WM-RES
WM Record File
B
ORN

WM Project 10, 11, 16
Docket No. ______
PDR ✓
LPDR B, N, S

Distribution:
______
(Return to WM, 623-SS)
L3

DISTRIBUTION
WM s/f (B0287)
WMGT r/f
NMSS r/f
REBrowning
MBell
JBunting
MRKnapp
DJBrooks & r/f
RJStarmer
PDR
LPDR (B,N,S)

426.1/DJB/85/03/04

MAR 14 1985

Susan K. Whatley, Manager
Engineering Analysis and Planning
Chemical Technology Division
Oak Ridge National Laboratory
P.O. Box X
Oak Ridge, TN 37830

Dear Ms. Whatley:

SUBJECT: CONTRACT NO. NRC--50-19-03-01, FIN B-0287, ORNL NO. 41-47-54-92-4, "TECHNICAL ASSISTANCE IN GEOCHEMISTRY," JANUARY, 1985, MONTHLY PROGRESS REPORT

I have reviewed the January monthly progress report dated 2/11/85. Based on my review, I have the following comments:

Task 1, 2, and 3 - BWIP, NNWSI and Salt Geochemical Technical Assistance

- Progress to date is satisfactory.
- We received ORNL comments on the BWIP DEA, the NNWSI DEA and seven salt DEAs. Your quick response was appreciated. Your comments were considered and factored into our draft comments as appropriate.
- A draft of our comments for each of the sites will be sent to you during the week of March 18, 1985.

Task 4 - Short-term Geochemical Technical Assistance

- Progress to date is satisfactory.
- During the week of January 29, 1985, Jim Blencoe, Gary Jacobs and Don Kelmers provided a valuable review of our draft EA comments. Gary Jacobs' follow-up letter report on solubility calculations was appreciated.

Task 5 - Project Management

- Progress to date is satisfactory.
- In your next monthly report, please let me know the progress of our planned work on anologs.

8504090075 850314
PDR WMRES EXIORNL
B-0287 PDR

FC	:WMGT	:	:	:	:	:
AME	:DJBrooks;mt	:	:	:	:	:
ATE	:85/03/	:	:	:	:	:

1956

- The critical review of the site documents in the ORLOOK data base is of very high priority this year. Attachment 1 is a review of BWIP document "...SA-219P." Please have this information incorporated into the data base. Also, from a scientific (technical) standpoint, the attached review is a good example of the kind of critical review that is needed. Our reviews need to focus on the weaknesses or over-simplifications of the results being published by the individual sites. Please distribute the attached review for information, and as an example of what is needed. It is my goal that the data base output for each document would serve as a mini "technical position" on the data presented, the data collection methods used, interpretations and conclusions.

The action taken by this letter is considered to be within the scope of the current contract (NRC-50-19-03-01/FIN B-0287). No changes to costs or delivery of contracted products is authorized. Please notify me immediately if you believe this letter would result in changes to costs or delivery of contracted products.

Sincerely,

Original Signed By

David J. Brooks
Geochemistry Section
Geotechnical Branch
Division of Waste Management
Office of Nuclear Material Safety
and Safeguards

Enclosures:
As Stated

FC	WMGT					
AME	DJBrooks;mt					
ATE	85/03/13					

Attachment -1

REVIEW OF RHO-BW-SA-219 P, Experimental Investigation of Sodium Bentonite Stability. . ., M. I. Wood

This brief review is intended to address the simplistic assumptions made by the author of this report rather than raise specific criticisms already made by R. A. Couture in a separate review. Wood reports on 1-3 month experiments on a bentonite at 300°C and 0.3 Kb in which he notes little change in the clay mineralogy but documents the approach of synthetic groundwater compositions to a steady state. The experiments provide valuable data on solution changes with time. However, Wood concludes without justification that bentonites will be stable as a backfill in a nuclear waste repository as will be discussed below.

