GAS GEOTHERMOMETRY FOR DRILLHOLE FLUIDS FROM VAPOR DOMINATED AND HOT WATER GEOTHERMAL FIELDS

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INTRODUCTION

The compositions of steam from the vapor-dominated geothermal systems of Larderello, Italy and The Geysers, California have been shown by previous investigators to vary with position in the field (Celati et al., 1973; Panichi et al., 1974; D'Amore et al., 1977; Truesdell et al., 1977; Truesdell and Nehring, 1978; Mazor, 1978). The most conspicuous chemical patterns observed in the Larderello and The Geysers vapor-dominated geothermal areas are strong increases or decreases from the center to the edges of constituents carried in the steam. The pattern of these parameters in vapor-dominated systems seem to be controlled mainly by a process of lateral steam movement and condensation (D'Amore and Truesdell, 1979; Calore et al., 1980).

The condensation process, at constant temperature and total pressure increases the partial pressure of CO_2 at the same rate as the gas/steam ratio, strongly affecting the composition of the total gas (including steam). The condensation effect should increase contents of CO_2, H_2S, H_2 and CH_4 in the residual steam to about the same degree because their solubilities are similar. However, the general trend observed is almost constant ratios of H_2S, H_2, CH_4 to H_2O as the CO_2/H_2O ratio increases in Larderello about 5-6 times from the center to the edges at constant temperature. This means that the H_2/CO_2, CH_4/CO_2, and H_2S/CO_2 ratios decrease with increasing CO_2/H_2O ratios. Apparently the only mechanism that can explain this behavior is reaction of these three gases with other gases and with reservoir minerals so that the partial pressures of these gases are buffered by temperature-dependent reactions with water and rock minerals.

GEOTHERMOMETERS

A system of appropriate equations involving H_2O, CO_2, H_2, CH_4, H_2S can be set up. Table 1 shows the equations and their equilibrium constants as functions of temperature in degrees Kelvin.

Equation 2 expresses a possible double origin for CH_4, from carbon dioxide and from free carbon via hydrogen reduction. Free carbon as carbonaceous minerals has been found in cuttings from wells at Larderello, Cerro Prieto, and elsewhere. Hydrogen partial pressure can be calculated from the dissociation reaction of water (reaction 1), by fixing temperature and oxygen fugacity.

Equations 3 and 4 express the control of sulfur fugacity by magnetite, pyrite and oxygen fugacity and the control of H_2S partial pressure by sulfur fugacity and hydrogen partial pressure. In general, gas equilibrium equations are expressed in fugacities but at the relatively low pressures of geothermal systems these are equivalent to partial pressures. The term fugacity is retained here only for oxygen and sulfur which are not measureable in the well discharges but important in expressing reservoir conditions.

Equation 5 expresses the partial pressures of oxygen determined by two extreme oxygen buffer systems:

$CaCO_3$-$CaSO_4$-Fe_3O_4-FeS_2-CO_2

C-(FeS)-FeS_2-Fe_3O_4-CO_2

and equation 6 the fugacity of saturated steam. Assuming ideal gas behavior, P_{CO_2} is a function of Nl_{CO_2}/Kg_w as in equation 7.

$$P_{CO_2} = P_{H_2O}\,(Nl_{CO_2}/Kg_w)/(55.51 \times 22.4) \qquad (7)$$

where (Nl_{CO_2}/Kg_w) is the gas/steam ratio as liters of CO_2 at standard conditions (STP) per kg of steam.

From equations 1 to 7 the following equations have been obtained, relating the gas composition to temperature (°K) and gas/steam ratio.

$$\log (P_{H_2}/P_{CO_2}) = 4.34 - 2396/T - \log (Nl_{CO_2}/Kg_w) \qquad (8)$$

$$\log (P_{CH_4}/P_{CO_2}) = 16.87 - 4318/T - 3.56 \log T - 1/2 \log (Nl_{CO_2}/Kg_w) \qquad (9)$$

$$\log (P_{H_2S}/P_{CO_2}) = 7.74 - 3024/T - 0.79 \log T - \log (Nl_{CO_2}/Kg_w) \qquad (10)$$

$$\log (P_{H_2}/P_{CO_2}) = 5.52 - 3406/T - 1/2 \log (Nl_{CO_2}/Kg_w) \qquad (11)$$

$$\log (P_{H_2S}/P_{CO_2}) = 8.92 - 4033.5/T - 0.79 \log T - 1/2 \log (Nl_{CO_2}/Kg_w) \qquad (12)$$

Equations 11 and 12 must be considered semi-empirical, because the H_2/CO_2 and H_2S/CO_2 parameters are arbitrarily taken as function of $(P_{CO_2})^{1/2}$ instead of P_{CO_2} equations 8 and 10.

