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HYDROGEN CHLORIDE IN SUPERHEATED STEAM AND CHLORIDE IN DEEP BRINE
AT THE GEYSERS GEOTHERMAL FIELD, CALIFORNIA

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ABSTRACT

Chloride (Cl) concentrations of 10-120 ppm$_w$ have been measured in superheated steam produced by wells at The Geysers, a vapor-dominated geothermal field in northern California. Corrosion of the well casing and steam-gathering system has been recognized in some parts of The Geysers, and is apparently related to the presence of Cl. Cl in the steam is in a volatile form, generated with the steam at reservoir temperatures, and probably travels to the wellhead as HCl gas. Published experimental data for partial pressures of HCl in steam over aqueous HCl solutions and for dissociation constants of HCl were used to calculate distribution coefficients for HCl. Reservoir liquid Cl concentrations capable of generating steam with the observed Cl concentrations were then calculated as a function of pH and temperatures from 250 to 350°C. Equilibrium mineral/liquid reactions with the K-mica and K-feldspar assemblage found in the wells limit the reservoir liquid pH values at various Cl concentrations to about 5 to 6 (near neutral at 250 to 350°C). Within this pH range, liquid at 250°C could not produce steam containing the high Cl concentrations observed. However, liquid at higher temperatures (300 to 350°C) with chloride concentrations greater than 10,000 ppm$_w$ could generate steam with 10 to over 200 ppm$_w$ Cl. There is a positive correlation between pH and the chloride concentrations required to generate a given Cl concentration in steam. The concentration of Cl in superheated steam constrains not only the reservoir liquid composition, but the temperature at which the steam last equilibrated with liquid.

INTRODUCTION

The Geysers is a vapor-dominated geothermal field in northern California which has been producing steam for generation of electricity since 1960. The Geysers reservoir has been modeled from geochemical, geological, and reservoir engineering viewpoints (e.g., White et al., 1971; Truesdell and White, 1973; D'Amore and Truesdell, 1979; Pruess, 1985; Ingebritsen, 1986, 1987; Truesdell et al.,

1987 and Walters et al., 1988). Although the estimated amount of liquid saturation varies widely, most workers agree that the vast majority of reserves in The Geysers are liquid, but little is known about the character or location of this liquid.

Although liquid samples have been collected from The Geysers reservoir (Table 1), these samples appear to be primarily steam condensate either from condensation in the wellbore or from the reservoir itself. Current models of The Geysers (cited above) emphasize the heat-pipe mechanism in which a deep boiling liquid produces steam that moves upward through major fractures in an otherwise predominantly liquid-saturated reservoir (as in the cracked sponge model, Weres et al., 1977). Steam condenses and transfers heat to the upper part of the system. Steam condensate then moves down to replenish the deep boiling zone. Thus, fluids in The Geysers system consist of steam, condensate, the liquid (probably condensate) in saturated blocks within the reservoir, and probably a deep boiling liquid (brine), which may be of varying degrees of salinity (Haizlip, 1985; Truesdell et al., 1987) and temperature (Drenick, 1986; Walters et al., 1988). Although required by the postulated convection in the reservoir, the deep liquids have never been sampled directly.

It is probable that The Geysers reservoir liquids cannot be sampled with conventional methods, but some characteristics may be calculated from the compositions of steam produced. Until now, most of the components recognized in steam were those which partitioned preferentially into the vapor phase, i.e., noncondensible gases. Recently, however, significant concentrations of chloride (written here as Cl with valence omitted except in chemical equations) have been observed in superheated steam produced in several different areas of The Geysers. Concentrations from 10-120 ppm$_w$ have been measured in deep wells producing highly superheated steam (Table 1, analyses 3-6). Some of these wells have produced for several years; others are at the edge of present development, and have not yet been produced.

Corrosion problems in the steam-gathering system connected to these wells appear to be related to the Cl production.

The purpose of this paper is to evaluate the physiochemical conditions that would produce these Cl concentrations, and thus provide some chemical constraints on the composition and chemical environment in the reservoir. Two-phase or "wet" wells that produce Cl are not discussed here.

