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Fluid-Mineral Equilibria in a  
Hydrothermal System, Roosevelt Hot Springs, Utah

by

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## ABSTRACT

The availability of fluids and drill cuttings from the active hydrothermal system at Roosevelt Hot Springs allows a quantitative comparison between the observed alteration mineralogy and the predicted mineralogy calculated from fluid-mineral equilibria relationships. Comparison of all wells and springs in the thermal area indicates a common reservoir source. Geothermometer calculations on these fluids predict the reservoir temperature to be slightly higher, at  $288^{\circ}\text{C} \pm 10^{\circ}$ , than the maximum measured temperature of  $268^{\circ}\text{C}$ .

The composition of the deep reservoir fluid was estimated from surface well samples, taking into account the effects of steam loss, gas release, mineral precipitation and ground-water mixing in the well bore. This deep fluid is sodium chloride in character, with approximately 9700 ppm total dissolved solids, a pH of 6.0, and gas partial pressures of  $\text{O}_2$  ranging from  $10^{-32}$  to  $10^{-35}$  atm,  $\text{CO}_2$  of 11 atm,  $\text{H}_2\text{S}$  of 0.020 atm and  $\text{CH}_4$  of 0.001 atm.

Comparison of the alteration mineralogy from producing wells 72-16 and 14-2 and nonproducing wells 52-21 and 9-1 allowed delineation of an alteration pattern characteristic of the reservoir rock. Theoretical alteration mineral assemblages in equilibrium with the deep reservoir fluid, in the temperature range from  $150^{\circ}$  to  $300^{\circ}\text{C}$  and in the system  $\text{Na}_2\text{O}-\text{K}_2\text{O}-\text{CaO}-\text{MgO}-\text{FeO}-\text{Fe}_2\text{O}_3-\text{Al}_2\text{O}_3-\text{H}_4\text{SiO}_4-\text{H}_2\text{O}-\text{H}_2\text{S}-\text{CO}_2-\text{HCl}$ , were calculated and compared with the observed alteration assemblages in the reservoir rock. Minerals theoretically in equilibrium with the calculated

reservoir fluid at  $>240^{\circ}\text{C}$  include sericite, K-feldspar, quartz, chalcedony, hematite, magnetite and pyrite. This assemblage corresponds with observed higher-temperature ( $>210^{\circ}\text{C}$ ) alteration assemblage in the deeper parts of the producing wells. The presence of montmorillonite and mixed-layer clays with the above assemblage observed at temperatures  $<210^{\circ}\text{C}$  corresponds with minerals predicted to be in equilibrium with the fluid below  $240^{\circ}\text{C}$ . Component activities calculated to correct for solid solution in sericite vary with depth and temperature and predict sericite equilibrium temperatures in agreement with those observed.

Alteration minerals present in the reservoir rock that do not exhibit equilibrium with respect to the reservoir fluid include epidote, anhydrite, calcite and chlorite. These may be products of an earlier hydrothermal event, or processes such as boiling and mixing, or a result of errors in the equilibrium calculations as a result of inadequate thermochemical data.

## INTRODUCTION

Active geothermal systems provide a unique glimpse at the chemical and physical processes that take place during hydrothermal alteration and the influence that variations in temperature, pressure and chemical composition of thermal fluid has on the formation of alteration minerals. Detailed studies of the relationship between fluid chemistry and alteration mineralogy in geothermal systems are, however, lacking. In part, this represents the unavailability of complete fluid analyses and of detailed petrologic studies for most geothermal systems.

Extensive exploration in recent years at Roosevelt Hot Springs has made the necessary data available to study these processes. Seven wells, up to approximately 2000 m in depth, currently tap thermal fluids (Fig. 1). Chemical analyses of fluids and petrographic analyses of drill cuttings from several of these wells have been described (Ballantyne and Parry, 1978; Ballantyne, G., 1978; Nielson et al., 1978; Parry, 1978; Rohrs and Parry, 1978; Glenn and Hulen, 1979; Bamford et al., 1980; Glenn et al., 1981). In most cases, however, these studies have focused on the individual wells rather than on the reservoir as a whole.

In this paper alteration mineralogy from the producing and nonproducing wells is compared and an alteration pattern characteristic of the reservoir rock is defined. The composition of the deep reservoir fluid is calculated from analyses of liquid and steam samples from production well 14-2. Corrections are made for the effects of ground-

water mixing, mineral precipitation, steam loss and gas release on the pH, gas partial pressures and element concentrations of the original reservoir fluid. Finally, mineral equilibria in the deep reservoir fluid are quantitatively evaluated at temperatures ranging from 150° to 300°C and compared to the alteration mineralogy of the reservoir rock.

#### GEOLOGY AND HYDROTHERMAL ALTERATION

Roosevelt Hot Springs thermal area, located in west-central Utah, covers approximately 32 sq km on the western margin of the Mineral Mountains (Fig. 1). The thermal reservoir occurs within fractured Precambrian gneisses and Tertiary granitic rocks of the Mineral Mountains pluton (Nielson et al., 1978; Sibbett and Nielson, 1980). At least ten rhyolite domes occur along the crest of the Mineral Mountains, representing igneous activity between 0.5 and 0.8 million years ago. A deep-seated magma body related to this young rhyolitic volcanism is a possible heat source for the present geothermal system (Smith and Shaw, 1975).

The western boundary of the geothermal system is defined by the northeast-trending Opal Mound fault (Fig. 1). Wells drilled east of this fault, except 52-21 and 24-36, produce commercial quantities of fluid, whereas the two wells drilled west of the fault (9-1 and 82-33) do not (Fig. 1) (Forrest, 1980).

Drill cuttings are available for study from four geothermal exploration wells, including the producing wells 72-16 and 14-2, and the

nonproducing wells 52-21 and 9-1. Mineralogic descriptions of the cuttings from these wells are taken from the work of Ballantyne and Parry (1978), Ballantyne, G. (1978), Nielson et al. (1978), Parry (1978), Rohrs and Parry (1978), Glenn and Hulen (1979) and Glenn et al. (1981). Petrographic studies of alteration mineralogy of these drill cuttings were limited by the small chip size which prohibited the accurate determination of paragenetic relationships.

Lithologies found in producing wells 72-16 and 14-2, and nonproducing wells 52-21 and 9-1, consist of arkosic alluvium overlying interfingering gneisses and granitic rocks. Alteration in these wells occurs mainly along faults and fractures that mark past and present fluid channels and appears to be largely independent of rock type.

Three alteration assemblages are recognized at depth in producing wells 72-16 and 14-2 (Fig. 2). These include an upper assemblage (I) characterized by the occurrence of montmorillonite, mixed-layer clays and epidote, a transition assemblage (II) present only in well 14-2, and a lower assemblage (III) that in contrast to the upper zones contains minor anhydrite and greater abundances of chlorite after plagioclase, pyrite and calcite. Despite these differences both zones contain chlorite after mafic minerals, limonite-hematite, quartz, sericite and traces of chalcopryite. Chalcedony is present in both zones of well 72-16 but absent from well 14-2. K-feldspar is common in rocks of the reservoir and is described as an alteration phase in both 72-16 and 14-2. It is very difficult, however, to distinguish hydrothermal K-

feldspar from perthitic, anti-perthitic and micrographic K-feldspar in gneisses and granitic rocks when examining only cuttings (Nielson et al., 1978), and therefore it is considered as a questionable alteration product. Magnetite-ilmenite, although present as primary phases in wells 14-2 and 72-16, are also described as alteration products in 72-16.

Although these zones occur at considerably different depths in wells 72-16 and 14-2, their measured temperatures are very similar (Fig. 2). The highest measured temperatures for assemblage I in wells 72-16 and 14-2 are 196° and 210°C, respectively. The lower-most assemblage (III) corresponds to temperatures ranging from 196°C to the bottom hole temperature of 243°C in well 72-16 and from 224°C to the bottom hole temperature of 268°C in well 14-2.

The alteration assemblage in nonproducing well 52-21 (assemblage IV) is markedly different from assemblages observed in the producing wells and includes sericite, chlorite (after mafic minerals), calcite and traces of hematite, pyrite, chalcopyrite, epidote and magnetite-ilmenite. The abundances of these alteration minerals are similar throughout the well with the exception of calcite and epidote which increase in concentration with depth, and hematite which only occurs above 762 m. Quartz and K-feldspar are present throughout the well, although not considered alteration products. The maximum measured temperature in well 52-21 is 204°C.

Alteration mineral assemblages in well 9-1 (although a nonproducing well) exhibit characteristics comparable to both nonproducing well 52-21 and the two producing wells 14-2 and 72-16. The upper mineral assemblage (I) in well 9-1 occurs above a major fault zone at 844 m and contains mixed-layer clays and epidote. This assemblage resembles the lower-temperature assemblages found in the producing wells (assemblages I and II). On the other hand, below 844 m in well 9-1, the lower alteration assemblage is characterized by the absence of chlorite after plagioclase and is most similar to the alteration assemblage observed in nonproducing well 52-21 (assemblage IV). An exception to this similarity, however, is the presence of trace amounts of anhydrite in assemblage IV of well 9-1. Measured temperatures in the upper zone of 9-1 (assemblage I) are less than 160°C, whereas in the lower portion of the well (assemblage IV) they range from 160°C to the bottom hole temperature of 224°C.

The apparent similarity between alteration minerals present in the upper portion of well 9-1 and the upper alteration assemblages found in the production wells suggests that at one time there was an influx of thermal fluid into the rocks above 844 m in well 9-1. Although well 9-1 is presently devoid of free-flowing thermal fluid, Glenn et al. (1981) have recognized a zone above the major fault at 844 m that is more fractured and altered than rock encountered in deeper portions of the well.



Several hydrothermal events have altered the rocks in the Roosevelt Hot Springs thermal area. Consequently, it is difficult to separate alteration assemblages produced by the present geothermal event from those of earlier events (i.e., Tertiary alteration associated with Cu-Pb-Zn mineralization (Bamford et al., 1980)). Nevertheless the mineralogic relationships described for wells 72-16, 14-2, 52-21 and 9-1 suggest that alteration assemblages characteristic of the producing wells are related to the present thermal event. This argument is supported by the similarity in the zoning sequences found within the two producing wells, 14-2 and 72-16, and the differences between alteration assemblages found in producing and nonproducing wells.