The geothermometers have been tested on data, reported in table 2, from the Italian fields of Larderello, Travale, Bagnore, Piancastagnaio and from The Geysers, U.S.A. For the central Larderello ("classic") area, 70 samples have been ranked for increasing gas/steam ratio. Table 3 shows the temperatures computed from equations 8, 11, 9, 10 and 12. Equation 8 can be used satisfactorily for the Italian geothermal fields, except where the gas/steam ratios are very high (more than 50 liters of gas per kg of steam). Equation 9 fails for CH_4/CO_2 ratios of more than 0.05. In the Larderello and Travale fields only, reliable results can be obtained computing the average of the three temperatures calculated from equations 8, 9 and 10. The most reliable results, however, are provided by equations 10 and 12. Equation 12 indicates reasonable temperatures for all these vapor-dominated geothermal fields.

In figure 1, equation 12 is plotted at selected temperatures along with the analytical data for Larderello, The Geysers and Travale fields which exhibit a wide range of gas/steam ratios. An attempt has been made to apply equation 12 to all of Larderello geothermal field. Figure 2 shows the computed isotherms at 250, 225, and 200°C. The full circles indicate temperatures of more than 250°C. The only wells shown are those for which recent (1978-1980) data are available.

RESERVOIR TEMPERATURE EVOLUTION OF SINGLE WELLS

Well-head temperatures of single wells change with time but these temperatures are a complex function of flow rate and do not directly indicate changes in reservoir temperatures. The gas geothermometers have been applied to steam samples from wells FBN and ALR in order to define changes in both well and reservoir temperatures. Both these wells are situated in the central part of the Larderello classic area and have produced steam for about 40 years. Gas/steam ratio and dry gas composition evolve with time. As an example, the evolution of steam H_2S contents in dry gas for well ALR and of gas/steam ratios for three wells in Larderello is shown in figures 3 and 4.

In figures 5 and 6 the well-head temperature, well-bottom temperature computed by the methods described by Nathenson (1975), and the average temperature computed using H_2S, H_2, CH_4 geothermometers (equations 8, 9, and 10) are plotted as function of time. The agreement of geothermometer temperatures with computed well-bottom temperatures is very good. The increase of temperature in the first 25 years could be related to a change in the source of steam from wetter to drier parts of the reservoir. During production, decrease of pressure causes the flashing of the liquid water to steam and a decrease of temperature (Truesdell and White, 1973).

A completely dry system will have the same temperature with exploitation and a constant decay of flow rate. The decrease of steam flow rate with time observed at Larderello is generally exponential and only a considerable amount of liquid water at starting production can explain this strong decrease in flow rate during the first years of exploitation. If the gases retain a memory of the original reservoir temperature and are not greatly influenced by the rapid decrease of temperature caused by flashing, the difference between the well-bottom temperature and that indicated by gas geothermometers should be related to the ratio of liquid water to rock in the producing reservoir. As shown in D'Amore et al. (1978) from an application of Rn^{222} as tracer, the transit time of steam in Larderello from the two-phase, vapor-dominated reservoir to the well bottom is of the order of a few days. This travel time is too short to allow significant reequilibration of gas reactions at the new temperature conditions.

In figure 7 the volume per cent of liquid water in total rock is plotted versus the difference between the reservoir temperature and well-bottom temperature affected by flashing (data from Truesdell and White, 1973). The starting temperature to plot the diagram is 240°C but the computed temperatures differences are also approximately correct for a starting temperature of about 250°C.

