Estimation of Reservoir Temperature of Geothermal Water in the Busan Area: Application of Mineral Equilibrium Temperature

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1. Introduction

The temperature of a geothermal reservoir can be estimated by using chemistry of water sampled from the geothermal area. A reservoir temperature experienced from geothermal waters can be recorded by ionic and stable isotope rations in solutes and the water itself. The depth of the geothermal reservoir also can be estimated from calculated reservoir temperature, based on an understanding of regional tectonics and geothermal gradients. Such informations are important to interpret origin and evolution of geothermal water.

The alkali element geothermometer developed over the past two decades rely on the assumption that the two species or compounds have coexisted and equilibrated within the geothermal reservoir. The temperature is the main control on their ratio, and re-equilibration has not occurred during ascent and discharge (Fournier and Truesdell, 1973; Truesdell, 1976; Tonani, 1980; Fournier, 1981; Fouillac and Michard, 1981; Kharaka et al., 1982; Arnorsson, 1983; Nieva and Nieva, 1987; Giggenbach, 1988; Kharaka and Mariner, 1989). The principal drawback to these alkali and silica geothermometers is that geochemical re-equilibration of geothermal waters can occur through exchange reactions such as mixing and dilution with groundwater at lower temperatures during ascent. Therefore, careful considerations are required in application of these geothermometers. Although they have problems, the alkali and silica geothermometers are useful tools to interpretate the temperature of geothermal reservoir and the evolution of geothermal water.

2. Calculation method of mineral equilibrium temperature

Using the activities of aqueous component species calculated for homogeneous equilibrium at a series of temperatures, it is possible to compute the degree of super- or undersaturation of the aqueous phase with minerals at each temperature (Reed and Spycher, 1984). Computed multicomponent chemical equilibria in the geothermal waters

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can be applied to chemical geothermometry. The method relies on using the composition of a natural water to find a temperature where a convergence of plausible alteration minerals in the geothermal system are computed to be in equilibrium with each other and with the aqueous phase. The study of mineral equilibrium temperature in many geothermal and hot spring waters has shown that most waters closely approach equilibrium with a subsurface mineral assemblage, and that the calculated mineral equilibration temperatures are within 10° to 20°C of the measured temperatures or those determined from conventional alkali ion and silica geothermometer (Reed and Spycher, 1984; Pang and Reed, 1998; Palandri and Reed, 2001).

In this study, for a given analysis of geothermal waters, mineral saturation indices (log Q/K = Log (AP/K), where Q and AP are the activity product, and K is the mineral-aqueous equilibrium constant) for probable secondary equilibrium minerals are calculated at many temperatures between 25° and 300°C. Equilibrium between the fluid and selected minerals is evaluated by plotting the saturation indices of the minerals vs. temperature (log Q/K graph). Agreement in the temperature at which log Q/K = 0 for the selected minerals is a good indiction of equilibrium between the minerals and geothermal water at the temperature where the curves converge. Most raw water analyses are computed to be at disequilibrium with possible secondary minerals, which results from problems with the analytical data for pH and concentration of silica, aluminum, bicarbonate, and acetate. These are recomputed during the calculations to identify equilibrium between the fluid and secondary minerals, if such equilibrium exists.

3. Application on geothermal waters in the Busan area

Various chemical geothermometers have been applied to predict the reservoir temperatures of geothermal water in the Busan area (Han et al., 1999). Application of the technique to various geothermal waters has shown that it yields useful information on the equilibrium status of the systems, and in many cases it provides an excellent prediction of equilibrium or reservoir temperatures (Tole et al., 1993). The reservoir temperatures calculated from available silica and alkali ion geothermometers are summarized (Table 1). Calculated temperatures from alkali ion geothermometers have various ranges. Notably, the temperatures calculated by using Li and Mg are higher than the temperatures calculated other ions, whereas the temperatures using Na and Li are lower. The temperatures calculated by quartz and chalcedony geothermometer range from 95.3 to 118.9°C and from 64.9 to 90.4°C, respectively. These temperatures are lower than some temperatures calculated by alkali ion geothermometers. These result, showing various temperatures, indicate that the chemical composition of geothermal waters in the Busan area is affected by different environments such as mixing and dilution during ascent.