Because Cl production appears to be related to corrosion problems, and because the chemical characteristics of the reservoir liquid (salinity in particular) strongly affect the boiling characteristics of a liquid, it is hoped that this paper will assist field operations and physical modeling of The Geysers reservoir.

BACKGROUND

The relation of Cl to corrosion of the production equipment associated with some wells has drawn attention to the concentration of Cl in the steam produced at The Geysers, and to the differences between Cl-producing wells (Table 1, analyses 3-6) and wells that produce low or zero amounts of Cl. Cl analyses in steam from some Geysers wells are available from the past several years. Most results indicated a few to 25 ppm_w Cl in steam samples (e.g., Table 1, analyses 13, 14), but the accuracy of these analyses may be doubtful. These levels were not previously considered significant, but it is now realized that such concentrations in steam can cause corrosion. Because such corrosion was generally not found, it appears that these measurements may have been in error. Recent analyses indicate <1 ppm_w Cl in steam from most wells (Table 1, analyses 1, 17-19), even some for which as much as 25 ppm_w Cl had been reported. Analytical methods used now are designed for the detection of low concentrations rather than accuracy at high concentrations.

THE FORM OF CHLORIDE TRANSPORT

Cl concentrations in steam at the wellhead in high-Cl Geysers wells have been reliably measured between 10 and 120 ppm_w (Table 1, analyses 3-6). Some of this steam is probably a mixture of high-Cl and low-Cl (<1 ppm_w) steam, so Cl concentrations in the high-Cl steam as generated in the reservoir could be 200 ppm_w or higher before mixing (Walters et al., 1988). In order to evaluate the chemistry of Cl in steam and reservoir liquid, the chemical form in which the Cl is transported in the vapor phase must be determined. If Cl was present in steam as an alkali salt (NaCl, KCl, etc.), it would have to move as aerosols or in water droplets, because the volatility of these compounds under reservoir conditions is negligible (Strykyovich, 1957).

Liquid samples from the steam-gathering system contain condensed steam and any materials soluble in liquid at line temperature (about 175°C) and pressure (about 10 bars; see Table 1 for a representative sample), such as chloride, boron, and silica compounds. The condensate collected downstream from a Cl-producing well contains Cl, B, NH4, and minor amounts of SO_4, HCO_3, Fe, Mn, and other heavy metals, with little or no alkali metals. The absence of alkalis indicates that Cl is not being transported from the reservoir as an alkali metal salt. Another possible Cl compound is ammonium chloride. Ammonia is a gas in steam, reacting with CO_2 in condensate to form NH4 and HCO3 ions. There is no indication that NH4Cl exists as a gas or has appreciable volatility (except when it dissociates at 300°C to NH3 and HCl) under reservoir or pipeline conditions (Cotton and Wilkinson, 1972, p. 349).

The most probable form in which Cl is carried in steam is as HCl gas. HCl is a gas at normal and high temperatures (condensing at -84°C) and is moderately soluble in water as HCl (Cotton and Wilkinson, 1972, p. 178; Ellis and Mahon, 1977, p. 288). Its apparent solubility is high at low and moderate temperatures (<250°C) because HCl in solution is a strong acid and dissociates almost completely into H^+ and Cl^-. HCl becomes a weak acid at high temperatures (>300°C) so that un-ionized HCl exists in solution and HCl gas will partition into vapor. HCl is thus found in superheated steam that does not equilibrate with dilute water at moderate (<300°C) temperatures. It is a well-known component of volcanic gases from lavas and high-temperature (>400°C) fumaroles.

Analyses of water from condensate traps of high-Cl wells give clear indications of the presence of HCl in steam (Table 1). The more concentrated of these waters consist almost entirely of Fe^{+3} and Cl with much lower concentrations of B and NH4. Analysis 3 in Table 1 has 185 milliequivalents (meq) of Cl, 215 meq of Fe^{+3}, 7 meq of Mn^{++}, 48 meq of B (H_3BO_3), and 41 meq of NH4. SO_4 and HCO3 were probably present but not analyzed, so exact charge balance is not possible, but the solution is predominantly Cl and Fe^{+3}. $FeCl_3$ is not volatile and Fe and Mn are constituents of steel pipes, so it appears most probable that Fe and Mn are dissolved by the action of H^+ formed by dissociation of HCl when it dissolved in condensate.