#### FLUID CHEMISTRY

Chemical analyses of fluids from wells and springs in the Roosevelt Hot Springs area (Table 1) indicate that four types of water are present: 1) deep circulating thermal fluid tapped by producing wells 14-2, 54-3, and 72-16; 2) hot water from nonproducing wells 9-1 and 52-21; 3) recent discharge from the Roosevelt seep; and 4) water from the now dry Roosevelt Hot Spring. Fluid from wells that do not produce has a composition clearly different from production well fluid. The temperature and concentration of Na, K, F, Cl and total dissolved solids are lower in fluid discharged from nonproductive wells, whereas Ca, Mg, Fe, SO<sub>4</sub> and HCO<sub>3</sub> are more concentrated. Spring water from the Roosevelt seep is similar to nonproducing well fluid, exhibiting greater concentrations of Ca, Mg and HCO<sub>3</sub> than producing well fluid. The total

dissolved solids content of fluid discharged from the seep, however, is in the same range as that measured for production wells. Fluid from the now dry Roosevelt Hot Spring is similar to producing well fluid but has higher Mg and lower Si concentrations.

Comparison of the compositions of the Roosevelt fluids, however, suggests they are derived from a common reservoir source and that variations in composition are due largely to ground water mixing. Local ground water is enriched in Ca, Mg, Fe,  $SO_4$  and  $HCO_3$  and depleted in Na, K, F and Cl relative to the thermal fluid (Mower and Cordova, 1974). This compares well with enrichments and depletions in nonproducing well fluids relative to production well fluid to indicate that nonproducing well fluid is mixed with local ground water.

The extent of ground water mixing with thermal fluid in the Roosevelt geothermal system tends to increase with distance from well 54-3 (Fig. 3). The minimum mixing percents for fluid from wells 14-2, 72-16, 9-1 and 52-21 average 7, 12, 17 and 21%, respectively, whereas the maximum amounts of ground water mixing with fluids discharged from the Roosevelt seep and hot spring average 25 and 11%, respectively. These relationships suggest that well 54-3 and other nearby producing wells have the most direct access to the reservoir. Contouring of heat flow data from the Roosevelt thermal area (Fig. 1) produces a similar pattern, with the highest heat flow corresponding to areas of least mixing.

An enthalpy-chloride diagram (Fig. 4) (Truesdell and Fournier, 1976; Fournier, 1979) is used to calculate the percentage of ground water mixing. This diagram employs the bottom-hole temperatures and fluid chloride concentrations listed in Table 1 and calculated percentages of steam loss listed in Table 2. Fluid from well 54-3 (sample 5) is taken as the well fluid having the lowest percentage of ground water because it exhibits the highest enthalpy and chloride concentrations in relation to the other wells. For the purposes of these calculations, 54-3(5) is designated as zero-percent mixed. This assumption allows minimum percentages of ground-water mixing to be calculated for other well fluids using Figure 4 (Fournier, 1979). Estimation of the percent mixing for surface seepages is complicated, however, by uncertainties in their cooling history. Assuming that surface seepages cooled entirely by steam loss to 100°C, the maximum percentages of ground water in fluids discharged from the Roosevelt seep and hot spring are calculated using Figure 4 (Fournier, 1979).

Fluids from producing wells have undergone single-stage liquid-vapor separation in the well bore. The fraction of isenthalpic steam separation,  $\chi_g$ , can be calculated from the relation

$$\chi_g = (h_{f,T_i} - h_{f,T_c}) / (h_{g,T_c} - h_{f,T_c}) \quad (1)$$

where  $h_g$  and  $h_f$  are the enthalpies of saturated steam and saturated liquid, respectively, at both the initial temperature,  $T_i$ , of the deep reservoir fluid and final temperature,  $T_c$ , of the fluid at the collection site. Steam-loss fractions calculated for fluid samples from

wells 14-2, 72-16 and 54-3 are listed in Table 2. Truesdell (Thermal Power Co., 1978) has calculated the presence of less than 1.5% steam in the Roosevelt reservoir, indicating that the fluid is very near liquid-vapor equilibrium. Therefore, it was not necessary to correct for excess or insufficient enthalpy in the reservoir fluid in these steam-loss calculations.

Interpretation of geothermometer calculations allows prediction of the deep reservoir fluid temperature to be  $288^{\circ}\text{C} \pm 10^{\circ}$ , compared to the maximum measured temperature of  $268^{\circ}\text{C}$  (Table 1). Calculated cation-geothermometer temperatures for producing well fluids, as listed in Table 1, range from  $284^{\circ}$  to  $297^{\circ}\text{C}$ . Cation-geothermometer temperatures, however, may be unreliable if uncorrected for the occurrence of calcite scaling (Fournier and Truesdell, 1973) in Roosevelt production wells. Sulfate water isotopic geothermometer temperatures for samples from two Roosevelt production wells predict reservoir temperatures of  $278^{\circ}$  and  $280^{\circ}\text{C}$  (Nehring and Mariner, 1979). In addition, a minimum temperature for the reservoir fluid of  $284^{\circ}$  is estimated from the point of intersection of lines A and B on the enthalpy-chloride diagram (Fig. 4) (see Fournier, 1979).

#### CALCULATION OF DEEP RESERVOIR FLUID CHEMISTRY

The composition of the deep reservoir fluid was estimated from calculations that account for the effects of ground-water mixing, steam loss, gas release and mineral precipitation in the well bore on the pH, gas partial pressures and element concentrations of the original

reservoir fluid. Fluid samples from well 14-2 are used for these calculations, because they are the only samples for which all the necessary data, including gas analyses (A. H. Truesdell, unpublished data, 1979), are available. In the absence of required data from other production wells, 14-2 well fluid was considered adequate to represent the reservoir fluid composition because, as concluded in the previous section, production well fluids all have similar compositions and are probably derived from a common source. These 14-2 samples, in particular (3) and (4), are reliable in that they were chemically preserved and filtered at the sample site. Also, chemical and isotopic data indicate that the steam and liquid samples are near complete separations (Thermal Power Co., 1978).

The composition of 14-2 well fluid used in the reservoir fluid calculations (Table 3) represents the average of analyses 14-2(1) through 14-2(4). The average analysis corrected for concentration by 18% steam loss is also given in Table 3. Because the  $\text{HCO}_3$  content of these samples was not measured, the  $\text{HCO}_3$  concentration present in the least mixed fluid 54-3 (5), corrected for 12% steam loss, is used.

An average fluid analysis corrected for both concentration by 18% steam loss and dilution through 7% mixing was also determined. Both of these fluids were considered because it is not known whether mixing occurred in the well bore or reservoir. Mineral-fluid equilibrium calculations on both these calculated fluids produced very similar results. Therefore, the fluid corrected only for steam loss is

discussed in this paper.

Because well 14-2 is cased to 551 m (Glenn and Hulen, 1979), sampled waters are assumed to represent a composite of fluids derived from 551 m to at least 1830 m, the total depth of the well. Well log temperatures in this depth interval vary from 210° to 268°C (Glenn and Hulen, 1979). The only significant hot-water entry occurs at 869-881 m (Bamford et al., 1980), where the recorded well log temperature is 250°C.

#### Methodology for Equilibrium Calculations

The distribution of element concentrations among aqueous species is calculated using a modification of the computer program PATH (Helgeson et al., 1970; Knight, 1976). The sources of thermochemical data for aqueous species considered in these calculations are Helgeson (1969), supplemented by data from Kharaka and Barnes (1973), Bladh (1978) and Rimstidt (1979). Thermochemical equilibrium constants for minerals and gases are calculated using data reported by Helgeson et al. (1978).

In these calculations the standard state for H<sub>2</sub>O and intercrystalline standard state for solids are consistent with unit activity of the pure component at any pressure and temperature. The intracrystalline standard state for minerals calls for all activity coefficients of atoms on the lattice sites of solid solutions to approach unity as the mole fractions of the atoms on the sites approach those in the thermodynamic components of the solution at any pressure and temperature. The standard state for aqueous species, other than

H<sub>2</sub>O, is one of unit activity in a hypothetical one molar solution referenced to infinite dilution at any pressure and temperature. For gases the standard state is one of unit fugacity of the hypothetical ideal gas at one bar and any temperature.

Component activities accounting for nonstoichiometry of sericite and chlorite in rock samples from well 14-2 are calculated from electron microprobe analyses reported by Ballantyne, J. (1978, 1980) and are shown in Figure 5. The specific expressions for calculating the activities of the components, which are listed in Table 4, are derived from the general equations relating site occupancy in a mineral to the activity of the thermodynamic component as presented by Helgeson *et al.* (1978, Equations (46) through (52)), and from preferential site occupancies as defined by Helgeson and Aagaard (1981). The thermodynamic components of the mineral sericite correspond to the chemical formula units of Al<sub>2</sub>Si<sub>4</sub>O<sub>10</sub>(OH)<sub>2</sub>, KAl<sub>2</sub>(AlSi<sub>3</sub>O<sub>10</sub>)(OH)<sub>2</sub> and NaAl<sub>2</sub>(AlSi<sub>3</sub>O<sub>10</sub>)(OH)<sub>2</sub>. For chlorite the activity of the component Mg<sub>5</sub>Al(AlSi<sub>2</sub>O<sub>10</sub>)(OH)<sub>8</sub> is calculated.

