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ORIGINAL ARTICLE

Estimation of reservoir temperature using silica and cationic solutes geothermometers: a case study in the Tengchong geothermal area

Xiaobo Zhang · Qinghai Guo · Jiexiang Li · Mingliang Liu · Yanxin Wang · Yijun Yang

Abstract Reservoir temperature estimation is vitally important for assessing the exploitation potential of a geothermal field. In this study, the concentrations of major chemical constituents in geothermal water sampled from boiling and hot springs in the Tengchong hydrothermal area were measured, and quartz and cationic solutes geothermometers were used to calculate subsurface temperatures. Log (Q/K) diagrams and Na–K–Mg triangular diagrams were applied to evaluating the equilibrium status of geothermal water samples with regard to reservoir minerals, and results were used to select suitable geothermometers. The results show that samples RH01, RH03, RH04, RH05, and LL16 were in or very close to full equilibrium with the selected minerals, and therefore a Na–K geothermometer is appropriate. A K/Mg geothermometer, however, is applicable to LP08 and PZH18 whose chemical compositions adjusted to the shallow reservoir temperatures during their re-equilibrium processes. In contrast, cationic solute geothermometers are unsuitable for SQ20 and RH07, which are categorized as immature water in the Na–K–Mg diagram; a quartz geothermometer was adopted to evaluate the corresponding subsurface temperatures of these samples. According to the reservoir temperature estimation made in this study, there is at least one high-temperature reservoir below Rehai with a possible temperature range of 210–270 °C.

Keywords Chemical equilibrium · Geothermometer · Geothermal spring · Reservoir temperature · Tengchong

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1 Introduction

Hot springs are typical surface indicators of subsurface geothermal activity. During the exploration and subsequent evaluation of geothermal resources, one of the most important procedures is to estimate reservoir temperature based on the chemical composition of geothermal waters (Fournier and Rowe 1966; Asta et al. 2012; Fournier and Truesdell 1973; Pang and Reed 1998). As an essential parameter, reservoir temperature can be used to categorize a geothermal system and to assess its exploitation potential (Mutlu and Gulec 1998; Pang 2001). Geothermometry is an economical and effective way to measure this parameter (Pasvanoglu 2011). Among all kinds of geothermometers, silica and cationic solute geothermometers are the most widely used, but have been proven to be useful only in specific situations (Ellis and Wilson 1960; Ahmad et al. 2002; Anzil et al. 2012; Mimi et al. 1998; Pirlo 2004). For example, a lack of full equilibrium of geothermal water with all minerals in reservoir hostrocks—which may be caused by low reservoir temperature or different types of cooling, including adiabatic cooling, conductive cooling and mixing with cold waters—may render cation geothermometry unreliable (Dulanya et al. 2010; Mohammadi et al. 2010). Thus, for a given geothermal water sample, a wide range of subsurface temperatures can be obtained with the use of different geothermometers. It is necessary to evaluate geothermal fluid–hostrock equilibrium before selecting an appropriate geothermometer for reservoir temperature estimation (Zheng et al. 2002; Gokgoz and Tarcan 2006; Majumdar et al. 2009).

The Tengchong volcanic geothermal area in western Yunnan of China is characterized by numerous hot springs and very high terrestrial heat flow values, and has a huge potential for geothermal resource exploitation (Shangguan

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et al. 2005; Du et al. 2005). A wide variety in the pH, temperature, and chemical composition of the hot springs in the Tengchong geothermal area makes it a prime location to comprehensively investigate the applicability of various solute geothermometers. The primary objective of this paper is to evaluate the feasibility of estimating the reservoir temperatures of several representative hydrothermal systems located in Tengchong (e.g. Rehai, Xiaotang, Shiqiang, etc.) using silica and cationic solute geothermometers. Based on this evaluation, principles for the selection of suitable geothermometers are proposed as well. The calculation results of subsurface temperatures are also useful for assessing the geothermal resource in Tengchong.