Although there are many studies on the dissociation of HCl in solution, there appears to be little data on the solubility of gaseous HCl at high temperatures. In the following section we use the available data to estimate the solubility of HCl and then, using dissociation constant data, estimate the chemical character of waters in equilibrium with high-Cl steam.

CALCULATIONS

A two-part calculation constrains the temperature and composition of liquids that could generate high-Cl Geysers steam. First, the distribution coefficient for HCl is calculated, then this data and the dissociation constants of HCl are used to calculate the concentrations of Cl in liquid in equilibrium with high-Cl (10-200 ppm_w) steam. Second, the pH of the reservoir liquid at various Cl concentrations is calculated from the reaction between K-mica and K-feldspar.

The Distribution Coefficient of HCl

The concentration of associated HCl in aqueous solution is determined by the dissociation reaction:

$$HCl = Cl^- + H^+, \qquad (1)$$

Therefore, $Kd = [Cl^-][H^+]/[HCl] \qquad (2)$

where Kd = the dissociation constant for HCl. The log of the concentration of HCl in the liquid phase is thus:

$$\log m_{HCl,1} = \log m_{Cl^-} + \log \gamma_{Cl^-}$$
$$- pH - \log Kd - \log \gamma_{HCl} \qquad (3)$$

where, $m_{HCl,1}$ = moles/kg HCl in solution; m_{Cl^-} = moles/kg Cl in solution; and γ_{Cl^-} and γ_{HCl} = the activity coefficients of Cl^- and HCl, respectively. The concentration of HCl in equilibrium with the liquid phase is determined by the distribution coefficient, B_{HCl}, where,

$$B_{HCl} = m_{HCl,v}/m_{HCl,1} \text{ and thus} \qquad (4)$$

$$\log m_{HCl,1} = \log m_{HCl,v} - \log B_{HCl}, \qquad (5)$$

where $m_{HCl,v}$ = moles/kg HCl in the vapor phase. Since γ_{Cl^-} and γ_{HCl} can be estimated at a given temperature using the Debye-Huckel theory, if Kd and B_{HCl} are known then m_{Cl}, representing the reservoir liquid chloride composition, can be calculated as a function of pH and T using the following equation:

$$\log m_{Cl^-} = \log m_{HCl,v} - \log B_{HCl} -$$
$$\log \gamma_{Cl^-} + pH + \log Kd + \log \gamma_{HCl} \qquad (6)$$

Perry and Chilton (1973) obtained experimental data on the partial pressure of HCl over various concentrated HCl solutions for the temperature range 0 to 110°C. The concentrations of undissociated HCl in solution were calculated using Kd values from Helgeson (1969) and equation (2). From HCl concentrations in the vapor phase (calculated from the partial pressure of HCl), and the liquid phase (HCl in solution, above), B_{HCl} was calculated as a function of temperature (Figure 1). B_{HCl} values lie between those of B_{H2S} and B_{NH3}, indicating that HCl partitions into the liquid phase slightly more readily than H_2S but not as easily as NH_3. These results are consistent with the suggestion of Ellis and Mahon (1977,

p. 288) that the distribution of HCl gas between liquid and vapor is similar to H_2S. The results were extrapolated to higher temperatures, using B equals 1 at 374°C (true for all gases) and making the curve similar to those of H_2S and NH_3 (Figure 1). A linear regression on the data gives:

$$\log B_{HCl} = 3.5597 - 0.00974 \, t \; (R^2 = .958), \quad (7)$$

where t = temperature in °C.