#### Gas pressures

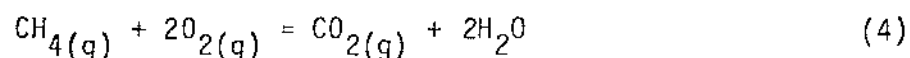
Calculated partial pressures of O<sub>2</sub>, CO<sub>2</sub>, H<sub>2</sub>S and CH<sub>4</sub> dissolved in the reservoir fluid are shown in Figure 6. The partial pressures of CO<sub>2</sub>, H<sub>2</sub>S and CH<sub>4</sub> are calculated using the Henry's law relation

$$f = K_T X \quad (2)$$

where *f* is the fugacity of the gas (at the temperatures and pressures

considered in this study the fugacity is essentially equivalent to the partial pressure of a gas in atmospheres),  $K_T$  is the Henry's law constant at temperature  $T$  (Table 5), and  $X$  is the mole fraction of the gas in the reservoir fluid. Mole fractions of these gases in the reservoir fluid are calculated from concentrations measured in the steam sample. The gas content of the steam fraction collected at the same time as liquid samples (1) through (4) from well 14-2 was provided by A. H. Truesdell (unpublished data, 1979). It is assumed for these calculations that  $\text{CO}_2$ ,  $\text{H}_2\text{S}$  and  $\text{CH}_4$  separate completely into the steam fraction. This is supported by the work of Drummond (1981, Fig. 4.5) in which he calculates that with 18% steam loss as the result of isenthalpic boiling of a  $250^\circ\text{C}$  (3m NaCl) fluid, less than 1% of these gases will remain in the liquid phase. This is further supported by a study of gas concentrations in geothermal discharges from the Wairakei system (Ellis, 1962) which has temperatures, pressures and fluid composition similar to those of Roosevelt Hot Springs.

The oxygen partial pressure ( $P_{\text{O}_2}$ ) of the reservoir fluid is approximated using both hematite-magnetite and methane-carbon dioxide equilibria, given by equations (3) and (4), respectively,



and from the relationship between temperature and  $P_{\text{O}_2}$  determined by D'Amore and Panichi (1980) (equation 5).



$$\log P_{O_2} = 8.20 - (23643 / T(^{\circ}K)) \quad (5)$$

At 288°C the  $P_{O_2}$  ranging from  $10^{-32}$  to  $10^{-35}$  atm,  $P_{CO_2}$  of 11 atm,  $P_{H_2S}$  of 0.020 atm, and  $P_{CH_4}$  of 0.001 atm in the Roosevelt reservoir fluid are similar in magnitude to calculated gas pressures in other high-temperature geothermal reservoirs such as Wairakei and Broadlands (Ellis, 1979; D'Amore and Panichi, 1980).

#### Ion concentrations corrected for gas losses and scaling

The concentrations of calcium, carbon and sulfur will decrease in the fluid by 5, 1753 and 119 mg/l, respectively, as a result of  $CO_2$ ,  $CH_4$  and  $H_2S$  gas release and calcite precipitation accompanying steam separation. The amounts of carbon and sulfur lost to the steam fraction are calculated from the concentrations of  $CO_2$ ,  $CH_4$  and  $H_2S$  released from the flashed fluid. The amounts of calcium and carbon removed from the reservoir through calcite precipitation are calculated from the change in calcite solubility as a result of boiling. The composition of the reservoir fluid corrected for these losses is given in Table 3.

As the reservoir fluid boils and thereby cools from the reported bottom-hole temperature of 265°C to the collection temperature of 192°C, the solubility of calcite decreases by  $1.31 \times 10^{-4}$  moles/l. This solubility decrease is largely a result of the  $CO_2$  pressure decrease in response to the removal of  $CO_2$  from the fluid by the gas phase. The solubility change of calcite can be determined using equation (6) (adapted from Segnit et al., 1962) which gives moles of  $Ca^{++}$  in a kilogram of fluid,  $m_{Ca^{++}}$ , in equilibrium with calcite.

$$m_{\text{Ca}^{++}}^3 = 13.9 K_1 K_c X_{\text{CO}_2} / K_2 \quad (6)$$

$$K_1 = a_{\text{H}^+} a_{\text{HCO}_3^-} / a_{\text{H}_2\text{CO}_3} \quad (7)$$

$$K_2 = a_{\text{H}^+} a_{\text{CO}_3^{--}} / a_{\text{HCO}_3^-} \quad (8)$$

$$K_c = a_{\text{Ca}^{++}} a_{\text{CO}_3^{--}} / a_{\text{CaCO}_3(\text{s})} \quad (9)$$

$K_j$  = equilibrium constant for the  $j^{\text{th}}$  reaction

$X_{\text{CO}_2}$  = mole fraction of  $\text{CO}_2$  gas in the reservoir fluid

$a_i$  = activity of the  $i^{\text{th}}$  species in solution

The variation in the mole fraction of  $\text{CO}_2$  in the reservoir fluid as a result of boiling is determined using the fraction of  $\text{CO}_2$  removed with each percentage of steam separated from the flashed  $260^\circ\text{C}$  reservoir fluid of the Wairakei geothermal system as reported by Ellis (1962, Fig. 3).

For the estimation of calcium and carbon loss due to calcite scaling, it is assumed the reservoir fluid is in equilibrium with calcite and that the fluid remains in equilibrium with calcite as it boils. The occurrence of calcite at depth in well 14-2 supports this assumption of equilibrium. Filtering of the sample upon collection corrects for removal of calcite precipitate that has not adhered to the piping.

#### Hydrogen ion concentration

The reservoir fluid pH is determined by the method of hydrogen ion

## DISCUSSION OF FLUID-MINERAL EQUILIBRIA

The equilibrium relationship between the alteration mineralogy and the deep reservoir fluid of the Roosevelt Hot Springs thermal system is quantitatively evaluated at temperatures ranging from 150° to 300°C. Species distribution calculations are used to calculate mineral equilibria in the deep reservoir fluid at 150°, 200°, 250° and 300°C. The results of these calculations are displayed on a plot of chemical affinity of each mineral in solution against temperature in Figure 7.

The chemical affinity,  $A_j$ , (Helgeson, 1979) indicates the equilibrium condition of the mineral in the fluid and is calculated using equation 10,

$$A_j = RT \ln(K_j/Q_j) \quad (10)$$

where  $K_j$  and  $Q_j$  represent the equilibrium constant and activity product for the  $j^{\text{th}}$  reaction,  $T$  is the temperature in °K and  $R$  is the gas constant. A positive value indicates the mineral is undersaturated with respect to the fluid. The chemical affinity is zero for mineral-fluid equilibrium and negative for supersaturation. Phases that satisfy equilibrium or supersaturated conditions are shown in the lower portion of Figure 7, with undersaturated conditions represented on the upper portion of the diagram.

– Minerals included on the chemical affinity versus temperature diagram are those described as alteration minerals in the Roosevelt system. Thermochemical data for montmorillonite and mixed-layer clays

are not supplied in the data compilation by Helgeson et al. (1978), nor are compositional data available to calculate component activities. Data for (Ca-) montmorillonite and illite equilibrium taken from Helgeson (1969) are, therefore, used to calculate the equilibrium trends of these minerals. Although these data are not entirely consistent with the Helgeson et al. (1978) data base, they will at least provide a reasonable approximation of the actual equilibrium conditions.

Microprobe analyses were available for chlorite and sericite from well 14-2 (Ballantyne, J., 1978, 1980), allowing calculation of the component activities to account for solid solution in these minerals. Using these calculated activities, the chemical affinity of the component in solution was calculated for the  $KAl_2(AlSi_3O_{10})(OH)_2$  component of sericite and the  $Mg_5Al(AlSi_3O_{10})(OH)_8$  component of chlorite. These component affinities are represented on Figure 7 by the dashed lines. For comparison, the chemical affinities of the pure end members muscovite and clinocllore are also shown on Figure 7.

The activities of the  $KAl_2(AlSi_3O_{10})(OH)_2$  component of sericite used to calculate the range in chemical affinities of this component are shown in Figure 7. The activity of the  $KAl_2(AlSi_3O_{10})(OH)_2$  component of sericite tends to decrease with increased depth (Fig. 5). A similar trend is noted for the activities of the other two components of sericite (Fig. 5),  $NaAl_2(AlSi_3O_{10})(OH)_2$  and  $Al_2Si_4O_{10}(OH)_2$ . The distribution of available compositional data with depth allows for a grouping of samples into those above 896 m and those below 1341 m in

mass balance (Truesdell and Singers, 1974; Bischoff and Dickson, 1975) (Fig. 6). This method of estimating pH is based on the assumption that the mass balance of hydrogen in a fluid is independent of temperature and that the hydrogen mass balance of the fluid corrected for gas separation equals that of the deep reservoir fluid. The method of hydrogen mass balance is used in this study because it is independent of mineral equilibrium relationships which the final calculated reservoir fluid will ultimately be used to predict.

The hydrogen mass balance for the fluid corrected for  $\text{CO}_2$  and  $\text{H}_2\text{S}$  loss is 0.340 moles/l. This corresponds to a pH of 6.0 at  $288^\circ\text{C}$  (Fig. 6) and is similar to pH values calculated for other high-temperature geothermal fluids. For example, New Zealand geothermal fluids having temperatures of  $220^\circ\text{C}$  exhibit a range in calculated reservoir pH from 5.9 to 7.1 (Ellis, 1979), and Icelandic geothermal fluids with temperatures of  $195^\circ$  to  $220^\circ\text{C}$  range in pH from 5.0 to 8.3 (Arnorsson et al., 1978; Ellis, 1979).

#### Effect of changing temperature

The effects of temperature change on the pH, gas partial pressures and distribution of aqueous species were determined at  $150^\circ$ ,  $200^\circ$ ,  $250^\circ$  and  $300^\circ\text{C}$  for fixed concentrations of elements in solution (Fig. 6). These calculations suggest that decreasing temperature produces a decrease in the fluid pH and  $P_{\text{O}_2}$ , an increase in  $P_{\text{CO}_2}$  and  $P_{\text{CH}_4}$ , and no consistent variation in  $P_{\text{H}_2\text{S}}$ .

well 14-2. The average activity of  $\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{OH})_2$  is 0.51 for the upper group and 0.36 for the lower. Measured well temperatures for these two groups is 210° to 250°C for the upper group and 255°C for the lower group (Fig. 2). The calculated equilibration temperature ( $A_j = 0$ ) of the  $\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{OH})_2$  component with the reservoir fluid for the upper group is 246°C and for the lower group is 254°C (Fig. 7). This suggests that solid solution in sericite is temperature dependent and that calculation of component activities to correct for its effects on fluid-mineral equilibrium calculations is appropriate.

The activity of the  $\text{Mg}_5\text{Al}(\text{AlSi}_3\text{O}_{10})(\text{OH})_8$  component of chlorite does not appear to vary consistently within the depth range for which microprobe analyses are available (Fig. 5). Therefore, the average activity of 0.013 for all depths is used to calculate the chemical affinity of this component in solution. Chlorites in the reservoir rock contain nearly equal atomic proportions of Fe and Mg (Ballantyne, J., 1978). Thermochemical data for the Fe component of chlorite, however, are not available and therefore only the Mg component is discussed.