LIQUID WATER IN THE RESERVOIRS OF LARDERELLO AND THE GEYSERS

Table 4 shows the approximate volume per cent of liquid water in the part of the reservoir feeding the wells ALR and FBN from 1940 to 1978, computed as described by Truesdell and White (1973) from differences in temperatures given by the H_2S geothermometer (equation 12) and the well-bottom temperatures calculated according to Nathenson (1975). The differences between the bottom-hole temperatures and a constant reservoir temperature of 255°C are also given.

As the flow rate decreases, the volume per cent of liquid water in the producing reservoir decreases regularly. After a broad maximum from 1950 to 1960, the computed temperatures in reservoir and at the well bottom decrease moderately probably due to the increasing contribution of the steam directly coming from flashing of deep brine. The temperature decrease is moderate because steam temperatures are buffered by passage through the dry but still hot reservoir. Geothermometer temperatures agree with bottom-hole temperatures because the long transit time has allowed reequilibration of the gases. When the steam temperature starts to drop, chloride ion in the steam for these wells starts to increase strongly. This is consistent with a contribution of steam coming from a chlorine-rich water such as the hypothesized deep brine. The flow rate continues to decrease because of the strong exploitation in this part of the field.

It is interesting to note that the leveling off of the flow rate from its initial large values coincides with the achievement of near maximum well-bottom temperatures. Flashing of liquid from the upper condensate layer near the well bottom produces a large flow of relatively low temperature steam. When this is exhausted steam is supplied from drier parts of the reservoir at greater distances from the well. Flashing of the deep brine produces no increase in flow because it is distant from the well bottom. Because the brine zone is saturated the steam flow path may be expected to increase more slowly and the rate of flow decline may decrease.

The calculated percentage of liquid water in the reservoir rock is shown for a number of The Geysers wells in Table 5 (data courtesy of Union Oil Co.). The temperature has been computed starting from a reservoir temperature of 247°C based on the maximum sea level pressure of 36 bars for the Sulfur Bank wells (Lipman et al., 1978), and the average depth of The Geysers wells assuming saturated steam. This temperature is consistent with the average temperature (249±10°C) computed by the H_2S geothermometer (equation 12). The computed liquid water contents in the reservoir increase with CO_2/steam ratio as would be expected with increasing condensation. The average water contents are close to 6.5 volume percent, similar to the figure of 5 per cent used by Nathenson and Muffler (1975) to calculate reserves at The Geysers.

The actual content of liquid water at The Geysers is much higher than the present content computed for wells GR1, ALR and FBN of Larderello, but of the same order of the content computed for these three wells in the early years of production. The thickness and water content of the vapor-dominated reservoir appears to differ in the two systems and it is not certain whether steam compositions at The Geysers will go through the same evolution as at Larderello.

The gas geothermometer provides indications of reservoir conditions distant from the well bottoms and allows estimates of the liquid water content of reservoir rocks. Since most steam must be produced from the vaporization of this liquid, the calculation of liquid water content allows estimates of steam reserves within a known volume of reservoir rock.

APPLICATION OF THE GEOTHERMOMETERS TO HOT-WATER FIELDS

Equation 10 has been applied to calculate the reservoir temperatures using computed H_2S/CO_2 ratios in the total fluid of samples from several wells of hot water fields.

The following parameters are used:

P_{sep}	measured pressure at separator (bars)
E_{tot}	measured enthalpy of total fluid at separator (J/g)
E_w	enthalpy of liquid water (J/g)
E_S	enthalpy of steam (J/g)
% H_2S	measured mole per cent of H_2S in the total noncondensable gas
X_{H_2S}	molar fraction of H_2S in total fluid (steam + liquid water)
X_w	fraction of liquid water
X_s	fraction of steam
Y	measured gas/steam ratio (moles/moles)
n	number of moles

The enthalpy balance is:

$$E_{tot} = E_w X_w + E_s(1-X_w)$$

Using the measured P_{sep} and E_{tot}, liquid and steam enthalpies can be computed from steam tables, and then the function of liquid water X_w and steam X_s:

$$X_w = (E_{tot}-E_s)/(E_w-E_s)$$

$$X_s = 1 - X_w$$

From X_s, the measured % H_2S and the gas/steam ratio Y, the molar fraction of H_2S in total fluid, X_{H_2S}, can be computed:

$$X_{H_2S} = \frac{100}{Y \cdot\ X_s\ \%H_2S} + \frac{100}{\%\ H_2S}\ ^{-1}$$

For water-dominated systems it is then convenient to have an H_2S geothermometer as a function of X_{H_2S}, taking into account the solubility of this gas.