2 Geological setting

Tengchong County, with a total area of 5,848 km^2, lies in the southwest of Yunnan province (Fig. 1). The elevation in the county ranges from 3,780 to 930 m. Tectonically, Tengchong is located in the collision belt between the Eurasian and Indian plates, where volcanism was frequent in the Late Cenozoic. Volcanic rocks, including Pliocene alkaline basalt and dolerite, Early Pleistocene calc-alkaline andesite and dacite, early Middle Pleistocene alkaline basalt, and late-Middle and Late Pleistocene calc-alkaline andesitic basalt are widely distributed in Tengchong (Shangguan 2000).

With an area of about 10 km^2, the Rehai geothermal field is located in the southern part of the Tengchong volcanic geothermal area. Rehai is the most important hydrothermal area in Tengchong in light of its high reservoir temperatures and strong geothermal manifestation including boiling springs, hot springs, geysers, hydrothermal explosions, and steaming ground (Zhang et al. 2008; Guo and Wang 2012; Shangguan et al. 2005). With increasing depth the following sequence of units is found at Rehai: Holocene alluvium, middle Pleistocene basalt, lower Pleistocene andesite, Miocene brecciated rocks, Cretaceous granite, and Proterozoic gneiss and migmatite. The caprocks of the Rehai hydrothermal system consist of intensively altered gravelstone and sandstone, while the reservoir itself is composed of the Yanshanian granite and Proterozoic metamorphic rocks. According to the carbon isotope fractional values of CO_2 and CH_4 from magma degassing, the present temperature of the deep magmatic heat source should be above 514 °C. The discharge of geothermal waters from Rehai is controlled by three groups of faults at different depths. A multi-layer reservoir structure has been identified below Rehai (Shangguan et al. 2005).
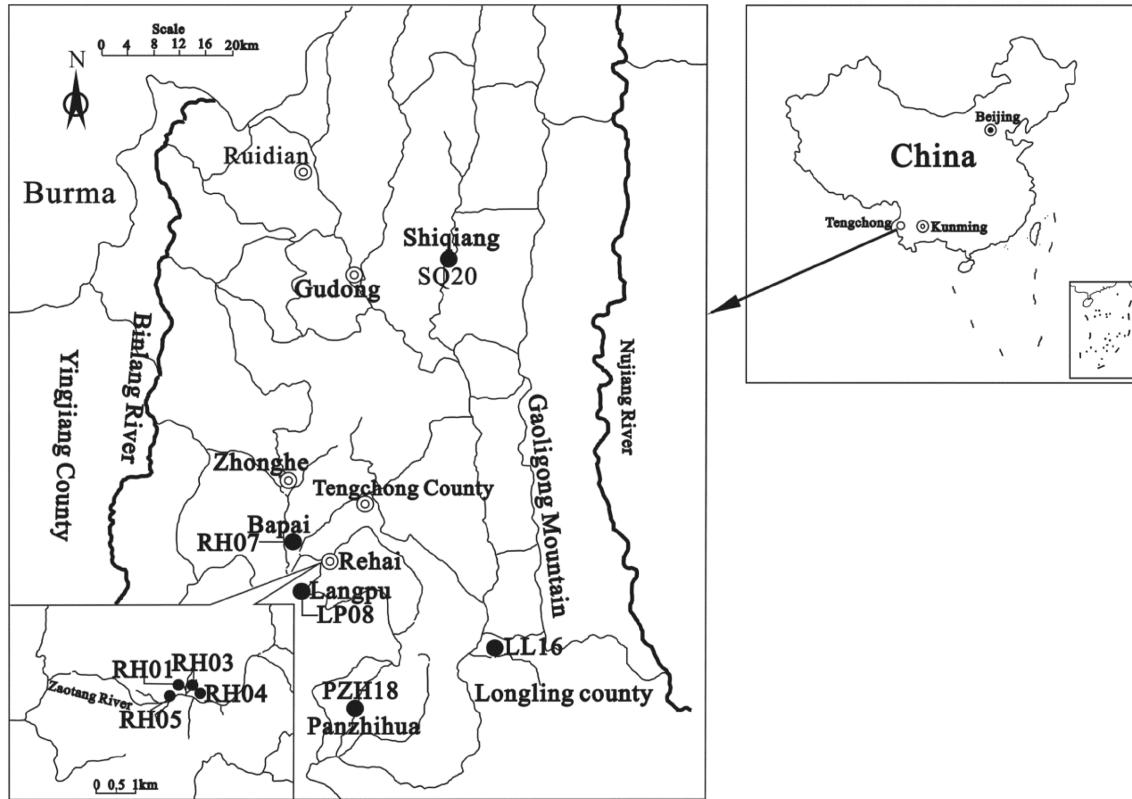
Fig. 1 Simplified map of the Tengchong geothermal area and sampling locations