Comparison of mineral equilibria in the calculated reservoir fluid (Fig. 7) with alteration mineral assemblages described in producing wells (Fig. 2) indicates that the majority of these minerals could have been produced through interaction of the reservoir rocks with the present thermal fluid. Phases such as hematite, magnetite, pyrite and quartz are saturated with respect to the fluid at nearly all temperatures. These minerals are present in the production wells at all

depths, except hematite which is absent from the deeper high-temperature alteration assemblage III in well 14-2.

A select group of phases including chalcedony, the  $KAl_2(AlSi_3O_{10})(OH)_2$  component of sericite, K-feldspar, (Ca-) montmorillonite and illite exhibit a crossover from undersaturated to saturated conditions in the temperature range from 229° to 260°C. Typically those phases that exhibit a crossover between 240 and 260°C occur in both the low (I and II) and high (III) temperature alteration zones of the producing wells. These phases include sericite, K-feldspar and chalcedony. A second group of minerals that equilibrate below 240°C is present only in the lower-temperature alteration zones (I and II) of the production wells and includes the clay minerals montmorillonite and illite.

Finally, there is a group of phases present as alteration minerals in production wells 72-16 and 14-2 that is undersaturated with respect to the reservoir fluid at all temperatures, suggesting that these minerals could be the result of a past thermal event. This group includes chlorite, calcite, epidote and anhydrite. Anhydrite and epidote are present in only trace amounts in producing wells, whereas chlorite and calcite are more abundant. All four of these minerals, however, are also present in the nonproducing wells, therefore their presence in the production wells could be the result of a past thermal event.

The abundance of calcite in the upper alteration zone (I) of wells 14-2 and 9-1, in contrast to the near absence of calcite in the upper 700 m of nonproducing well 52-21, however, suggests that calcite in assemblage I is a product of the present thermal event. Because calcite equilibrium is strongly dependent on those chemical characteristics of the reservoir fluid most difficult to quantify, pH,  $P_{CO_2}$  and total carbon, it is possible that the error in calculating these fluid components could indeed allow for calcite equilibrium with the fluid. On the other hand, the presence of calcite in the upper 400 m of the reservoir rock can be explained by boiling of the fluid at these shallower depths. According to data from Mahon et al. (1980, Fig. 1), the present Roosevelt Hot Springs reservoir fluid, with 0.64 wt %  $CO_2$ , will be very close to, if not at, its hydrostatic boiling point in the upper portions of the reservoir (less than 400 m). Boiling of this fluid in the formation rocks can precipitate calcite, a condition analogous to steam loss in well bores in the Roosevelt Hot Springs geothermal system that produces a calcite scale.

Chlorite after plagioclase is present in both wells 72-16 and 14-2 but is lacking from nonproducing well 52-21 and found only rarely and in trace quantities in nonproducing well 9-1. Chlorite after mafics, however, is present in all wells. This suggests that chlorite after plagioclase is a product of the present thermal event, despite the apparent nonequilibrium of chlorite with the reservoir fluid. The lack of thermochemical data for the Fe component of chlorite, however, does not allow a complete evaluation of chlorite equilibrium relationships



with the known reservoir fluid. Bird and Norton (1981) in evaluating fluid-mineral equilibria in the Salton Sea geothermal system found a similar situation of nonequilibrium of the thermal fluid with chlorite which appeared related to the present thermal event.

These fluid-mineral equilibrium calculations do not take into account the effects of mineral precipitation on the composition of the fluid as it travels along its flow path in the cooling process. The possibility of mixing with nonthermal ground waters, which is undoubtedly occurring at the margins of the system, or the mineralogic consequences of boiling are also not considered in detail. It is realized, however, that in the Roosevelt thermal system the effects of these processes could be significant.

This study suggests, however, that the observed sequence of a lower to upper mineral assemblage in the production wells can be produced by the present reservoir fluid as it cools from approximately 300°C during its migration to the surface. The mineral equilibrium calculations suggest fluid temperatures of 240° to 260°C in rocks containing the lower mineral assemblage, and temperatures ranging from approximately 229° to 240°C in rocks of the upper mineral assemblage. These predicted temperatures agree within 40°C with the maximum measured temperatures of 210°C in the upper production well alteration assemblage I and 268°C in the lower production well assemblage III (Fig. 2). The highest temperature of 260°C predicted for fluid-mineral equilibrium in mineral assemblage III is remarkably close to geothermometer temperatures for

the fluid. For example, geothermometer temperatures estimated from the elemental composition of the surface sample, sulfate water isotopes and enthalpy-chloride relationships give a temperature of  $288^{\circ}\text{C} \pm 10^{\circ}$ . Also, the Na-K-Ca geothermometer temperature determined from the calculated composition of the deep reservoir fluid is  $277^{\circ}\text{C}$  (Table 3).

#### CONCLUSION

In the Roosevelt Hot Springs geothermal system, the interaction of the country rock with the present high-temperature thermal fluid will result in a complex series of dissolution and deposition reactions as the fluid and rock attempt to equilibrate. The extent to which the thermal fluid is in equilibrium with the country rock is estimated by comparing actual alteration assemblages with predicted fluid alteration products.

The composition of the high-temperature fluid at depth is estimated from well samples. The effects of nonthermal ground-water mixing, mineral precipitation in the well bore, steam loss and gas release on these surface samples are considered. The predicted composition of the deep fluid is similar in composition to fluids found in other high-temperature geothermal systems, such as Wairakei and Broadlands (Ellis, 1979).

Comparing predicted alteration mineral assemblages for the calculated reservoir fluid, in the temperature range from  $150^{\circ}$  to  $300^{\circ}\text{C}$ , with alteration assemblages described in drill cuttings of geothermal

production wells indicates that several alteration minerals are equilibrated with the reservoir fluid. These include hematite, magnetite, pyrite and quartz which are saturated with respect to the fluid at all temperatures, and chalcedony, sericite, K-feldspar, (Ca-) montmorillonite and illite which are saturated in the fluid at temperatures less than 260° to 229°C. The calculated equilibration temperatures of the last group of minerals predicts to within 40°C the transition zone observed between shallow (lower temperature) and deep (higher temperature) alteration zones. Furthermore, the variation in temperatures of equilibration of the  $KAl_2(AlSi_3O_{10})(OH)_2$  component of sericite with the reservoir fluid with depth in well 14-2 is in agreement with observed temperatures in the well. This indicates that sericite solid solution is temperature dependent and that it is necessary to correct for its effects in equilibrium calculations.

Phases that do not exhibit equilibrium with respect to the reservoir fluid, such as epidote, anhydrite, chlorite and calcite, could be remnants of an earlier event. Disequilibrium of the reservoir fluid with chlorite, however, may be the result of inadequate thermochemical data, as the presence of chlorite after plagioclase as a characteristic alteration product in production wells suggests it is a product of the present thermal event. Calcite, on the other hand, could be a product of another process inherent to geothermal activity, such as boiling.

It is suggested, therefore, that the geothermal reservoir fluid at Roosevelt Hot Springs has probably not changed character significantly

in the recent past. This is supported by the similarity between the observed alteration mineralogy and fluid-mineral equilibrium calculated for the reservoir fluid and the near agreement between predicted equilibration temperatures and observed temperatures.

## ACKNOWLEDGEMENTS

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#### FIGURE CAPTIONS

Figure 1. Generalized geologic map of the Roosevelt Hot Springs thermal area, taken from Nielson and Moore (1979). Closed circles represent geothermal production wells and open circles represent nonproducing wells, as defined by Forrest (1980). Triangles indicate areas of surface seepage. The heat flow contours are taken from Wilson and Chapman (1980).

Figure 2. Alteration mineralogy and temperature log data for wells 72-16, 14-2, 52-21 and 9-1, Roosevelt Hot Springs thermal area. Mineralogic and temperature-log data taken from Ballantyne and Parry (1978), Ballantyne, G. (1978), Nielson et al. (1978), Parry (1978), Rohrs and Parry (1978), Glenn and Hulen (1979) and Glenn et al. (1981). When more than one temperature log was available, the log that recorded the highest overall temperatures was used. A solid line indicates minor abundances; a dotted line indicates trace abundances.

Figure 3. Average percentage mixing of nonthermal groundwater with the thermal reservoir fluid, Roosevelt Hot Springs thermal area. The percentage of mixing is included in parentheses after the well number. ND indicates not done. Refer to Figure 1 for the location of this area.

Figure 4. Enthalpy-chloride diagram, Roosevelt Hot Springs thermal area. Data and sample numbers on this figure are taken from Tables 1 and 2. The local nonthermal ground water composition is taken from Mower and Cordova (1974). Triangles represent producing well fluid compositions corrected from steam loss, squares represent nonproducing well and surface seep samples assuming conductive cooling. Curves "A" and "B" are the upper bounding boiling and mixing curves, respectively. "RHS" denotes Roosevelt Hot Spring. This diagram was constructed employing the methods described by Fournier (1979).

Figure 5. Variation with depth of the activities of the  $KAl_2(AlSi_3O_{10})(OH)_2$ ,  $NaAl_2(AlSi_3O_{10})(OH)_2$  and  $Al_2Si_4O_{10}(OH)_2$  components in sericite, and the  $Mg_5Al(AlSi_3O_{10})(OH)_8$  component in chlorite from well 14-2. These activities are calculated using electron microprobe analyses of these minerals taken from Ballantyne, J. (1978, 1980). Hexagons indicate the average value for that depth and the bars represent the range in values. Refer to the text for the method of calculation.

Figure 6. The variation with temperature of pH and partial pressures of  $O_2$ ,  $CO_2$ ,  $H_2S$  and  $CH_4$  in the reservoir fluid. See text for methods of calculation.

Figure 7. Chemical affinities of minerals with respect to the Roosevelt reservoir fluid from  $150^\circ$  to  $300^\circ C$ . See text for discussion.