The theoretical partial pressure of H_2S is given by developing equations 1, 3, 4, 5, 6 as a function of temperature (°K)

$$\log P_{H_2S} = 10.11 - 5043.5/T - 0.79 \log T$$

By Henry's law, the partial pressure of a gas can be related to the molar fraction.

$$P_{H_2S} = X_{H_2S} \cdot K_{H_2ST}$$

Henry's law constant, K_{H_2S}, is a function of temperature (Kozintseva, 1964). Then:

$$\log X_{H_2S} = 10.11 - 5043.5/T - 0.79 \log T - \log K_{H_2S}(T)$$

The theoretical log X_{H_2S} given by the previous equation is plotted in the figure 8.

The results of these calculations are shown in table 6. The H_2S geothermometer (equation 13) appears to give results consistant with liquid-based geothermometers.

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Table 1.--Basic chemical reactions and equilibrium constants.

Reactions	Equilibrium constants	Regression coefficients A	B	C	Ref
(1) $H_2O_v = H_2 + 1/2\ O_2$	$\log K_1 = A_1 + B_1/T$	2.652	-12,776		a
(2) $C + CO_2 + 6H_2 = 2CH_4 + 2H_2O_v$	$\log K_2 = A_2 + B_2/T + C_2 \log T$	6.82	11,801	-7.11	b
(3) $H_2S = H_2 + 1/2\ S_2$	$\log K_3 = A_3 + B_3/T + C_3 \log T$	-0.13	-4,394.5	0.79	c
(4) $1/3\ Fe_3O_4 + S_2 = FeS_2 + 2/3\ O_2$	$\log K_4 = A_4 + B_4/T$	-4.683	-3,796		d
(5) $f_{O_2} = f_1(T)$	$\log P_{O_2} = A_5 + B_5/T$	2.80	-20,760		
(6) $f_{H_2O} = f_2(T)$	$\log P_{H_2O} = A_6 + B_6/T$	5.451	-2,020		

References: a, Barin and Knacke (1973); b, Ellis (1957), Kubaschewski (1951); c, Holland (1965); d, Helgeson (1969).

Table 2a. Average (and standard deviation) of volume per cent and log P_{gas}/P_{CO_2} for 70 wells of the central "classic" Larderello and Serrazzano zones divided into groups according to gas/steam ratio.

									S-1	S-2	S-3
log Nl/Kg_w (avg.)	1.25	1.36	1.46	1.56	1.66	1.77	1.87	2.02	1.23	1.25	1.41
(V%)H_2	2.9±0.4	2.7±0.4	2.1±0.4	1.8±0.2	1.6±0.2	1.2±0.2	1.0±0.3	0.8±0.2	3.6±0.3	3.8±0.5	3.1±0.2
log(P_{H_2}/P_{CO_2})	-1.50	-1.54	-1.65	-1.72	-1.78	-1.91	-1.99	-2.09	-1.40	-1.38	-1.46
(V%)CH_4	1.5±0.3	1.4±0.2	1.4±0.1	1.4±0.5	0.9±0.4	0.6±0.3	0.4±0.2	0.6±0.4	1.4±0.1	1.7±0.2	1.8±0.1
log(P_{CH_4}/P_{CO_2})	-1.79	-1.82	-1.83	-1.83	-2.03	-2.21	-2.39	-2.21	-1.85	-1.73	-1.71
(V%)H_2S	2.5±0.6	2.3±0.5	1.9±0.1	1.7±0.3	1.4±0.4	1.0±0.3	0.7±0.2	0.5±0.3	3.2±0.2	3.0±0.1	3.1±0.2
log(P_{H_2S}/P_{CO_2})	-1.57	-1.61	-1.69	-1.74	-1.84	-1.99	-2.15	-2.29	-1.45	-1.48	-1.47
(V%)$_{CO_2}$	92.4±1	93.0±1	93.7±1	94.2±1	95.8±1	96.7±1	98.2±1	97.5±1	91.1	90.8	91.7

Table 2b. Carbon dioxide/steam and gas ratios for some high-temperature wells at Serrazzano, for some wells at The Geysers and for reference wells at Monte Amiata and Travale.