First, smectites are *not* stable at 300°C for geological times. Many studies of natural clays metamorphosed to 200-300°C (the temperatures of prehnite-pumpellyite to lower greenschist facies metamorphism) show loss of smectites with formation of micas, feldspars and/or chlorites depending on the chemistry of the smectite precursor (Winkler, 1979; Kisch, 1980, 1981; Turner, 1981). Similar observations are available for hotsprings metamorphic systems which were subjected to pressures similar to Wood's experiments (McDowell and Elders, 1980; Henley and Ellis, 1981). While it is difficult to judge the exact time-temperature relations in metamorphic systems, geologic observations clearly indicate that smectites are not stable above 200°C and perhaps not above 150°C, in direct conflict with Wood's conclusions.

Secondly, the transformation of complex clay minerals to illite or white mica and other phases, is an irreversible, kinetically controlled process dependent at least on time, temperature, groundwater chemistry and the kinetics of nucleation, growth and dissolution (e.g. Hower et al., 1982; Lee et al., 1982). Therefore short-term experiments dealing with the stability of phases must be interpreted with extreme caution. Certainly the rate-controlling activation energy in terms of the reconstructive transformations involved will vary strongly as time varies by 10^5 (3 months to 3000 years). This is especially true insofar as the smectite to illite transformation includes two important aspects other than simple replacement of interlayer cations by potassium. These are: (1) substitution of Al for Si in tetrahedral sites, and (2) reorganization and annealing of structure layers from highly imperfect layers in smectite to relatively defect-free layers in illite. Both of these factors involve the breaking of Al- and Si-0 bonds and diffusion of those cations, in part. These factors are well known to inhibit reaction and to prevent attainment of equilibrium at low temperatures. It is not surprising, therefore that three month experiments show little change in the materials.

Thirdly, the steady-state approach of the interstitial solutions in Wood's experiments *in no way* indicates anything about the stability of the bentonite itself. One would expect a new composition solution to be attained in equilibrium with illite. Indeed the solution may be buffered by interstitial glass instead of the bentonite.

Fourthly, Wood's own experiments may be used to demonstrate that bentonite is beginning to react in three months because he observes growth of new albite. This suggests that bentonite is reacting to yield albite + chlorite ± paragonite ± illite ± quartz [redacted] + H_2O, a typical low-grade metamorphic assemblage. The new sheet silicates may well be "hidden" as mixed layers in the reacting bentonite. The observance of even 0.1% reaction in three months is extremely critical. For instance if this amount of reaction were to proceed at a constant rate, all bentonite would be gone in 100 years. As the water molecules are released significant shrinkage, *not* expansion, would occur. The observation of neomorphic albite suggests that sodium bentonite is *not* stable at 300°C for 100 year periods of time.

REFERENCES

Henley, R. W. and A. J. Ellis (1983) Geothermal systems ancient and modern: a geochemical review. Earth Sciences Reviews 19, 1-50.

Hower, J., S. A. Attaner, J. L. Aronson, and C. G. Whitney (1982) Kinetics and compositional controls of the smectite to illite reaction. Geol. Soc. Amer. Abstr. w. Prog. 14, 519.

Kisch, H. J. (1980) Illite crystallinity and coal rank associated with lowest-grade metamorphism of the Taveyanne greywacke. Ecologae Geologicae Helvetiae 73, 753-777.

__________ (1981) Coal rank and illite crystallinity associated with the zeolite facies of Southland and the pumpellyite-bearing facies of Otago. N.Z. Jour. Geol. Geophys. 24, 349-360.

Lee, J. H., D. R. Peacor, and R. P. Wintsch (1982) TEM and AEM study of interlayering and along-layer transitions in tri- and di-octahedral phyllosilicates. Geol. Soc. Amer. Abstr. w. Prog. 14, 544.

McDowell, S. D. and W. A. Elders (1980) Authigenic layer silicate minerals in Borehole Elmore 1, Salton Sea geothermal field, California. Contr. Min. Pet. 74, 293-310.

Turner, F. J. (1981) Metamorphic Petrology (2nd edition). McGraw-Hill, N.Y.

Winkler, H. G. F. (1979) Petrogenesis of Metamorphic Rocks (5th edition). Springer-Verlag, N.Y.