3 Sampling and analysis

Nine geothermal water samples were collected in Tengchong in July 2012; sampling locations are shown in Fig. 1. The major characteristics and hydrochemical types of the nine samples are presented in Table 1 and Fig. 2. All the water samples were collected from the hot springs emerging across the Tengchong geothermal area and added to 500 mL polyethylene bottles which had been rinsed with distilled water three times before sampling.

For cation analysis, reagent-grade HNO_3 with a concentration up to 14 M was added to each sample to bring the pH below 1. Unstable hydrochemical parameters, including temperature, pH, and electrical conductivity (EC), were measured with hand-held meters that had been calibrated beforehand. Alkalinity was measured using titration within 24 h after sampling. The HCO_3^-, CO_3^{2-}, and CO_2 concentrations of all samples were calculated from the measured values of alkalinity and pH by PHREEQC. The SO_4^{2-}, Cl^-, and F^- concentrations were analyzed by IC; Ca^{2+}, Mg^{2+}, Na^+, and K^+ by ICP-OES; and the other metal elements by ICP-MS. The concentrations of major chemical components in the samples are listed in Table 2. The SiO_2 concentrations of the Tengchong samples were not determined in this study, and the measurements made by other authors were used (Shangguan et al. 2005).

4 Results and discussion

4.1 Geothermometry

Many dissolved constituents in geothermal water and their concentration ratios can be used as geothermometers to estimate the temperature of a reservoir. The most widely used geothermometric methods are silica and cationic solute geothermometers. The former is based on temperature-dependent variation in the solubility of silica (Pang et al.

1990), whereas the latter uses the fact that the equilibrium for exchange reactions of some cations is controlled by reservoir temperature. The expressions for quartz, Na–K, and K–Mg geothermometers are presented in Table 3 and were used to calculate the reservoir temperatures corresponding to the nine geothermal water samples collected in this study (Tong and Zhang 1989). The results are shown in Table 4 and also in Fig. 3.

It is not surprising that different results were obtained through the use of the four geothermometers. As mentioned earlier, whether or not a geothermometer is appropriate for reservoir temperature calculation depends on a series of factors. For the application of cationic solute geothermometers, the most important precondition is that the geothermal water should be in equilibrium with the relevant hydrothermal minerals (Mutlu 1998; Reed and Spycher 1984). Thus, an evaluation must be conducted to judge
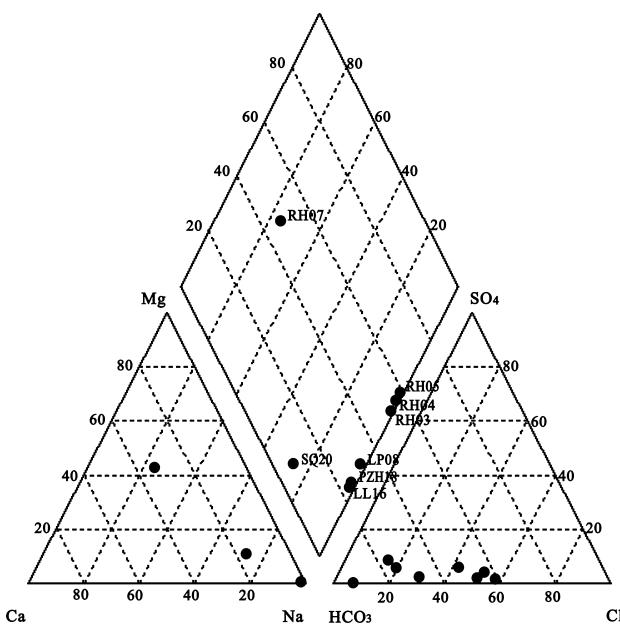
Fig. 2 Piper diagram of geothermal water samples