	G-1	G-2	G-3	G-4	G-5	G-6	G-7	G-8	G-9	A-1	A-2	T-1
log(Nl_{CO_2}/Kg_w)	0.33	0.12	0.38	0.69	0.56	0.53	-0.73	0.64	-0.62	1.56	2.16	1.66
log(P_{H_2}/P_{CO_2})	-0.45	-0.87	-0.64	-0.66	-0.47	-1.21	-1.42	-0.90	-0.70	-2.44	-2.29	-2.25
log(P_{CH_4}/P_{CO_2})	-0.79	-0.92	-1.39	-0.96	-0.53	-1.70	-1.34	-0.73	-0.15	-1.84	-1.56	-1.58
log(P_{H_2S}/P_{CO_2})	-1.02	-1.16	-1.06	-1.21	-1.15	-1.56	-1.12	-1.11	-0.64	-2.97	-2.37	-1.97

Well names:

Serrazzano area, Larderello
S-1 VC/10
S-2 Querciola 1
S-3 Lustignano 5

The Geysers
G-1 Rorabauch-10
G-2 La Koma-Fame-15
G-3 DX-State-30
G-4 Ottobone-State-15
G-5 Rorabaugh-7A
G-6 DX-27
G-7 CA 956
G-8 Aidlin 1
G-9 CA 1892

Monte Amiata - Travale
A-1 Bagnore 2b
A-2 Piancastagnaio 8
T-1 Travale 23/D

Table 3. Temperatures computed by gas geothermometers (°C)

Larderello (classic area)	T_{H_2}		T_{CH_4}	T_{H_2S}		T_{meas}
log (Nl_{CO_2}/Kg_w)	eq. (8)	eq. (11)	eq. (9)	eq. (10)	eq. (12)	
1.22	245	250	241	235	248	108-260
1.33	253	251	243	241	249	234-252
1.43	252	246	246	243	247	229-244
1.53	256	245	250	248	247	223-245
1.64	262	244	239	249	244	206-242
1.76	260	239	230	246	238	210-222
1.86	263	237	221	240	231	201-222
2.01	269	235	239	241	226	198-233
Serrazzano						
Well VC/10	253	265	235	243	256	260
Querciola 1	258	268	246	242	255	265
Lustignano 5	268	269	253	260	262	270
M. Amiata - Travale						
Well Bagnore 2b	184	200	249	153	173	160
Piancastagnaio 8	263	233	302	248	226	210
Travale 23/D	213	218	276	239	236	240
The Geysers (table 2)						
1	264	313	286	206	256	252±
2	198	265	266	180	239	
3	248	297	240	207	255	
4	283	310	287	220	255	
5	290	323	323	214	255	
6	204	253	222	180	225	
7	96	193	204	128	214	
8	248	285	307	224	260	
9	150	225	221	133	249	247

Table 4. Reservoir temperatures computed from equation (12), well-bottom temperatures and volume percentages of liquid water in the reservoir for two wells of Larderello from 1940-1978.

Well	Year	Q_{steam}	N_{CO_2}/Kg_w	$\log(P_{H_2S}/P_{CO_2}$	T_{H_2S}	T_{WB}	$(T_{H_2S}-T_{WB})$	$(255-T_{WB})$	V% liq. H_2O
FBN	1942	80-97	24	-1.59	252	215	37	40	6.1-6.5
	1951	51	26	-1.56	257	235	22	20	3.3-3.6
	1960-63	28	29	-1.65	252	248	4	7	0.7-1.2
	1970-71	18	29	-1.65	252	251	1	4	0.2-0.7
	1977-78	15	28	-1.69	248	245	3	10	0.5-1.7
ALR	1941	120	16	-1.51	252	216	36	39	5.9-6.4
	1951	38	26	-1.57	255	239	16	16	2.6-2.6
	1965-66	20	43	-1.74	251	250	1	5	0.2-0.9
	1970-71	15	28	-1.67	249	247	2	8	0.4-1.3
	1977-78	10	25	-1.71	246	247	0	8	0 -1.3

Table 5. Reservoir temperatures computed from equation (12), well-bottom temperatures and percentage of liquid water in the reservoir for nine wells at The Geysers in 1977.

