding consensus that the repository program needs to identify an overall conceptual model of repository behavior which would help to focus on the most important questions to be answered. It was also perceived that modelers and experimentalists needed to work more closely together in attempting to identify key data needs as well to correctly interpret test results in light of conceptual models of repository behavior. It was the overall impression of the non-repository participants that the work currently being conducted is not directed toward problem solving, but rather is a series of unrelated studies of dubious quality proceeding in a manner which will not produce useful and defensible information in a timely manner.

CONFERENCE SCHEDULEMonday, September 10, 1984

8:30 a.m.	Introduction to Conference	<u>D. G. Coles</u>
8:45	General Review of the Environment for a Saturated, Unsaturated, and Salt Repository from a Thermal- Hydrologic and Engineering Point of View	<u>J. O. Duguid</u>
9:30	Discussion	
10:00	Break	

Geochemical Environment of a RepositorySITE REPRESENTATIVES

10:20	• Basalt	J. Myers, BWIP
10:40	• Salt	T. L. Steinborn, ONWI
11:00	• Tuff	K. G. Knauss, NNWSI
11:20	• Granite	A. A. Bauer, OGRD
11:40	Discussion	J. O. Duguid
12:30	Lunch	

AFTERNOON FREE

Water Migration in a Thermal Field

7:00 p.m.	Porous Media	<u>J. D. Byerlee</u>
7:45	Discussion	
8:15	Fractured Media	<u>D. L. Norton</u>
9:00	Discussion	
10:00	End of Session	

Tuesday, September 11, 1984

Experimental Considerations

8:00 a.m.	Overview of Hydrothermal Experimentation	<u>H. L. Barnes</u>
8:45	Discussion	
9:15	Use of Sampling Hydrothermal Apparatuses to Assess Natural Hydrothermal Processes	<u>W. E. Seyfried, Jr.</u>
10:00	Discussion	
10:30	Break	
10:50	Use of Sampling Hydrothermal Apparatuses Applied to Nuclear Waste Testing and Natural Systems	<u>J. R. Holloway</u>
11:35	Discussion	
12:00	Lunch	

Experimental Considerations, continued

1:15 p.m.	Use of Flowing Autoclave Experiments for Investigating Water/Rock Interactions and Nuclear Waste Package Interactions	<u>J. Potter</u>
2:00	Discussion	
2:30	Theory, Interpretation and Problems Inherent in the Concept of Eh	<u>D. D. Runnells</u>
3:15	Discussion	
3:45	Break	
4:05	Control and Monitoring of Eh and pH in Hydrothermal Experimentation	<u>G. C. Ulmer/ D. E. Grandstaff</u>
4:50	Discussion	
5:30	End of Session	

Wednesday, September 12, 1984

Field trip to Ruth Mine area of the North Santiam Mining District of the Western Cascades of Oregon to investigate the character and controls on hydrothermal alteration.

M. L. Cummings and
J. M. Pollock

Thursday, September 13, 1984

8:00 a.m.	Low Temperature Diagenetic/ Metamorphic Reactions	<u>W. F. Giggenbach</u>
8:45	Discussion	
9:15	Kinetic Reaction Models Applied to Hydrothermal Reactions Within a Nuclear Waste Repository	<u>W. E. Dibble, Jr.</u>
10:00	Discussion	
10:30	Break	
10:50	Geochemistry of Metal Corrosion and Transport	<u>T. M. Seward</u>
11:35	Discussion	
12:00	Lunch	
AFTERNOON FREE		
6:15 p.m.	Hydrothermal Systems-Analogies to Nuclear Waste Repositories	<u>R. M. Capuano</u>
7:00	Discussion	
7:30	A Natural Analogue of a Salt Repository, the Salton Sea Geo- thermal Field, Imperial Valley, California	<u>W. A. Elders</u>
8:15	Discussion	
8:45	Mineralogy and Geochemistry of Hydro- thermal Alteration at Glass Buttes, South Central Oregon - a Potential Analogy for a Basalt Repository	<u>M. L. Cummings</u>
9:30	Discussion	
10:00	End of Session	

Friday, September 14, 1984

8:30 a.m. Wrap-up Session

12:00 Lunch

2:00 - Informal discussion for participants with late
4:00 airline schedules

Field Trip

The field trip is being lead by Dr. Michael Cummings and graduate student Mike Pollock, both from Portland State University. The route for this trip will be south along the west side of the Cascades to the North Santiam River mining district. Mike Pollock is currently doing field work in this area and will show the conference participants some interesting hydrothermal ore deposits. The conference focuses on the similarity between natural hydrothermal geochemistry and that induced by a nuclear waste repository. The field trip will allow participants to investigate an example of a natural hydrothermal area. The round-trip distance is about 300 miles through some of Oregon's most beautiful mountain scenery. Vans will be required for transportation because the roads near the field area are not wide enough for buses.