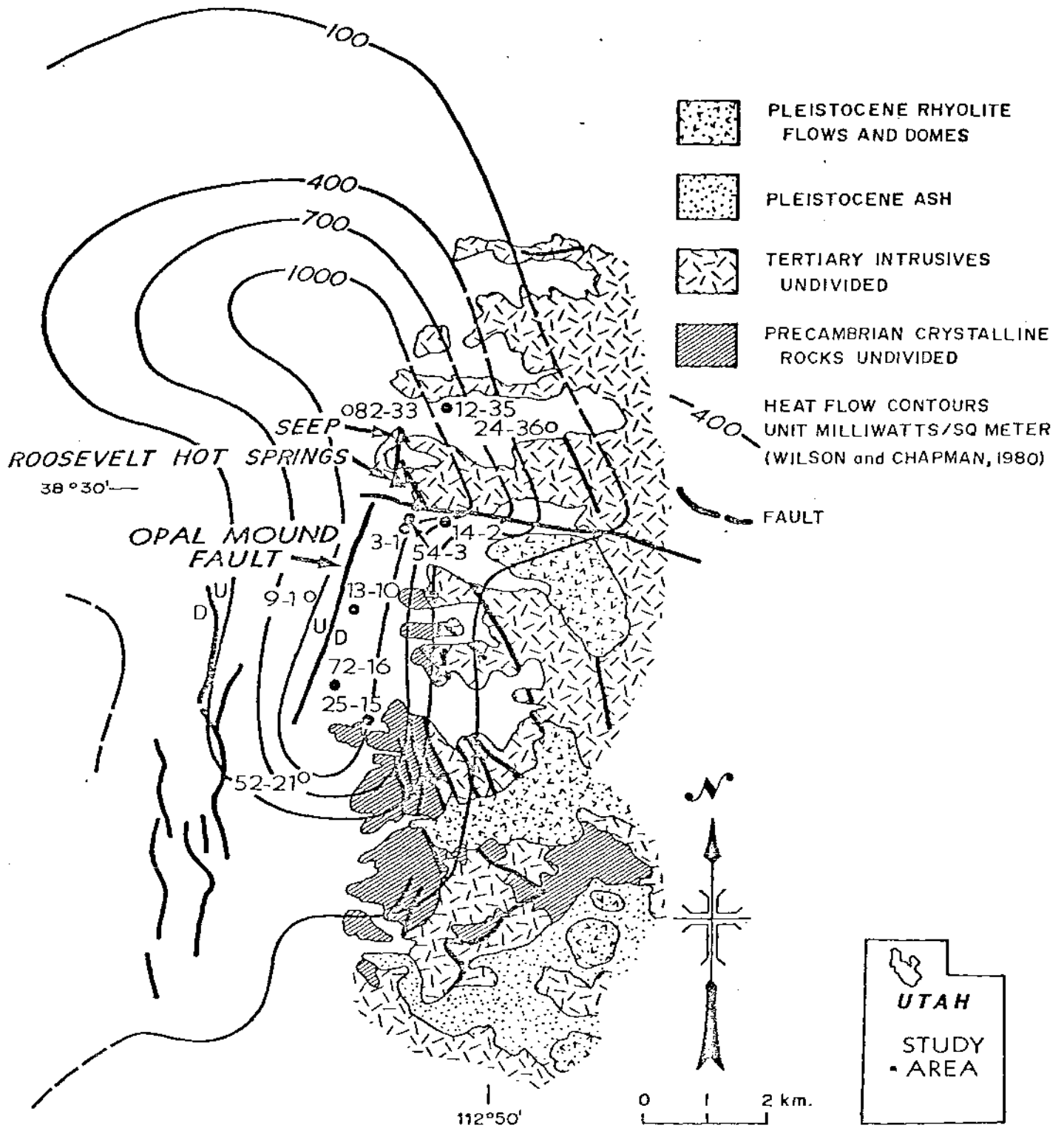


Figure 1



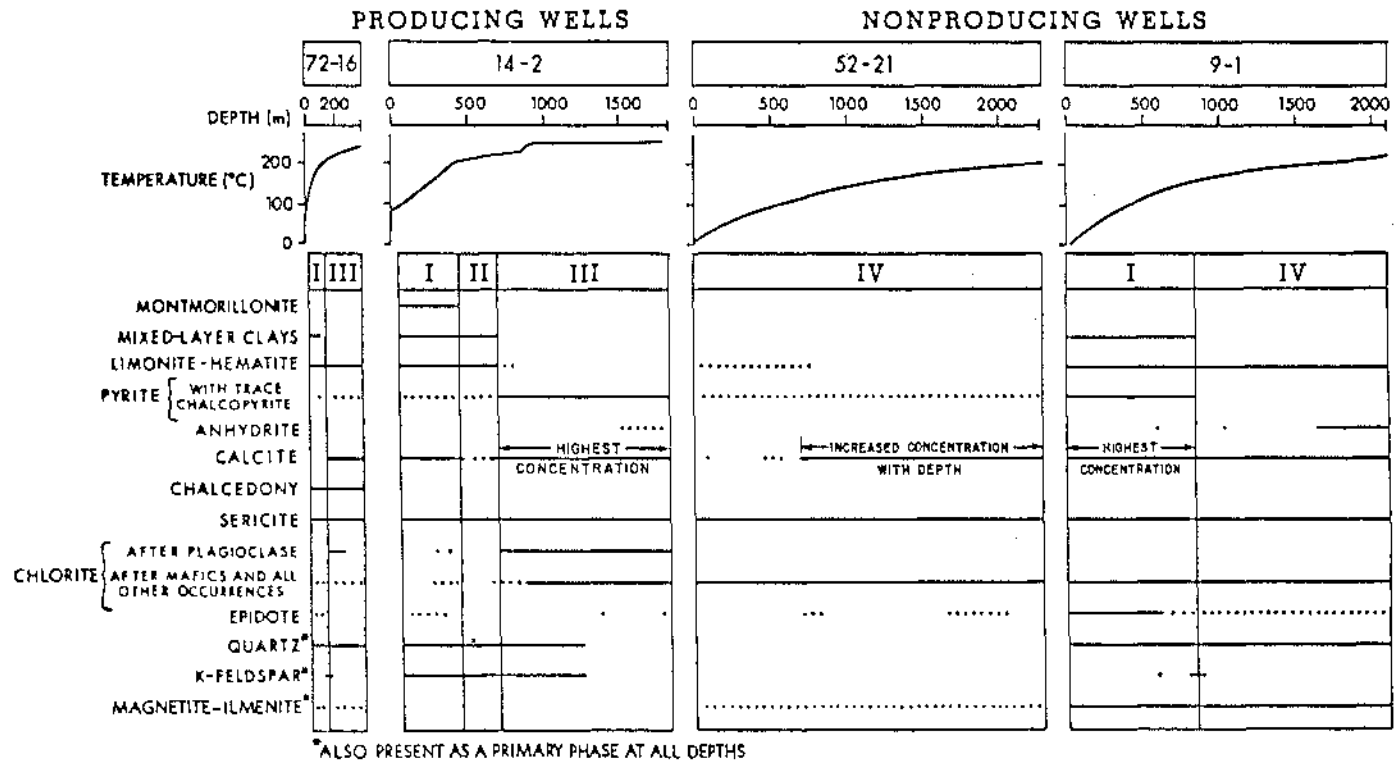


Figure 2

T26S  
T27S

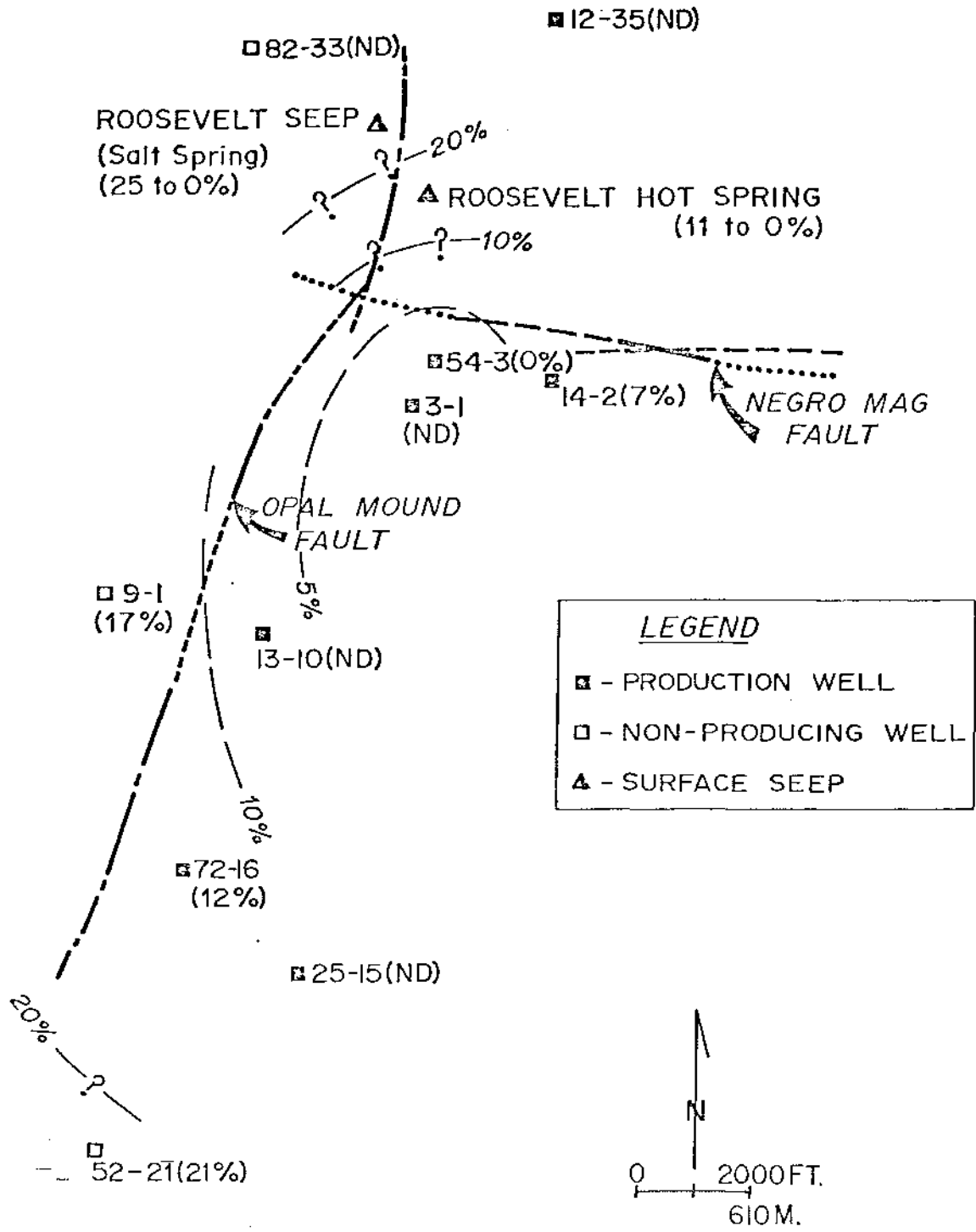


Figure 3

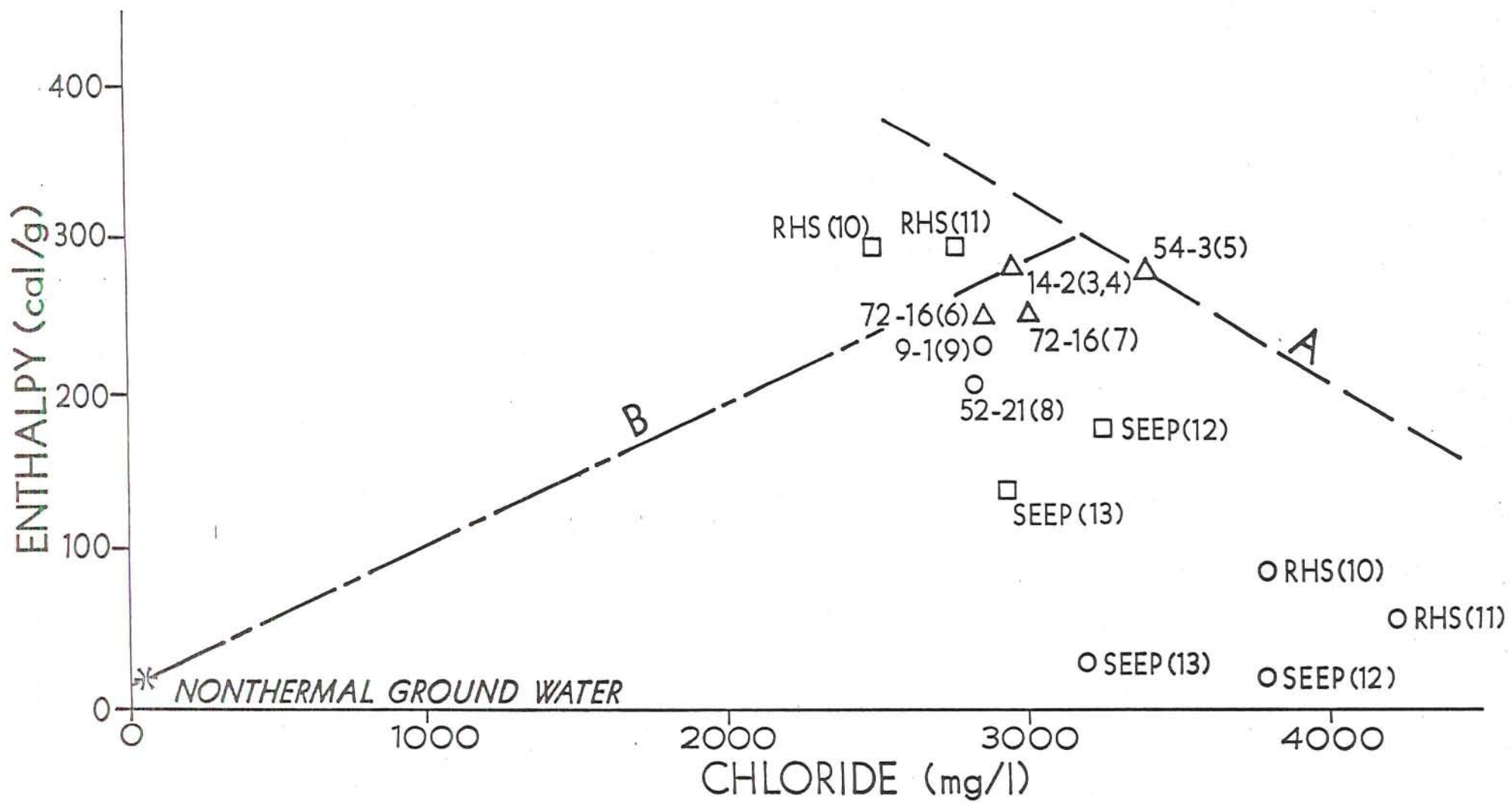