Sample	Nl_{CO_2}/Kg_w	$\log(P_{H_2S}/P_{CO_2})$	T_{H_2S} °C	T_{WB}°C	$(V\%)_{liq.\ w}$ 0.5
e	0.79	-1.01	241	232	2.4
a	1.10	-0.95	250	220	4.5
f	1.34	-0.99	251	207	6.5
c	1.53	-1.37	227	207	6.5
b	1.58	-0.98	254	207	6.5
d	1.76	-1.04	251	204	7.0
g	2.08	-1.36	232	204	7.0
h	2.08	-1.22	241	214	5.5
i	2.73	-1.06	257	179	11.1

Table 6 Reservoir temperatures of water-dominated geothermal systems calculated from equation (10) compared with measured temperatures and temperatures from other geothermometers. Data from Truesdell and Singers (1971) except for Cerro Prieto (Nehring and Fausto, 1979) and Namafjall (Arnorsson, 1974).

Well	Temperatures					
	Na-K-Ca	Na-K	Measured	Maximum	H_2S	SiO_2
Cerro Prieto 8	273	278	295	340	298	270
27	268	277	284		312	279
35	287	301	299		291	299
21A	283	301	279		301	280
20	258	262	243		289	256
Wairakei 20	254	262	260	270	231	246
Broadlands 2	297	287	287	307	265(290)	261
Tauhara 1	259	257			248	248
Rotakawa 1	259	255			247	254
Kawerau 8	282	258	260		291	282
Orakeikorakei 3	245	242	237		233(232)	234
Kizildere 1A	243	193		210	216	199
Matsao E205	234	248	240	300	290	245
Otake 7	230	212	195		180(201)	217
Hveragerdi G7	217	193			211(22)	222
4	187	165			219	200
Reykjanes 8	240	235	270		225(233)	251-256
Namafjall 4	237	256	280	290	290	252-207
5	221	237	250		278	264-243
6	245	257	270		306	264-243
7	247	265	290		317	280-243
8	236	257	275		277	241
9	244	266	260		295	254-259
Tatio 2	291	213	226	260	206	218

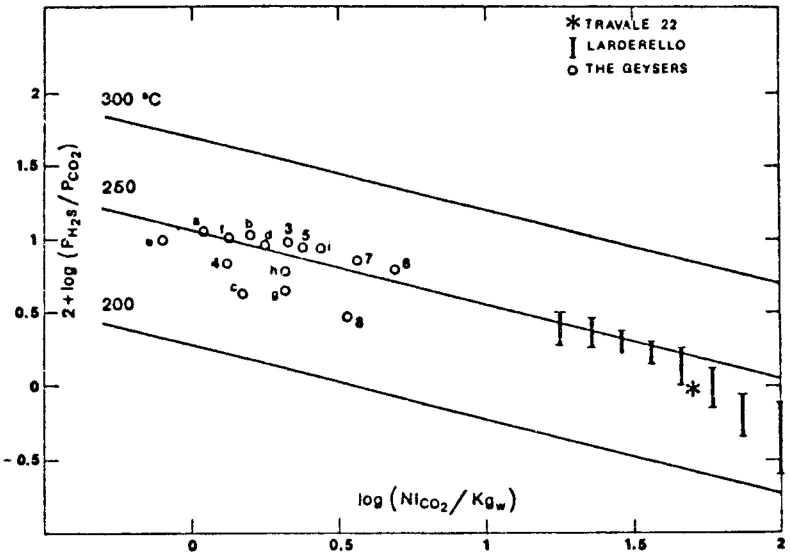

Figure 1

Figure 1. Application of equation 12 to Larderello, Travale, and The Geysers. Actual H_2S/CO_2 ratios (as $\log(P_{H_2S}/P_{CO_2} + 2)$) are plotted vs. gas/steam ratios. Continuous lines show predicted values at selected temperatures.