Table 1 Characteristics of water samples from Tengchong

No.	Location	Date	Sample type	Temperature(°C)	PH	Hydrochemical type	EC (μs/cm)
RH01	Dagunguo	2012.07.16	Hot spring	96.6	8.34	Na–HCO₃–Cl	4,210
RH03	Huaitaijing-L	2012.07.16	Hot spring	88.0	8.35	Na–HCO₃–Cl	3,350
RH04	Huaitaijing-R	2012.07.16	Hot spring	88.0	7.61	Na–HCO₃–Cl	2,464
RH05	Gumingquan	2012.07.16	Hot spring	96.0	8.87	Na–HCO₃–Cl	3,560
RH07	Bapai	2012.07.17	Hot spring	20.1	7.65	Na–Ca–HCO₃	163.6
LP08	Langpu Dagunguo	2012.07.17	Hot spring	70.0	8.16	Na–HCO₃–Cl	3,020
LL16	Banglazhang	2012.07.20	Hot spring	66.0	8.77	Na–HCO₃	1,021
PZH18	Xiaotang	2012.07.20	Hot spring	96.0	8.00	Na–HCO₃	1,078
SQ20	Shiqiang	2012.07.21	Hot spring	62.0	6.93	Na–Ca–HCO₃	2,456

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Table 2 Concentrations of major chemical constituents in water samples from Tengchong (mg/L)

No.	Alkalinity	SiO$_2$	SO$_4^{2-}$	Cl$^-$	F$^-$	NO$_3^-$	Ca	Mg	Na	K	Li
RH01	987.1	700	35.2	725.0	18.2	92.5	0.97	0.02	689.0	122.8	0.00
RH03	770.7	304	20.1	558.6	13.8	19.6	1.54	0.06	538.3	96.8	0.00
RH04	515.2	491	38.3	454.6	8.1	15.4	1.61	0.05	400.4	71.1	0.00
RH05	826.5	483	18.6	651.0	16.1	5.6	1.38	0.03	573.2	107.1	0.00
RH07	75.0	55	8.0	45.0	0.0	14.1	10.33	8.21	9.1	4.9	0.01
LP08	1217.9	150	28.2	336.9	11.7	5.1	3.84	0.98	573.8	55.9	2.89
LL16	505.2	172	38.9	54.5	19.3	0.0	1.36	0.01	203.5	15.2	3.03
PZH18	445.2	244	30.0	83.4	11.7	5.3	1.67	0.13	209.9	19.0	1.44
SQ20	1623.1	79	1.8	82.8	3.3	2.6	70.45	28.08	395.5	68.9	2.03

Table 3 Expressions for different geothermometers

Geothermometer	Expression	
Quartz(a)	$\theta_{\mathrm{SiO_2}}\,°\mathrm{C} = \frac{1309}{5.19-\log\mathrm{SiO_2}} - 273.15$	(1)
Quartz(b)	$\theta_{\mathrm{SiO_2}}\,°\mathrm{C} = \frac{1522}{5.75-\log\mathrm{SiO_2}} - 273.15$	(2)
Na/K	$\theta_{\mathrm{Na}/K}\,°\mathrm{C} = \frac{1217}{\log(Na/\mathrm{K})+1.483} - 273.15$	(3)
K^2/Mg	$\theta_{K^2/Mg}\,°\mathrm{C} = \frac{4410}{14-\log\left(K^2/Mg\right)} - 273.15$	(4)

if a geothermometer is suitable for estimating reservoir temperature.

