

# Evaporation of J13 and UZ Pore Waters at Yucca Mountain

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# Evaporation of J13 and UZ pore waters at Yucca Mountain

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## Introduction

This work is motivated by a need to characterize the chemistry of aqueous films that might form at elevated temperatures on engineered components at the potential high-level, nuclear-waste repository at Yucca Mountain, Nevada. Such aqueous films might form through evaporation of water that seeps into the drifts, or by water vapor absorption by hygroscopic salts directly deposited on these components (possibly from previous evaporation events or possibly from air-blown particles drawn into the drifts through a drift ventilation system).

There is no consensus at this time on the chemical composition of water that might come in contact with engineered components at Yucca Mountain. Two possibilities have received the most attention: well J13 water and pore waters from the unsaturated zone (UZ) above the repository horizon. These waters represent the two major types of natural waters at Yucca Mountain<sup>1</sup>. Well J13 water is a dilute Na-HCO<sub>3</sub>-CO<sub>3</sub> water, representative of regional perched water and groundwater. The UZ pore waters are Ca-Cl-SO<sub>4</sub>-rich waters with a higher dissolved ion content. These waters are less well-characterized.

We have studied the evaporative evolution of these two major types of waters through a series of open system laboratory experiments, with and without crushed repository-horizon tuff present, conducted at sub-boiling temperatures (75°C-85°C).

## Work Description and Results

Our experiments are based on synthetic waters representative of Yucca Mountain well J13 and UZ pore waters<sup>2,3</sup>. Details about the experimental methods are described elsewhere<sup>4</sup>. The solids acquired during these experiments were analyzed using standard X-ray diffraction (XRD) methods. The water chemistry results of these experiments are shown in Figs. 1-2.

*J-13 water* - The data clearly indicates loss of Ca, Mg and HCO<sub>3</sub> from solution as carbonates, with Mg likely going into a slightly disordered CaCO<sub>3</sub> phase. The pH stabilizes at about 10 after an approximately 50-fold concentration. Evaporation has produced an alkaline Na-HCO<sub>3</sub>-CO<sub>3</sub> brine. The presence of tuff had no significant, discernable effect on the water composition. (These reactions may become important in longer-term, higher-temperature evaporation

experiments.) The dissolved  $\text{SiO}_2$  remains high in these solutions, probably due to the high pH of the evaporated solution.

The minerals recovered after complete evaporation of the J13 water alone experiments contain amorphous silica, aragonite, calcite, halite, niter, thermonatrite and, possibly, gypsum, anhydrite and hectorite. After “1000x” concentration, the only minerals present were amorphous silica, aragonite and calcite. The XRD analysis of the solids sampled from the tuff plus water run after “1000x” concentration show the presence of trona, thermonatrite, halite, anhydrite, smectite and niter in addition to amorphous silica, aragonite, calcite. The minerals recovered after complete evaporation contain amorphous silica, aragonite, calcite, trona, thermonatrite, halite, anhydrite and smectite. The clays found in the 1000-fold sample were separated and found to be a mixture of dioctohedral smectite (probably montmorillonite) and trioctohedral smectite (hectorite, stevensite or saponite).

*UZ pore water* - After concentration approximately 1000-fold, the solution has lost Ca and  $\text{SO}_4$  suggesting removal of these species as gypsum, which is in agreement with the XRD data. Aqueous concentrations of Mg,  $\text{HCO}_3$ , F and  $\text{SiO}_2$  also decrease with evaporation. Carbonates are expected to precipitate with increasing evaporation, however because of the limited amount of  $\text{HCO}_3$  present initially, these carbonates may not be present in quantities significant enough to be detected by the XRD analysis. In contrast with the J13 water evaporation experiments, there is a large loss of  $\text{SiO}_2$  from these near-neutral pH solutions. The pH initially increases to about 9 and then decreases to values between about 5.5-6.5. The initial pH increase may be due to the  $\text{HCO}_3$  content increase until precipitation of carbonate species removed it from solution. Evaporation of this water has produced a Na-K-Ca-Mg-Cl- $\text{NO}_3$  brine. As with the J13 waters, the presence of tuff has no significant, discernable effect on the water composition.

The minerals recovered after complete evaporation of the UZ pore water alone experiments include tachyhydrite in addition to gypsum. Other mineral phases may be present but in amounts too low to be detected by this analysis. The XRD analysis of the solids sampled from the tuff plus water experiment show the presence of halite in addition to gypsum. The minerals recovered after complete evaporation contain a Mg-smectite and possibly kenyaite as well. We note that the hygroscopic nature of the salts formed in the UZ pore water experiments made them difficult to analyze.