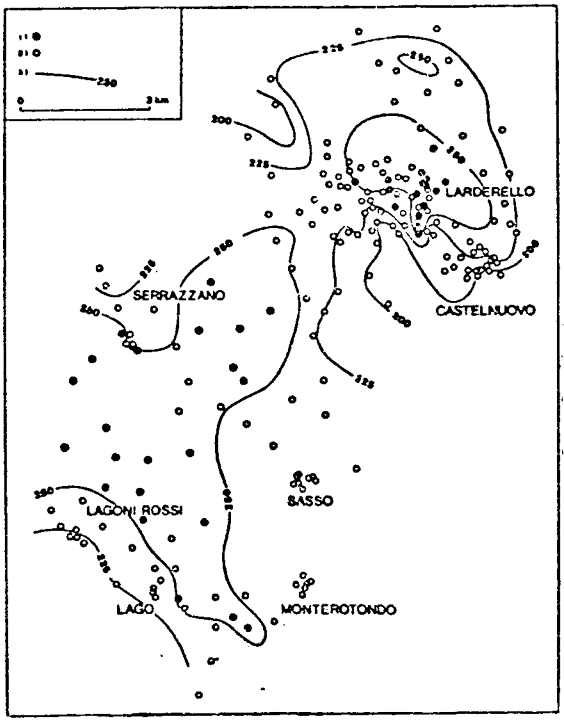

Figure 2

Figure 2. Application of equation 12 to all of Larderello field.
(1) computed T greater than 250°C
(2) computed T less than of equal to 250°C
(3) computed isotherms at 250, 225 and 200°C

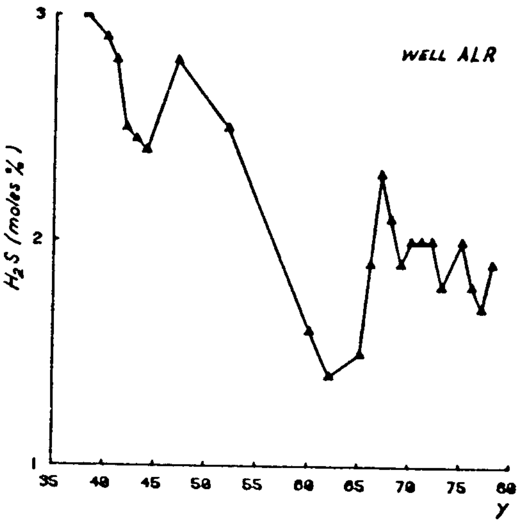

Figure 3

Figure 3. Evolution of H_2S percentage in the dry gas is plotted vs. time (years) for well ALR.

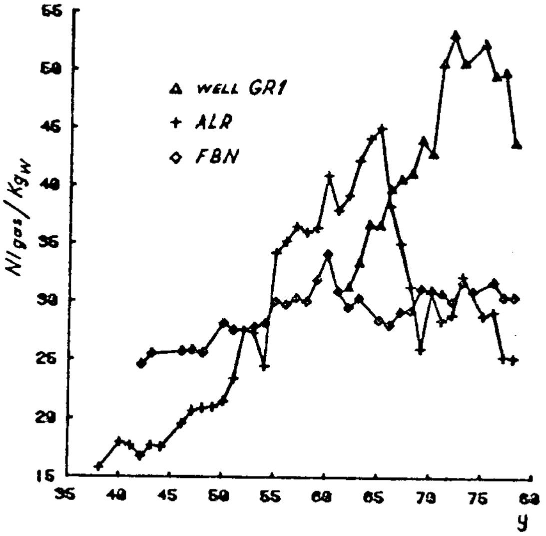

Figure 4

Figure 4. Change with time of gas/steam ratio (Nlgas/Kg steam) for three Larderello wells.

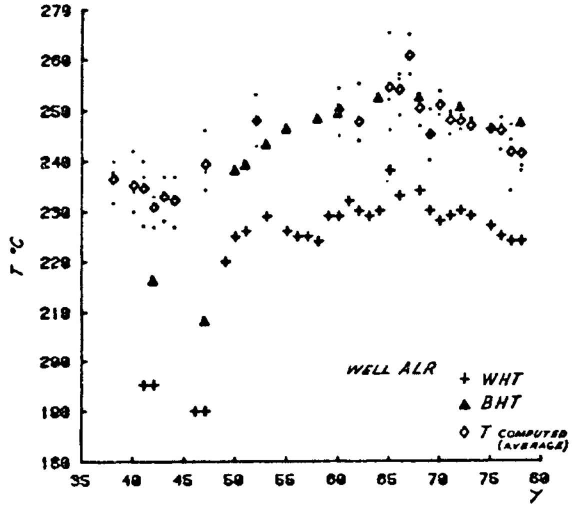

Figure 5

Figure 5. Evolution with time of well-head temperature (crosses), well-bottom temperature (triangles), and average reservoir temperature computed using H_2S, H_2, CH_4 geothermometers (diamonds) for well ALR.