Kim and Choi (1998) interpreted the high concentration of Na in geothermal waters originates from seawater mixing. If Na from seawater intruded into a geothermal water during ascent, the reservoir temperature will be over-calculated by the alkali ion

geothermometer using Na. In order to prevent such an error, we need to carefully elucidate geochemical environment of any geothermal system before applying alkali ion geothermometers.

Temperatures of multicomponent equilibrium using chemical composition each of the Haeundae (HW-01) and Dongrae (DW-01) geothermal water are estimated by using SOLVEQ (Reed, 1982). In cases where the Na-K-Ca and related methods fail to yield correct temperatures, the mineral equilibration method often still yields good temperature estimates because it is independent of assumptions of any particular mineral equilibria (Reed and Spycher, 1984). A plot of log Q/K vs. temperature for HW-01 is shown in Fig. 1a, from which this water equilibrated with a subsurface mineral assemblage within a temperature range of 80 to 150°C. Calcite is strongly supersaturated with respect to field pH, alkalinity and chemical composition. Zhong and Mucci (1993), and Zhang and Dawe (1998) reported that calcite precipitation is quite rapid even for the time scale of laboratory experiments, and that calcite is therefore almost certainly not supersaturated in a fluid at reservoir conditions. Calcite supersaturation probably results from degassing of CO₂ (Pang and Reed, 1998). Degassing of CO₂ is accompanied by increase in pH and decrease in bicarbonate activity. In calculation of temperature for mineral equilibration, degassed CO2 is replaced by adding in analytical data until the fluid is at the heretofore unknown temperature where Q/K curves for silicates converge with calcite to equilibrium (Fig. 1b). The calculation shows a knot of curve intersections suggesting a reservoir temperature of approximately 130°C. Also, in the same method, A plot of log Q/K vs. temperature for DW-01 is shown in Fig. 2a, calcite is strongly supersaturated. Degassed CO₂ is added in analytical data until silicates converge with calcite to equilibrium (Fig. 2b). The result of calculation shows that the knot of curve intersections suggest a reservoir temperature of approximately 110°C.

Table 1. Estimated reservoir temperatures of the thermal waters on the Busan area, using different gethermometers.

Sample no.	pН	Temp.	Estimated temperature (°C)													
			QC ⁱ⁾	QA ¹⁾	CA ^{I)}	NaK ¹⁾	NaK ²⁾	NaK ³⁾	NaK ⁴⁾	NaK ⁵⁾	NaK ⁶⁾	KMg ⁶⁾	NaKCa ⁷⁾	NaLi*)	NaLi ⁹⁾	LiMg ¹⁰⁾
Thermal water	from Ha	eundae					***************************************									
HW-01	7.3	55.1	115.2	114.1	86.4	143.7	102.6	110.9	132.1	99.8	163.0	111.3	90.8	30.1	103.9	191.7
HW-02	7.5	43.6	110.3	109.8	81.0	143.4	102.3	110.5	131.8	99.5	162.8	109.6	83.3	30.2	104.1	193.8
HW-03	7.8	40.0	118.9	117.3	90.4	137.0	94.7	103.5	125.6	92.2	156.6	107.7	88.4	-40.8	31.3	318.1
Thermal water	from Do	ngrae														
DW-01	7.9	68.1	103.6	104.1	73.9	133.5	90.6	99.6	122.2	88.3	153.2	116.1	74.7	41.2	114.7	167.3
DW-02	7.8	63.0	102.9	103.5	73.0	130.0	86.5	95.8	118.7	84.3	149.8	108.4	72.6	30.9	104.7	185.4
DW-03	7.8	46.2	95.3	96.9	64.9	126.8	82.8	92.3	115.6	80.8	146.8	95.3	66.1	55.9	128.5	179.0
DW-04	7.8	59.3	109.2	108.9	79.8	111.0	64.7	75.2	100.2	63.4	131.5	99.2	62.3	51.7	124.6	165.6
DW-05	7.7	52.9	99.9	100.9	69.8	137.5	95.3	104.0	126.1	92.8	157.1	106.3	74.1	37.3	110.9	187.2
DW-06	8.1	56.4	100.9	101.7	70.9	135.2	92.6	101.5	123.8	90.2	154.9	102.0	72.0	45.4	118.6	184.2
DW-07	7.2	29.1	98.8	100.0	68.7	105.8	58.9	69.6	95.1	57.7	126.4	67.7	39.5	47.9	121.0	218.5