Cl in Reservoir Liquid

The Cl concentration of the reservoir liquid can be calculated using equation (6). We used the HCl dissociation constant data of Ruaya and Seward (1987), and calculated activity coefficients of Cl and HCl from the extended Debye-Huckel equation (Henley et al., 1984). Using B_{HCl} values calculated from (7), the Cl concentrations in reservoir liquid as a function of HCl concentrations in vapor were calculated at 250°C, 300°C, and 350°C, and at pH values from 2 to 7 (Figures 2-4). The results indicate that at 250°C (Figure 2), even with 100,000 ppm_w Cl in the liquid, the pH would have to be less than or equal to 3.5 to generate 100 ppm_w Cl in steam. However, at 350°C (Figure 4) vapor with 100 ppm_w Cl or greater can be in equilibrium with liquids of pH from 5 to 6 and chloride concentrations of 10,000 to 80,000 ppm_w. Liquids at 300°C (Figure 3) are intermediate. Thus, the calculations indicate that there is a positive correlation between liquid pH and the chlorinity values required to generate steam with a given Cl concentration. The higher the pH, the higher Cl concentration required to volatilize the same amount of HCl. There is also a positive correlation between pH and the temperature required to generate steam with a given Cl concentration. At lower temperatures, a lower pH is required to volatilize HCl. The activity coefficients used are probably not acceptably accurate for solutions with Cl >35,000 ppm_w (1 m), although such hypersaline brines could be generated in the reservoir by evaporation.

pH Buffered by Mineral/Water Reactions

Potassium mica and potassium feldspar are present as hydrothermal alteration minerals in The Geysers reservoir at all temperatures encountered (Walters et al., 1988). If it is assumed that the pH of the liquid is buffered by the reaction between potassium mica and potassium feldspar in the presence of quartz, then

$$3 \, KAlSi_3O_8 + 2 \, H^+ = KAl_3Si_3O_{10}(OH)_2 +$$
$$6 \, SiO_2 + 2 \, K^+, \qquad (8)$$
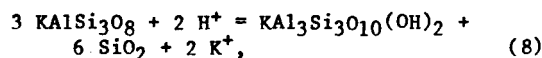
and the activity ratio of K^+ to H^+ is a function of temperature. Using this ratio, electrical neutrality, and the Na/K ratio from the empirical Na-K geothermometer, a

relation between chlorinity (Cl) and pH can be derived at various temperatures (Henley et al., 1984). The results (Figure 5) indicate that at Cl concentrations less than 50,000 ppm_w Cl, the pH of the reservoir liquid is greater than 6.25.

The pH required at 250°C to generate significant Cl in steam cannot exist in the reservoir in equilibrium with K-feldspar and K-mica; therefore it is unlikely that steam generated (boiled) at this temperature would contain Cl. If Cl concentrations in reservoir liquid are between 10,000 and 25,000 ppm_w, and the temperature is 300°C or 350°C at a pH of 6, steam with Cl concentrations between 10 and 200 ppm_w could easily be generated.

DISCUSSION AND CONCLUSIONS

Steam with negligible (<1 ppm_w) Cl has probably been generated by boiling from liquid in parts of The Geysers reservoir at less than 250°C, or has passed through zones of liquid saturation at those temperatures. Condensate contains little Cl (Table 1) and can be in equilibrium with 10 ppm_w Cl steam only at very low pH (<1?; Figure 2). Waters of this acidity would alter aluminosilicate minerals to kaolinite or other aluminum oxides according to temperature (Henley et al., 1984). Since feldspars and mica, rather than kaolinite or other Al oxides, are found in The Geysers reservoir (Sternfeld, 1981; Walters et al., 1988), it is unlikely that such acid reservoir waters exist. Mineral-liquid equilibrium for the reaction of potassium mica and potassium feldspar indicate that pH values for reservoir waters with reasonable chloride concentrations are near neutral (neutral pH = 5.6 at 250°C; 5.7 at 350°C). Thus, steam and steam condensate at typical Geysers reservoir temperatures (220-250°C) probably contain negligible Cl. Because steam condensate is nearly ubiquitous, Cl would be absorbed in the liquid before reaching a well.

Steam that contains significant (greater than 10 ppm_w) Cl must have been generated at temperatures above typical Geysers reservoir temperatures and must have followed a dry or high-temperature path to the wellhead. Specifically, at 350°C, steam with greater than 100 ppm_w Cl can be generated (boiled) from liquid at pH 5 with greater than 10,000 ppm_w Cl and at pH 6, greater than 90,000 ppm_w Cl. At this temperature and range of pH, liquid with greater than 10,000 ppm_w Cl would generate steam with greater than 10 ppm_w Cl. At 300°C, liquid with pH less than 5 and Cl greater than 70,000 ppm_w is required to generate steam with 100 ppm_w Cl (Figures 3 and 4). For the high-temperature (350°C) steam with 200 ppm_w Cl suggested by Walters et al. (1988) to exist in the high-temperature reservoir of the Northwest Geysers, liquid Cl concentrations must be greater than 25,000 ppm_w.