5 Evaluation of water–rock chemical equilibrium

5.1 Log (Q/K) diagrams

Reed and Spycher (1984) proposed a method to assess the state of chemical equilibrium between geothermal water and minerals in reservoir hostrocks. The principle is that the dissolved states of most minerals in water can be regarded as a function of temperature. If a geothermal water sample is close to saturation with regard to a group of minerals at a specific temperature, it is believed that this water sample was in equilibrium with hostrock minerals

and this temperature is the most probable temperature of the reservoir. A geothermal water that has never been in full equilibrium with hostrock minerals or has experienced a cooling process that was not followed by a complete re-equilibrium cannot be saturated with all minerals at any specific temperature (Zheng and Liu 1996).

To evaluate the status of a geothermal water sample with regard to hostrock minerals in the reservoir, we introduce the concept of saturation index (SI):

$$\mathrm{SI} = \log\frac{Q}{K} = \log Q - \log K$$

where Q and K represent the ion activity product in solution and the solubility constant of a mineral, respectively. Based on the measured hydrochemical composition, the variation of a geothermal water sample's SIs with respect to K-feldspar, chrysotile, quartz, chalcedony, calcite and aragonite can be delineated in a Log (Q/K) versus temperature diagram. Due to the reasons mentioned above, a water sample may not fully equilibrate with all hostrock minerals at a given temperature. However, if the SI trendlines for most minerals intersect near the horizontal axis, the sample can be regarded to be in full equilibrium with these minerals within the corresponding temperature range. Among the samples used in this study, RH01, RH04, and PZH18 are nearly in equilibrium with all the evaluated

Table 4 Reservoir temperatures calculated using different geothermometers

No.	Location	Temperature/°C				Estimated temperature (°C)
		Quartz(a)	Quartz(b)	Na–K	K–Mg	
RH01	Dagunguo	285.1	250.8	272.1	264.4	250 ∼ 270
RH03	Huaitaijing-L	210.4	192.7	273	227.4	210 ∼ 270
RH04	Huaitaijing-R	250.7	224.4	271.6	217.4	220 ∼ 270
RH05	Gumingquan	249.2	223.3	277.2	248.9	250 ∼ 277
RH07	Bapai	106.3	106.4	422.8	52.9	80 ∼ 100
LP08	Langpu Dagunguo	161.2	152.7	214.8	147.1	150 ∼ 210
LL16	Banglazhang	170.2	160.2	193.3	176.9	160 ∼ 190
PZH18	Xiaotang	193.9	179.5	208.6	144	180 ∼ 200
SQ20	Shiqiang	124.9	122.3	269.7	101.5	100 ∼ 120