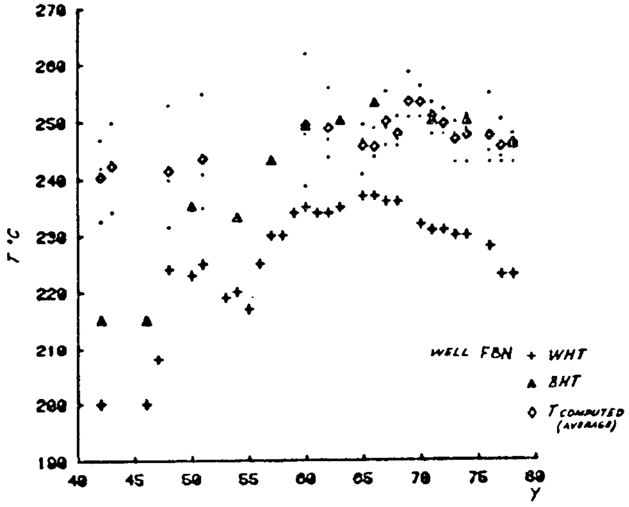

Figure 6

Figure 6. Same as Figure 5, for well FBN.

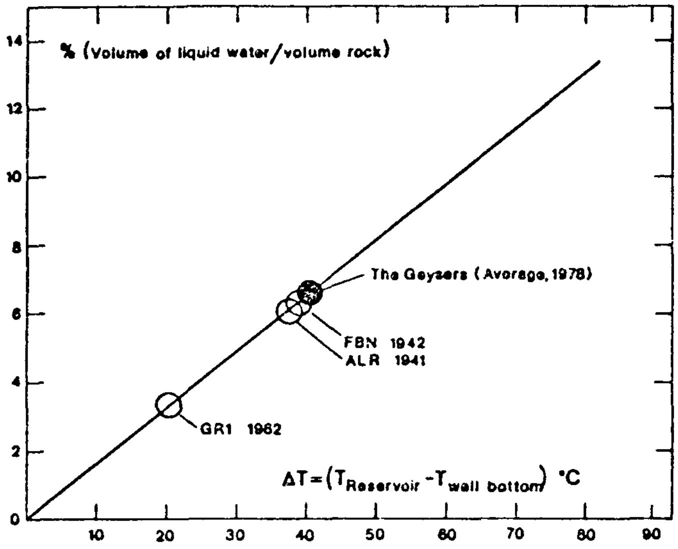

Figure 7

Figure 7. Computed volume per cent of liquid water in total rock plotted vs. the difference between the reservoir and the well-bottom temperatures. Starting production conditions are located on the diagram for 3 wells at Larderello.

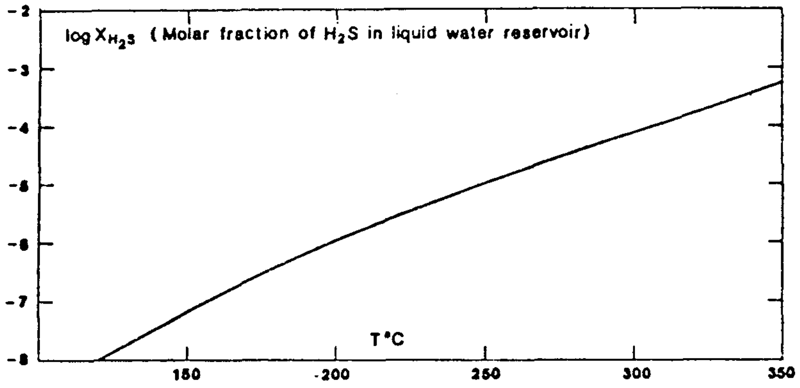

Figure 8

Figure 8. Computed molar fraction of H_2S in liquid water reservoir as function of temperature used as geothermometer for water-dominated geothermal systems (Table 6).