From the limits on pH from reservoir mineralogy, temperatures, and the inferred concentration of Cl in the reservoir liquid, we reach the following conclusions:

1. Reservoir pH is around neutral at The Geysers at 250 to 350°C.

2. Steam containing significant (>1 ppm_w) Cl concentration is not generated at typical Geysers temperatures.

3. Steam boiled at higher temperatures can contain the observed concentrations of Cl if near-neutral reservoir liquid has chlorinity between 10,000 and 80,000 ppm_w.

Thus, the source of the chloride is a hot (>300°C) saline (10,000-80,000 ppm_w) brine. Wells at The Geysers that produce chloride must have some steam derived from this brine. The existence of such a deep boiling brine is proposed by many models of The Geysers (e.g., White et al., 1971; Pruess, 1985) but without direct evidence. These conclusions also support the results of isotopic and noncondensible-gas studies, which have proposed that The Geysers contains reservoir liquids of a variety of salinities, with saline, possibly connate, water in parts of The Geysers and meteoric water in others (Haizlip, 1985; Truesdell et al., 1987). Water from hot springs to the east (Table 1) may be similar in composition to the saline reservoir water, as suggested from isotopic evidence.

Evidence presented by Drenick (1986) and Walters et al. (1988) suggests that Cl-bearing steam is produced from higher temperature reservoirs at The Geysers and that some wells produce mixed steam from both the high-temperature and typical reservoirs. Our work shows that steam passing through a two-phase zone containing condensate at around 250°C would be scrubbed of chloride. Therefore, the suggested mixing must occur in the well; or else the conduit through the typical reservoir must be dry. In newly developed areas like the Northwest Geysers, the former is probably true. In wells that have produced for a long time, the shallower typical reservoir may have become dry due to production.

Our results are in agreement with observations of corrosion from HCl-bearing superheated steam at well 12 of the Krafla, Iceland field (Haldur Armannsson, pers. commun., 1987) and with the occurrence of acid, high-Cl condensate from steam at Tatun, Taiwan (Ellis and Mahon, 1977, p. 289), although liquids in these systems may be much more acid than those at The Geysers. The appearance of Cl in steam at Larderello in the 1960s (D'Amore and Truesdell, 1979) probably resulted from the drying out of the 240±°C reservoir rather than from a sudden shift to production from deep brine.

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Table 1. Chemical Analyses of Water and Steam Condensate from The Geysers and Surrounding Areas [in mg/L, except $\delta^{18}O$ in °/°°]