Figure 4

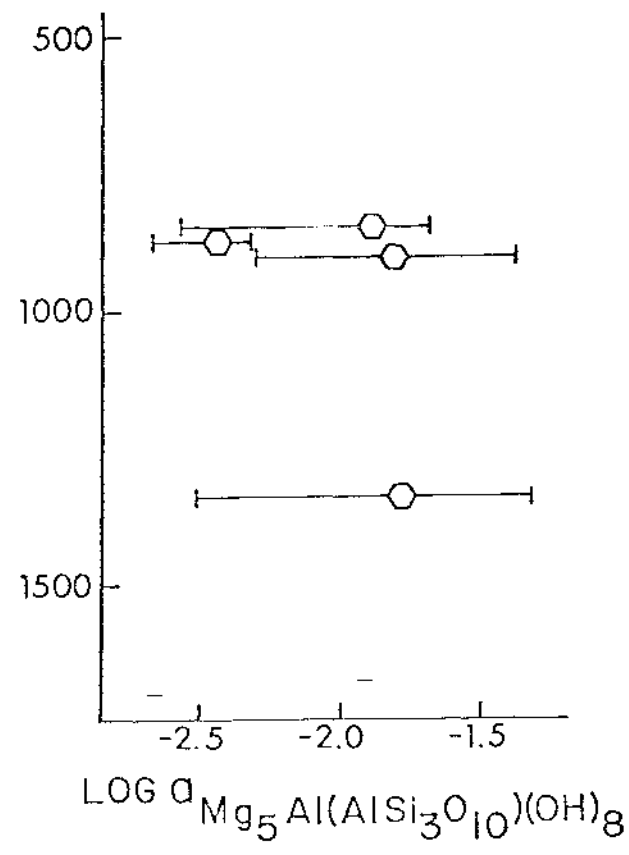
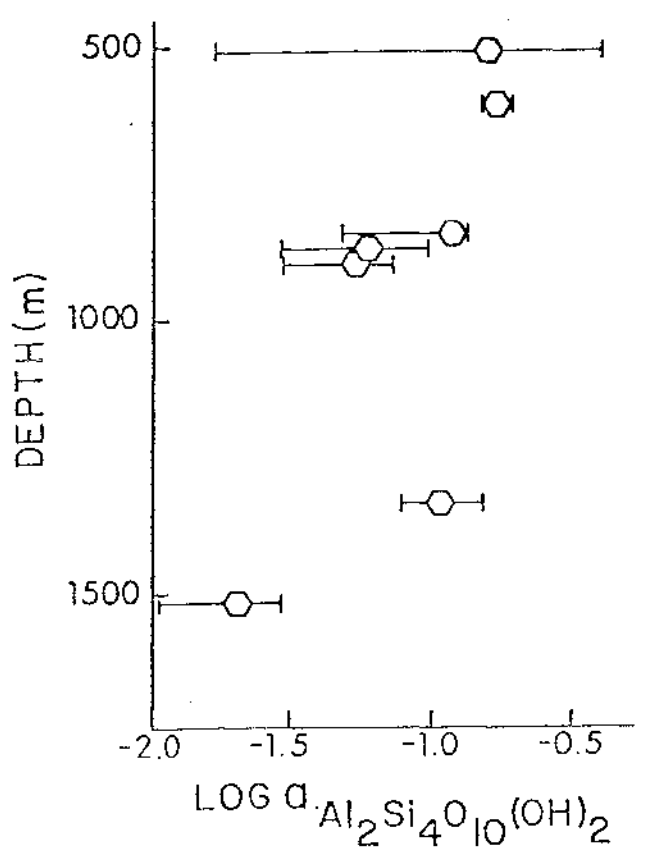
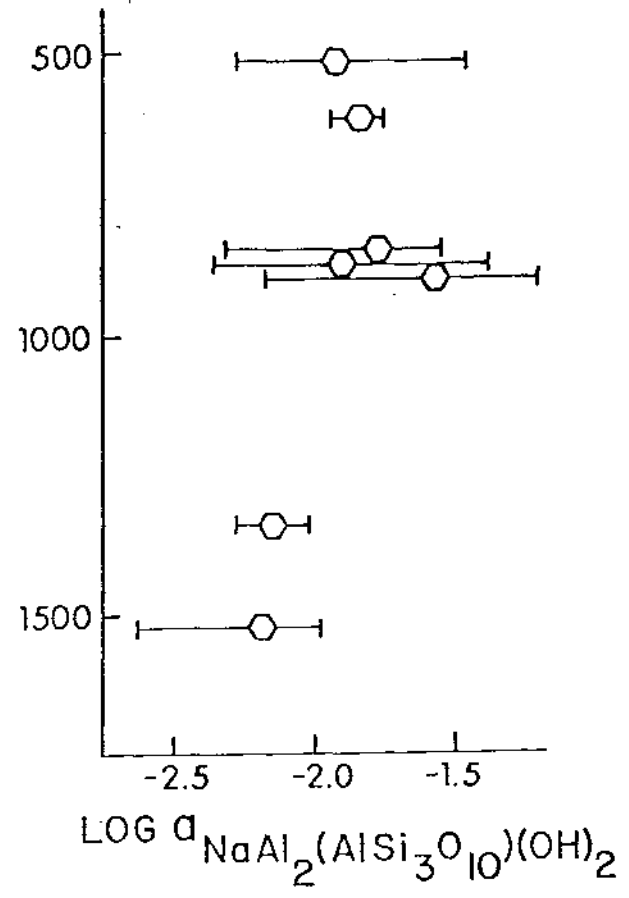
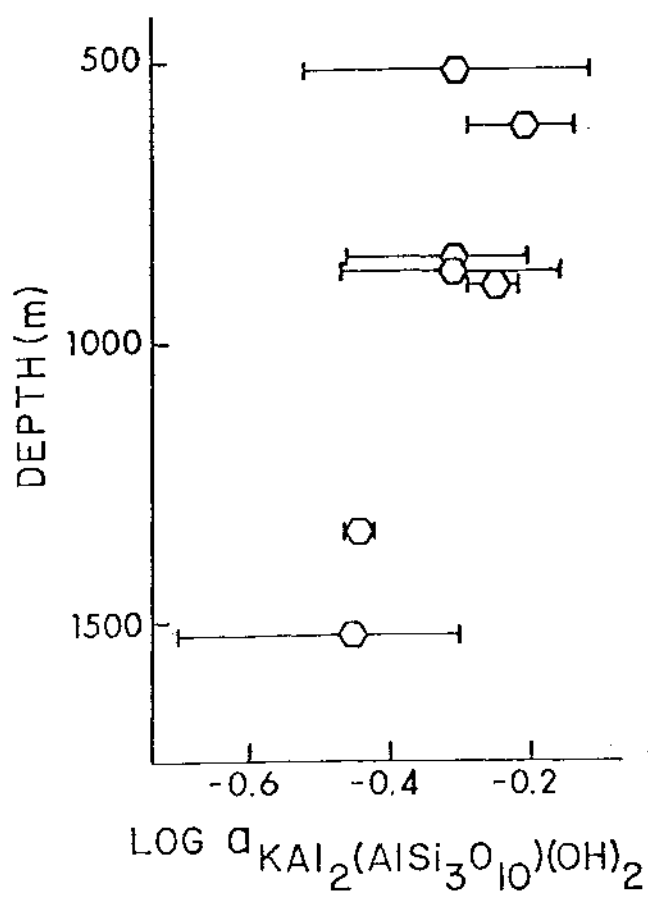