Location

The conference site we have selected is the Rippling River Resort located on the southwest flank of Mount Hood in northern Oregon. The resort is located 35 miles east of Portland, Oregon at Welches, OR. It has new meeting and convention facilities. It is a place of beauty and of considerable interest to the geologist and geochemist.

A professional meeting coordinator (Lois Elms) has been contracted by GSA assist us in setting up the conference.

Date

The date of the conference is September 9-14, 1984.

Number of Participants

The number of participants to the conference will be limited to about 70, including the speakers.

Sponsors

The NRC, DOE's Pacific Northwest Laboratory, and the DOE Office of Basic Energy Science provided funds to help defray the cost of this conference.

The co-sponsors recognize that no publications will result from this conference and that the content of the conference will be solely determined by the conveners, GSA, and the speakers.

Conveners

David G. Coles	-	Pacific Northwest Laboratory
Michael J. Apted	-	" " "
Floyd N. Hodges	-	" " "
Donald H. Alexander	-	Department of Energy/Headquarters

Coles, Hodges, and Alexander are members of GSA.

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"Geochemistry of the Environment Near a High-Level
Nuclear Waste Repository", September 9-14, 1984

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Description of a Penrose Conference Entitled:

"Geochemistry of the Environment Near a
High-Level Nuclear Waste Repository"

Purpose

The science of permanent disposal of high-level nuclear waste in geologic repositories has been evolving at an accelerating pace since the mid-seventies. Nuclear-waste disposal is a problem of concern for the public and a technical challenge to the scientific community because of the need to assure safe isolation of disposed nuclear waste for thousands of years. Only during the last few years has the emphasis shifted from engineering aspects of waste disposal to geologic/geochemical aspects. That emphasis is increasing because an understanding of the geology and geochemistry of waste disposal is essential for not only establishing the safety of nuclear waste storage but also for predicting the long-term (>100's of years) behavior of the waste, and hence it's integrated probability for returning to the biosphere, during the long-time period before nuclear decay reactions alleviate the radiologic hazard.

It is the opinion of the conveners that much expertise relevant to understanding waste disposal exists within the geoscience community and that much of this expertise may not have yet been applied directly to the waste disposal problem. Geoscience disciplines that are applicable to studying the area of the repository/waste package include field studies of low-grade metamorphic rocks, laboratory investigations of hydrothermal reactions between rocks and fluids, reactions and transport mechanisms for hydrothermal ore deposits, clay stability, metallic corrosion in a geological environment, water migration in a thermal field, solution chemistry effects (i.e., pH, Eh, complexants) on mineral phase stability--particularly radionuclide-bearing phases, expermental determination of sorption and solubilities of radionuclides under geological conditions, and colloid formation as a potential nuclide transport mechanism.

The emphasis on hydrothermal conditions is because the thermal load caused by the waste emplacement will initially raise temperatures up to ~300°C during its first few hundred years after disposal and because of the need to

accelerate otherwise sluggish reactions in the laboratory. The repository zone immediately surrounding and including the waste is emphasized due to the need for understanding what constitutes the source for radionuclide release. Can the complex series of reactions and consequent formation of alteration and corrosion products be simulated in laboratory tests? How can such test data be meaningfully extrapolated over time scales approaching hundreds of thousands of years? What confidence can be placed on such results and can these extrapolations be made with reasonable assurance? Are there dominant processes that will control the release and migration of radionuclides? What are the state-of-the-art test methods that can be used to obtain the most relevant and defensible data on expected rock/waste package interactions? These are some of the questions currently facing the waste disposal community.

The purpose of the conference is to assemble a group of speakers who have expertise in the experimental and theoretical technologies related to nuclear waste disposal but who may have never applied their expertise to the problem of nuclear waste isolation. Other attendees would be expected to have research interest or experience in the technology of nuclear waste disposal. Such a group of geoscientists would promote considerable technical exchange. The waste disposal community would benefit by gaining new ideas and techniques for tackling their extremely complex experimental problems. The speakers would benefit by having the opportunity to meet fellow scientists who work in the waste community, by having an opportunity to apply their skills to a new field of research, and by contributing to the solution of an international problem. The NRC, as well as the DOE, would benefit by assuring themselves that theoretical and experimental approaches pertinent to the goals of safe nuclear waste disposal in geologic repositories could be identified early in order to obtain consensus with expert professional judgement and to assure the highest technical quality to the data to be used in the federal licensing phase of a waste repository. An absolute criterion for this conference is that it is technical and not programmatic. Participants will be chosen with strong consideration of this proviso.