No.	Fluids from Wells	pH	SiO_2	Ca	Mg	Na	K	Li	HCO_3	SO_4	Cl	F	B	Fe	Mn	NH_4	$\delta^{18}O$
	CENTRAL GEYSERS																
1	Condensed steam typical well[1]	~6.3	0.8	–	–	–	–	–	*	*	≤1.0	*	21	–	–	*	-4.7
2	Water from condensate trap – typical well[1]	~5.9	4.1	1.29	–	–	–	–	*	*	6.0	*	521.5	–	–	*	*
3	Condensed steam Cl well[1]	*	–	–	–	–	–	–	*	*	13.0	*	34	–	–	*	-2.7
4	Water from condensate trap – Cl wells[1]	*	14.7	–	–	–	–	–	*	*	4900	*	11	1901	67	*	*
5		4.3	48.0	47.3	27.7	21.1	*	–	*	*	3885	*	664	3011	180	313	*
6		3.1	88.6	34.0	20.2	16.8	*	–	*	*	6575	*	520	4006	186	748	*
	NORTHWEST GEYSERS																
7	Condensed steam	~5.9	–	–	–	0.08	–	–	*	*	46	*	58	–	–	*	+3.0
8	Cl wells[1]	*	–	–	–	0.15	–	–	*	*	120	*	106	–	–	*	+2.0
9		*	0.8	–	–	0.3	–	–	*	*	88	*	80	–	–	*	+1.2
	SOUTHEAST GEYSERS																
10	Water entry original hole[2]	*	*	<10	*	146	110	*	*	*	41	*	82.8	*	*	*	*
11	Water entry redrill[2]	*	400	10	37	117	86	*	12.2	414	10	8	24.1	*	*	43.1	*
12	Steam condensates[2]	*	1.2	<0.1	0.005	<0.1	<0.1	*	80	10	2	<0.01	2.4	1.0	*	25.2	*
13	Steam condensates[3]	6.1	4.3	0.1	0.04	<0.5	<0.5	<0.01	95	4	7.1	<0.1	0.1	–	–	5	-6.3
14		5.7	4.3	<0.1	0.04	<0.5	<0.5	<0.01	46	7.3	10	<0.1	1.2	–	–	9	-4.8
15	Ejected waters[3]	8.9	42	1.8	1.0	18	7.8	0.12	64	9	7.1	0.5	42	–	–	–	-1.3
16		7.8	80	2.9	0.7	71	12	0.6	162	40	18	3.7	–	–	–	–	+0.8
	GEYSERS																
17	Condensed steam Well 1[4]	5.3	<1.0	*	*	<0.1	<0.1	*	*	*	<0.01	<0.04	0.47	*	*	*	*
18	Water from condensate trap – Well 1[4]	5.5	27.0	*	*	4.5	0.5	*	*	*	0.08	0.4	150	*	*	*	*
19	Condensed steam Well 2[4]	*	<1.0	*	*	<0.1	<0.1	*	*	*	0.09	<0.04	2.8	*	*	*	*
20	Water from condensate trap – Well 2[4]	*	15.0	*	*	0.2	0.2	*	*	*	0.14	0.1	150	*	*	*	*
	NATURAL WATERS																
21	Witches' Cauldron, The Geysers[5]	7.0	66	58	108	18	6	*	176	776	1.5	*	15	*	*	111	*
22	Devil's Kitchen, The Geysers[5]	1.8+	225	47	281	12	5	*	0	5710	0.5	*	3.1	63	1.4	1400	*
23	"Geyser" Spring Sulfur Bank Mercury Mine[6]	6.8	42	20	55	1190	23	4.4	3290	598	644	1.0	620	0.0	0.1	464	+5.41
24	Wilbur Hot Springs	6.9	200	28	38	8500	440	*	7310	390	9700	2.5	310	0.04	*	280	+3.24

NOTES
–, Not detected
*, Not analyzed
[1], Data available courtesy of Geothermal Resources International, Inc.
[2], Frye, 1975
[3], Truesdell et al., 1981
[4], Smith et al., 1987
[5], White et al., 1963
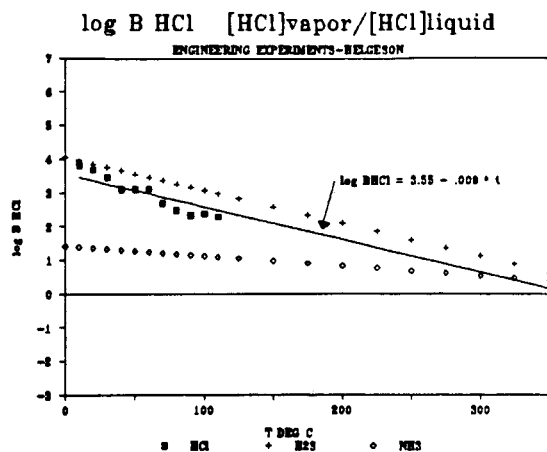[6], White et al., 1973

log B HCl   [HCl]vapor/[HCl]liquid

Figure 1. Log of the distribution coeffi-
cients of HCl, B_{HCl}, calculated from vapor
pressure data (Perry and Chilton, 1973), and
the Kd of Helgeson (1969). B_{H2S} and B_{NH3}
added for reference are from Giggenbach
(1980). Linear regression line calculates
to be: $\log B_{HCl} = 3.56 - .0097$ (t in °C).