Figure 5

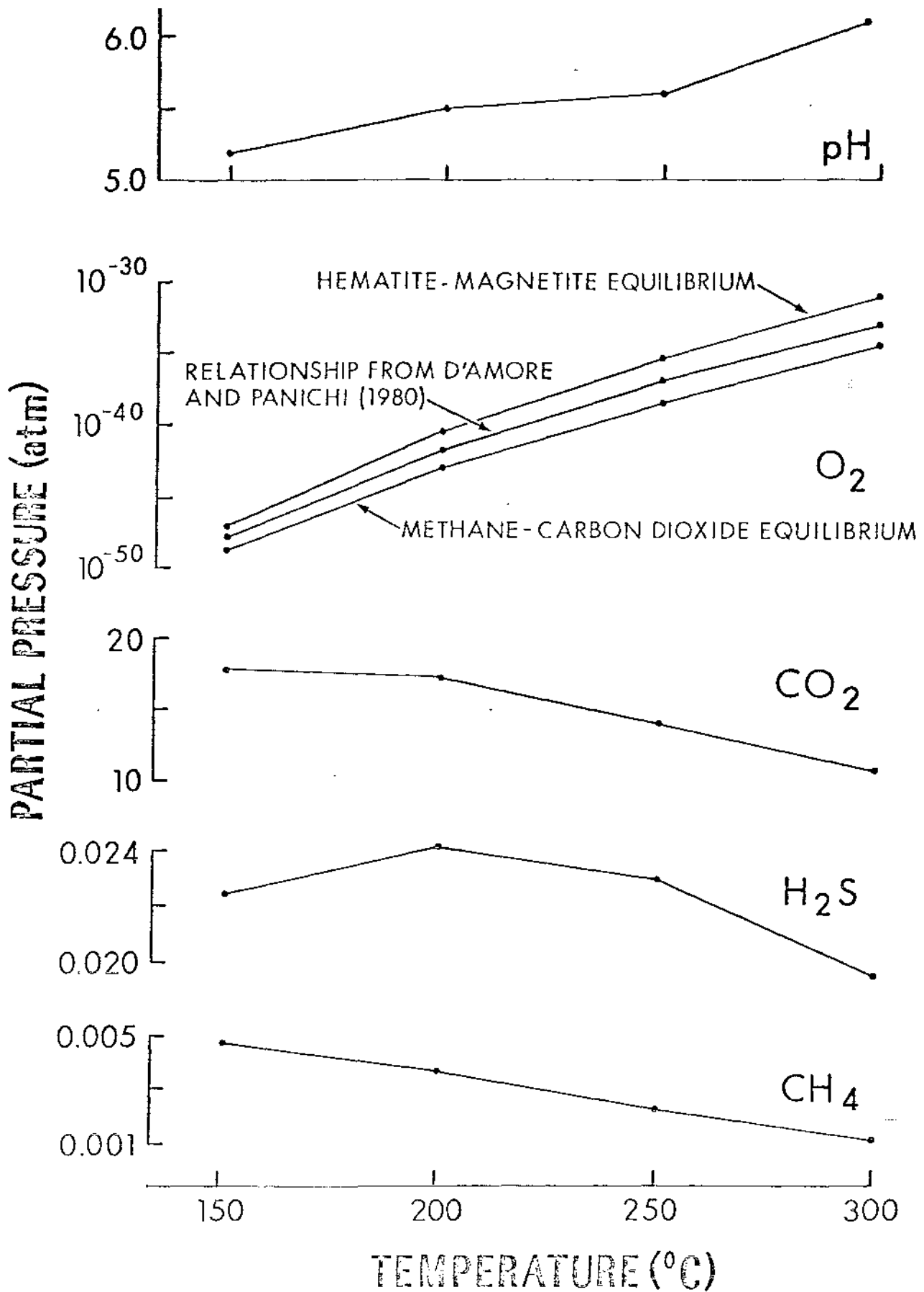


Figure 6

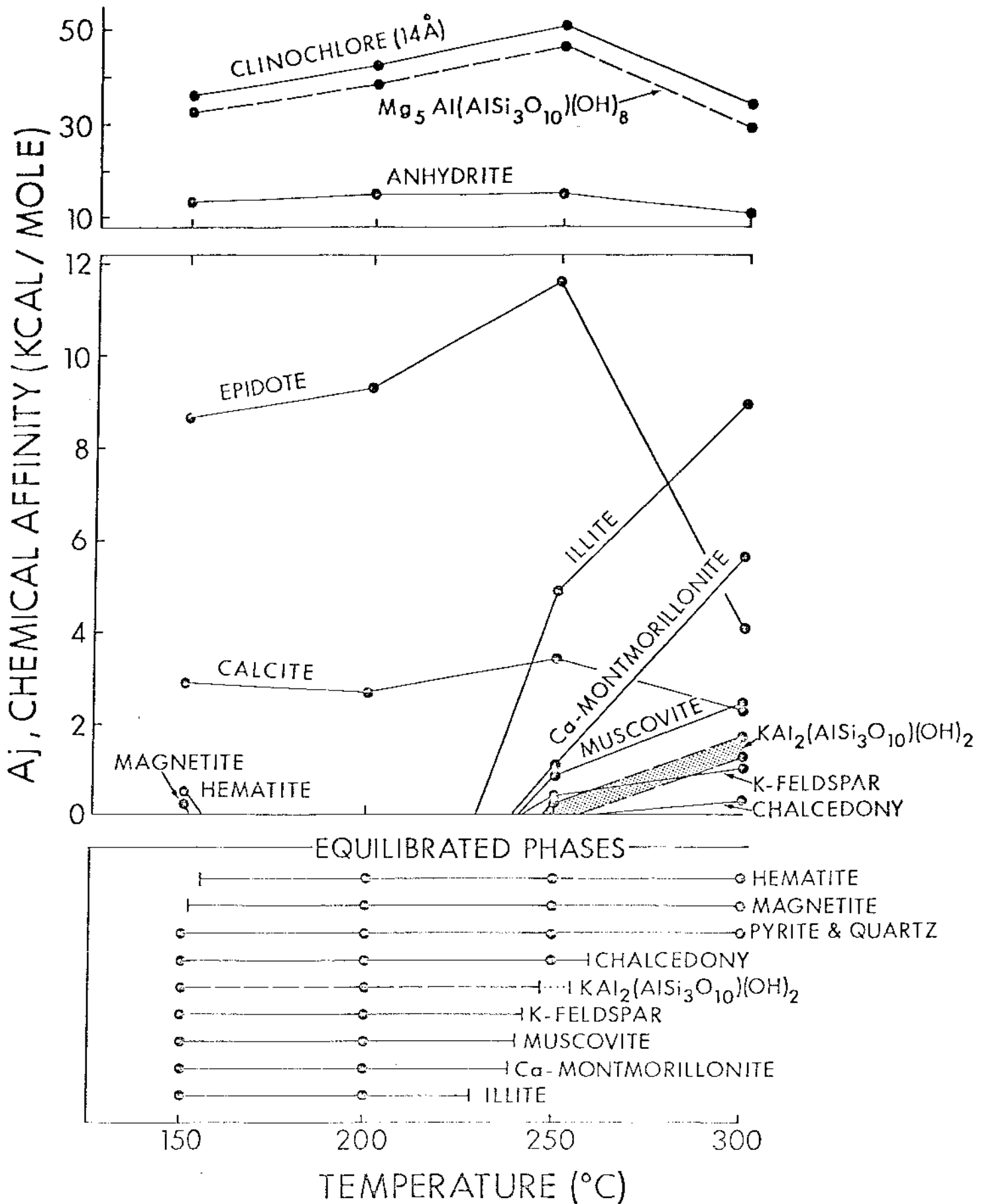


Figure 7

Table 1. Chemistry of Thermal Water<sup>1</sup>

Sample No	1 <sup>9,10</sup>	2 <sup>9,11</sup>	3	4	5	6	7 <sup>9,12</sup>	8	9	10	11	12	13
Well: Utah State	14-2	14-2	14-2	14-2	54-3	72-16	72-16	52-21	9-1	Hot Spring	Hot Spring	Seep	Seep
Reference <sup>2</sup>	1	1	2	2	3	2	2	4	5	6	6	7	7
Collection Date	5/78	7/78	11/77	11/77	11/79	4/77	4/77	11/78	10/75	11/50	9/57	5/73	8/75
Na	2070	2340	2150	2200	2320	1800	2000	1900	1780	2080	2500	2400	1800
K	384	419	390	410	461	380	400	216	440	472	488	378	280
Ca	11	6.8	9.2	6.9	8	12.4	12.20	107	69.1	19	22	113	107
Mg	0.28	<0.24	0.6	0.08	<2	0.29	0.29	4.0	1.0	3.3	0	17	23.6
Fe	0.13	<0.02			0.03			6.3	0.370				
Al	0.31	<0.28			<0.5			<0.1			0.04		
Si			229	383	263	238	244	65	178	189	146	36	50
Sr	1.44	1.28			1.2	1.36	1.20						
Ba	0.24	<0.24			<0.5								
As	3.2	3.5	3.0	2.2	4.3								
Li	25	28			25.3	15.0	16.0				0.27		17
Be	0.004	<0.004			0.005								
B	23	25	29	28	29.9	26.4	27.2	27.0	28.2		38	37	29
Ce	<0.20	<0.20			0.27								
F			5.2	4.8	6.8	5.2	5.3	3.6		7.1	7.5	5.2	3.3
Cl			3650	3650	3860	3110	3260	2881.6	2860	3810	4240	3800	3200
HCO <sub>3</sub>					232	181	181	615.0	485	158	156	536	300
SO <sub>4</sub>			78	60	72	33	32	85	120	65	73	142	70
NO <sub>3</sub>							1.3	<0.2	1.9	11	tr.	tr.	
T.D.S. <sup>3</sup>			>6614*	>6745*	7504	6074	6444	5677	5715*	7040	7800	7506	5948
pH (collection T)			5.9	6.2		7.83	7.53	6.8	7.3		7.9	8.2	6.43
T (collection)			14	9				204	225 <sup>6</sup>	85	55	17	28
T (bottom hole) <sup>4</sup>	268	268	268	268	>260 <sup>5</sup>	243	243	204	225 <sup>6</sup>				
Total depth (m) <sup>5</sup>	1862	1862	1862	1862	878	382	382	2289	2098				
Geothermometers													
T (Na-K-Ca)(-Mg) <sup>7</sup>	284	291	286	293	297	289	288	209*	278	283*	284	181*	141*
T (quartz cond) <sup>8</sup>			276	302	263	254	256	156	228	234	212	123	141
T (quartz diab) <sup>8</sup>			244	268	234	227	229	149	207	211	194	121	136

<sup>1</sup>For well locations, see Figure 1. Element concentrations are reported in mg/l and temperatures in °C. A blank indicates data not determined or information not available, and tr indicates trace amounts measured.

<sup>2</sup>References: 1 = Bamford et al. (1980), 2 = Thermal Power Co. (1978), 3 = This report, 4 = Getty Oil Co. (1978), 5 = S. D. Johnson (personal communication, 1980), 6 = Mundorff (1970), 7 = Lenzer et al. (1976).

<sup>3</sup>Total dissolved solids. Starred values were calculated in this study by summing ion concentrations (Hem, 1970).

<sup>4</sup>Glenn and Hulén (1979).

<sup>5</sup>Koenig and Gardiner (1977).

<sup>6</sup>Geothermal Resources Council Bulletin, 1979, P. O. Box 98, Davis California, Vol. 8, No. 8, p. 6.

<sup>7</sup>Calculated using the methods of Fournier and Truesdell (1973, 1974). The starred values are magnesium corrected (Fournier and Potter, 1979).

<sup>8</sup>Calculated using the methods of Fournier (1977).

<sup>9</sup>Elements analyzed for but present at concentrations less than ICPC limits of quantitative detection (Bamford et al., 1980) include Mn, Cu, Pb, Zn, P, Ti, V, Cr, Co, Ni, Mo, Cd, Ag, Au, Sb, Bi, U, Te, Sn, W, Rb, La, and Th.

<sup>10</sup>Sample supplied by J. R. Bowman, Univ. of Utah. Silica was not preserved in this sample and therefore is not reported.

<sup>11</sup>Sample supplied by A. H. Truesdell, U.S.G.S. Silica was not preserved in this sample and therefore is not reported.

<sup>12</sup>Chemical analyses on this sample were completed on fluid filtered to 0.45µ in the field as follows: fluoride, chloride, and total dissolved solids were determined employing specific ion electrode, silver nitrate titration, and gravimetric methods, respectively; sulfate was determined gravimetrically on samples treated with 1% acid in the field; all other elements were determined by Inductively Coupled Plasma Quantometer on fluid diluted with 20% nitric acid in the field.

Table 2. Fraction of Steam Separated from Flashed Well Fluids