## Conclusions and Discussion

These experiments indicate the evolution of high pH, Na- $\text{HCO}_3$ - $\text{CO}_3$  brines from water representative of perched water and groundwater (e.g., J13 well water). We expect similar behavior for nearly all perched water or groundwater at Yucca Mountain. These experiments indicate the evolution of near-neutral pH brines from UZ pore water from the units above the potential repository horizon. We expect that the evaporative evolution of the UZ pore waters will be more sensitive to variability/uncertainty in initial water composition, specifically, the ratio of Ca to  $\text{SO}_4$ . Because solution chemistry is sensitive to  $\text{CO}_2$  fugacity and  $\text{CO}_2$  levels in emplacements drifts at Yucca Mountain may be very different from atmospheric levels, at least transiently, open system experiments with a variety of  $\text{CO}_2$  gas levels may be of interest. In general, we expect that pH will increase as  $\text{CO}_2$  fugacity decreases and decrease as  $\text{CO}_2$  fugacity increases.

These experiments address a subset of relevant thermohydrologic-chemical conditions that might exist in a Yucca Mountain repository environment. One of us (Gdowski) has recently completed experiments that consider other possible conditions; we hope to report on these results in the near future. These experiments include studies of the evaporative evolution of J13 water at or near the boiling point of the concentrated solution (up to 112°C) and the evaporative evolution of J13 water at high-temperature, low-*RH* conditions (85-110°C, *RH* 50-90%).

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## Figure Legends

Fig. 1. Ratio of major ion concentration ratio to Cl concentration ratio for J13 and UZ pore water experiments. The bars are, from left to right, for the water alone experiments (“100x”, “1000x”, rewet) and water + tuff experiments (“1000x”, rewet).

Fig. 2. pH vs. concentration for evaporation experiments.

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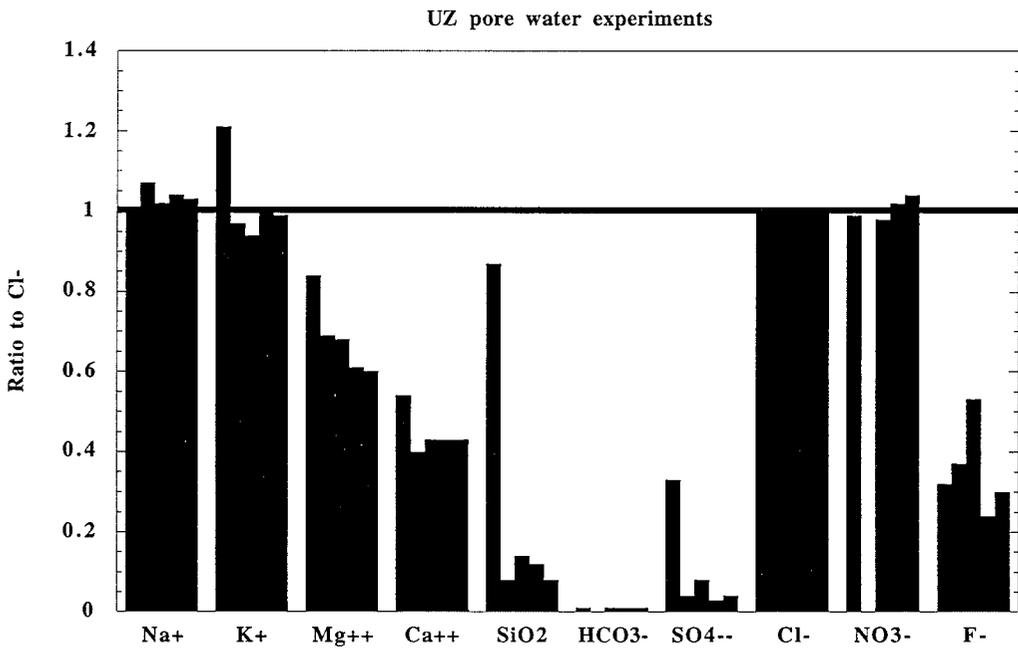
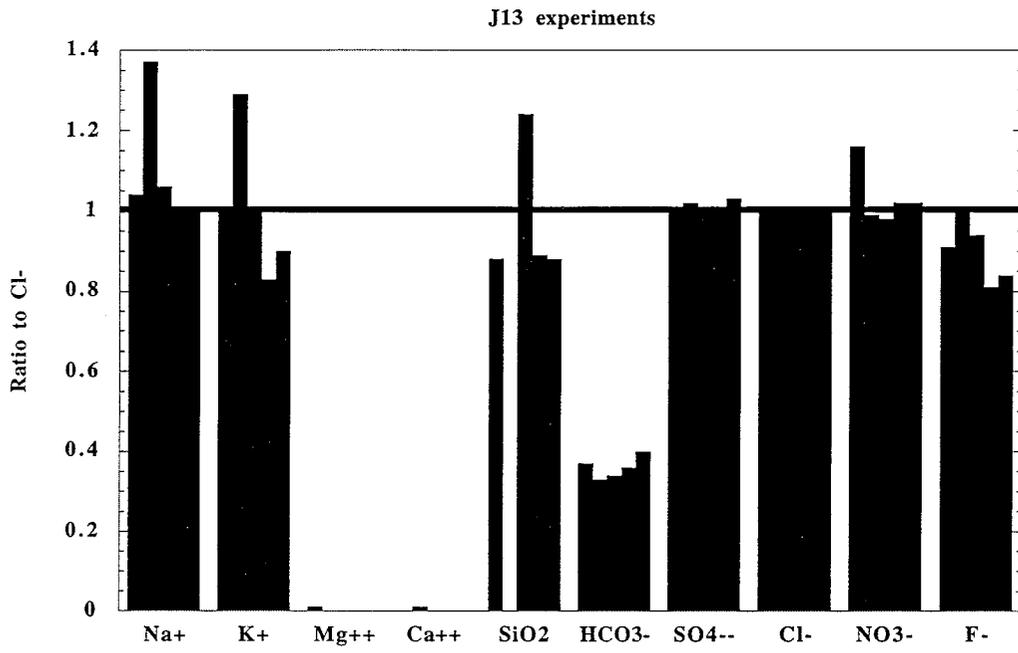