HCl PARTITIONING
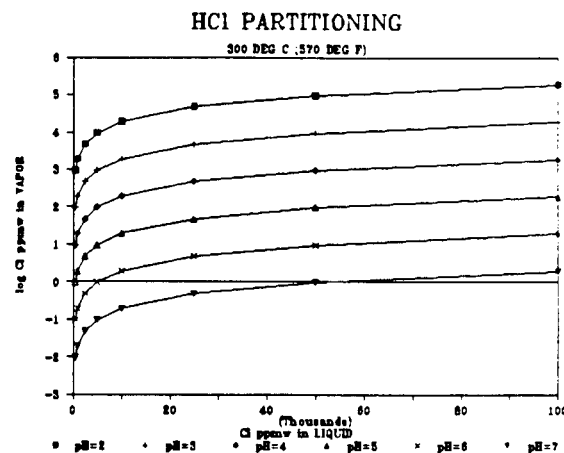

Figure 3. Cl in reservoir liquid versus Cl in
reservoir vapor, in ppm_w. Calculated using
B_{HCl} derived from experimental data and Kd
of Ruaya and Seward (1987) at 300°C.

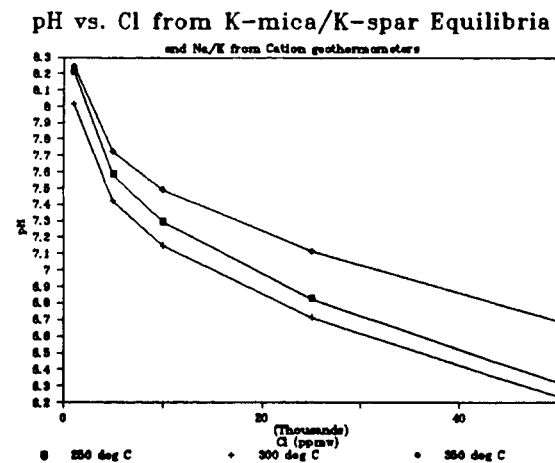## pH vs. Cl from K-mica/K-spar Equilibria

Figure 5. pH versus Cl in reservoir liquid
in equilibrium with potassium feldspar and
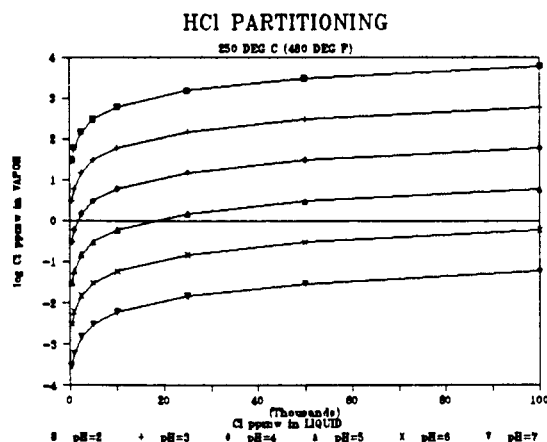potassium mica. Equilibrium constants from
Henley et al., (1984).

HCl PARTITIONING

Figure 2. Cl in reservoir liquid versus Cl
in reservoir vapor, in ppm_w. Calculated
using B_{HCl} derived from experimental data
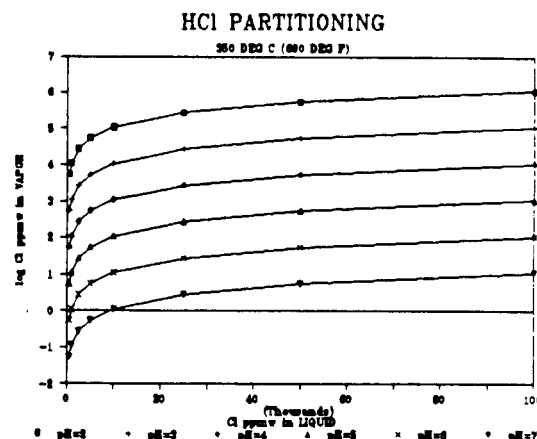and Kd of Ruaya and Seward (1987) at 250°C.

HCl PARTITIONING

Figure 4. Cl in reservoir liquid versus Cl
in reservoir vapor, in ppm_w. Calculated
using B_{HCl} derived from experimental data
and Kd of Ruaya and Seward (1978) at 350°C.