Well	Sample No. <sup>1</sup>	Collection Pressure <sup>2</sup> (atm)	Collection Temperature <sup>3</sup> (°C)	Reservoir Enthalpy (cal/g)	Reservoir Temperature <sup>4</sup> (°C)	Steam Fraction
14-2	1,2,3,4	12.83 <sup>5</sup>	192	277 <sup>5</sup>	265	0.18
54-3	7	23.61 <sup>6</sup>	222	277 <sup>7</sup>	265	0.11
72-16	8,9	20.38 <sup>5</sup>	214	250 <sup>5</sup>	242	0.07

<sup>1</sup>See table 1.

<sup>2</sup>At the separator. These are absolute values excepting for 54-3 (see footnote 6).

<sup>3</sup>Determined from the collection pressure assuming liquid-vapor equilibrium (Keenan et al., 1969).

<sup>4</sup>Determined from the reservoir enthalpy assuming liquid-vapor equilibrium (Keenan et al., 1969).

<sup>5</sup>Thermal Power Co. (1978).

<sup>6</sup>Written communication (S. D. Johnson, 1980). This is a gauge pressure rather than absolute, therefore the calculated steam fraction is a minimum value.

<sup>7</sup>Data is lacking on the reservoir enthalpy at the base of well 54-3, therefore the enthalpy is assumed equivalent to that of the nearby producing well 14-2.



Table 3. Composition of Reservoir Fluids from Well 14-2

Element	Average analysis	Average analysis corrected for	
		steam loss	steam loss gas loss scaling
Na (mg/l)	2190	1796	1796
K	401	329	329
Ca	8	7	12
Mg	0.32	0.26	0.26
Fe	<0.08	<0.06	<0.06
Al	<0.30	<0.24	<0.24
Si	341	280	280
Cl	3650	2993	2993
C	206	206	1793
S	69	57	138
T.D.S. <sup>a</sup>	6680	-	9707
Geothermometers			
T(Na-K-Ca) <sup>b</sup> °C			277
T(SiO <sub>2</sub> cond.) <sup>c</sup> °C			269

<sup>a</sup>T.D.S. represents total dissolved solids calculated by the method of Hem (1970). In accord with Hem's treatment of HCO<sub>3</sub><sup>-</sup>, H<sub>2</sub>CO<sub>3</sub> is converted by a gravimetric factor (H<sub>2</sub>CO<sub>3</sub> (mg/l) x 0.4837 = CO<sub>3</sub> (mg/l)) which assumes half of the H<sub>2</sub>CO<sub>3</sub> is volatilized as CO<sub>2</sub>. This value is used in the summation.

<sup>b</sup>Calculated using the methods of Fournier and Truesdell (1973, 1974).

<sup>c</sup>Calculated using the methods of Fournier (1977).

Table 4. Specific Equations for Calculating Component Activities

Component	Activity <sup>a</sup>
$KAl_2(AlSi_3O_{10})(OH)_2$	$(X_{K^+,A}) (X_{Al^{+3},M(2)})^2 (X_{Al^{+3},T10}) (X_{Si^{+4},T1m}) (X_{Si^{+4},T2})^2 (X_{OH^-})^2$ <sup>b,c</sup>
$Al_2Si_4O_{10}(OH)_2$	$(X_{V,A}) (X_{Al^{+3},M(2)})^2 (X_{Al^{+3},T10}) (X_{Si^{+4},T1m}) (X_{Si^{+4},T2})^2 (X_{OH^-})^2$ <sup>b,c</sup>
$NaAl_2(AlSi_3O_{10})(OH)_2$	$(X_{Na^+,A}) (X_{Al^{+3},M(2)})^2 (X_{Al^{+3},T10}) (X_{Si^{+4},T1m}) (X_{Si^{+4},T2})^2 (X_{OH^-})^2$ <sup>b,c</sup>
$Mg_5Al(AlSi_3O_{10})(OH)_8$	$(X_{Mg^{+2},M})^5 (X_{Al^{+3},M}) (X_{Al^{+3},T}) (X_{Si^{+4},T})^3 (X_{OH^-})^8$ <sup>b</sup>

<sup>a</sup> $a_i = k_i \prod_s \prod_j x_{j,s}^{v_{s,j,i}}$  (equation (46) from Helgeson et al. (1978)), where:  $a_i$  is the activity of the  $i^{th}$  component;  $k_i$  is the proportionality constant (defined by equation (48) for Helgeson et al. (1978)) relating the intracrystalline and intercrystalline standard states;  $x_{j,s}$  is the mole fraction of the  $j^{th}$  species on the  $s^{th}$  site; and  $v_{s,j,i}$  is the stoichiometric number of  $s^{th}$  energetically equivalent sites occupied by the  $j^{th}$  species in one mole of the  $i^{th}$  component.

<sup>b</sup>These equations are consistent with random mixing and equal interactions of atoms on energetically equivalent sites.

<sup>c</sup>These equations are consistent with ordered standard state site distributions, equations and data reported by Helgeson et al. (1978) for muscovite, paragonite and pyrophyllite.

Table 5. Henry's Law Constants

Gas	$K_T$ (atm/mole fraction)				Reference
	150°C	200°C	250°C	300°C	
CO <sub>2</sub> <sup>1</sup>	6750	6500	5380	4100	Ellis and Golding (1963)
H <sub>2</sub> S	1860	2000	1900	1620	Kozintseva (1964) <sup>2</sup>
H <sub>2</sub>	78387	49459	27813	13939	Naumov et al. (1974)
CH <sub>4</sub>	87952	70000	44000	22093	Naumov et al. (1974)

<sup>1</sup>For 0.1 moles/l NaCl solution.

<sup>2</sup>After Ellis and Mahon (1977, p. 132).