QC (quartz, no steam loss), QA (quartz, steam loss) and CA (chalcedony) = silica geothermometers; others = alkali-ion geothermometers [Data sources:

1) Fournier (1981), 1) Tonani (1980), 1) Arnorsson (1983), 1) Nieva and Nieva (1987), 1) Truesdell (1976), 1) Giggenbach (1988), 1) Fournier and Truesdell (1973), 1) Truesdell (1976), 1) Truesdell (1976), 2) Giggenbach (1988), 2) Fournier and Truesdell (1976), 2) Truesdell (1976), 3) Giggenbach (1988), 3) Fournier and Truesdell (1976), 3) Giggenbach (1988), 3) Fournier and Truesdell (1976), 4) Giggenbach (1988), 3) Giggenbach (1988), 4) Fournier (1988)

²⁾ Fouillac & Michard (1981), ⁹⁾ Kharaka et al. (1982), ¹⁰⁾ Kharaka & Mariner (1989)]

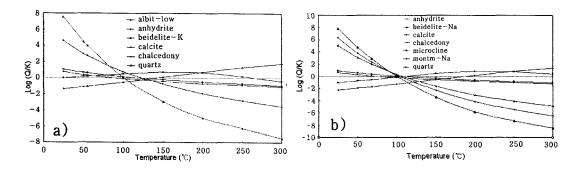


Fig. 1. Regular Q/K graphs for the Haeundae geothermal water. a) used raw water composition, strong supersaturation with calcite, b) sufficient CO_2 added to saturate the water with calcite

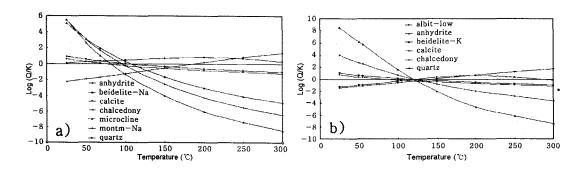


Fig. 2. Regular Q/K graphs for the Dongrae geothermal water. a) used raw water composition, strong supersaturation with calcite, b) sufficient CO₂ added to saturate the water with calcite

calcite, b) sufficient CO2 added to saturate the water with calcite Log (Q/K) Log (Q/K) 9used raw water composition, strong supersaturation with 6 2 0 2 00 9-10 4 ი **∞** Regular Q/K graphs for the Haeundae geothermal water. 50 50 100 100 - anhydrite - beidelite-K → albit-low chalcedony anhydritebeidelite-Na - microcline - calcite 150 Temperature (°C) → montm-Na - quartz - chalcedony calcite Temperature (°C) quartz 150 200 200 250 250 300 Log (Q/K) Log (Q/K) -109 b) sufficient CO2 added to saturate the water with calcite used raw water composition, strong supersaturation with calcite, Fig. 2. Regular Q/K graphs for the Dongrae geothermal water. a) 10 6 00 50 50 - beidelite-Na - chalcedony - calcite - montm-Na microcline anhydrite quartz 100 100 - anhydrite - beidelite-K - chalcedony - albit-low - calcite Temperature (°C) Temperature (°C) - quartz 150 150 200 200 250 250 300

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