Figure 1

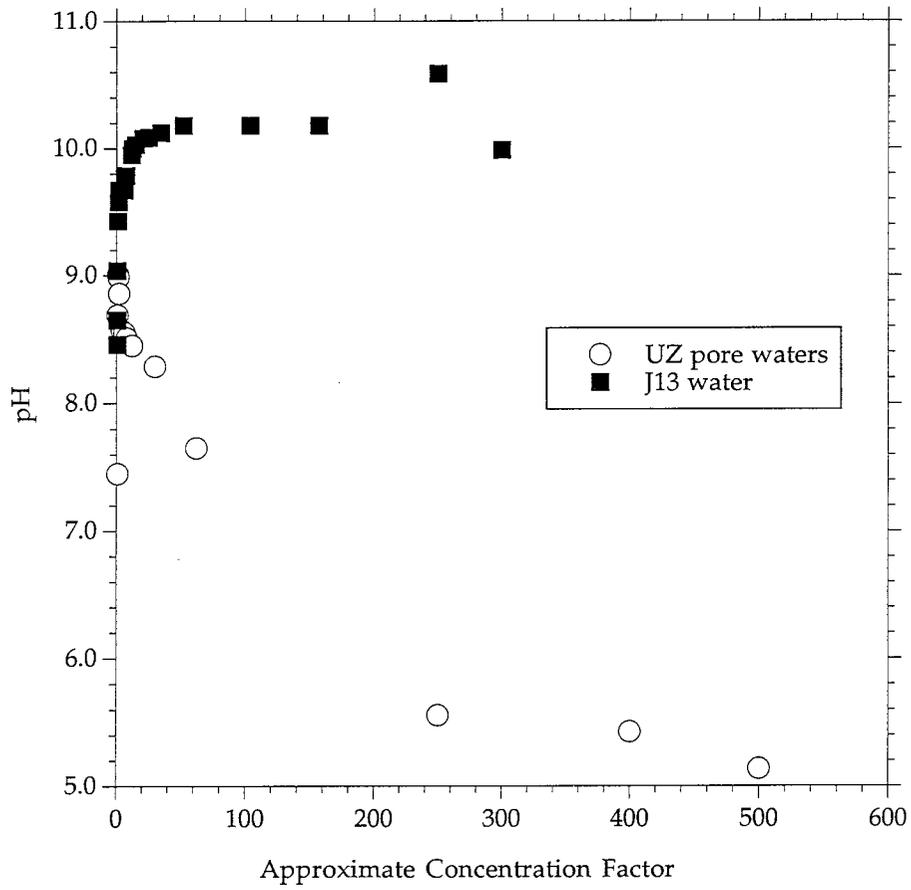


Figure 2

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