**Fig. 3** Reservoir temperatures calculated using different geothermometers
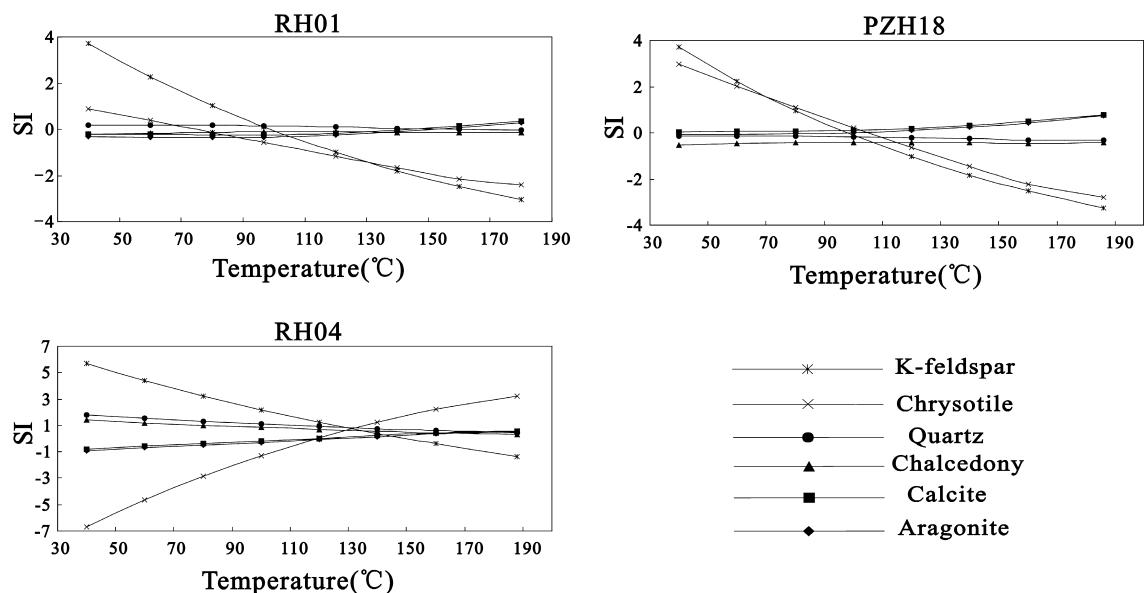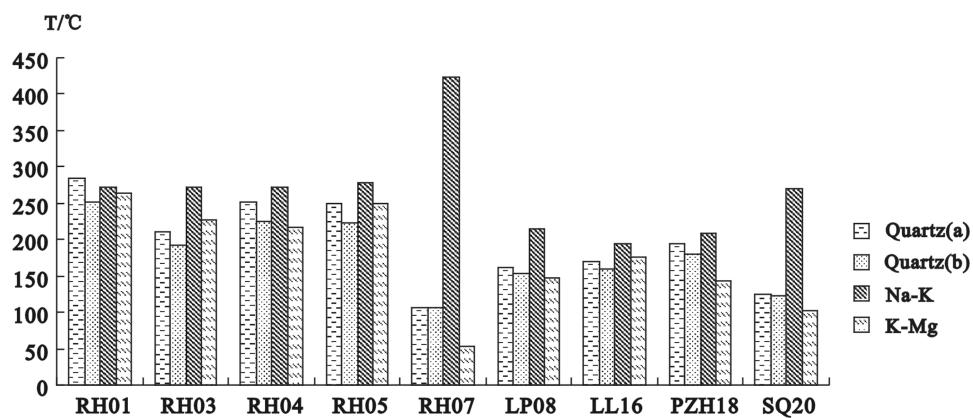
Fig. 4 Log (Q/K)—temperature diagrams for geothermal water samples in equilibrium with selected minerals. The lines show saturation index (SI) variation of the water samples with regard to K-feldspar, chrysotile, quartz, chalcedony, calcite, and aragonite with temperature. An SI greater or less than zero implies that it is oversaturated or undersaturated, respectively, with respect to the involved mineral

minerals (Fig. 4), while the disequilibrium of other samples is clearly documented by Fig. 5. Figure 4 shows that the equilibrium temperatures of samples RH01, RH04, and PZH18 are around 100, 130, and 108 °C, respectively. However, the Log (Q/K) diagrams do not show the real equilibrium temperatures. Hence, this method is possibly not suitable for the high-temperature geothermal waters in Tengchong, which leads us to seek other means to judge whether the Tengchong samples are in equilibrium with reservoir minerals.

6 Na–K–Mg triangular diagram

The Na–K–Mg triangular diagram, proposed by Giggenbach, is widely used to evaluate the state of water–rock equilibrium and to distinguish different types of geothermal water (Sun and Wu 1999; Giggenbach 1988). The diagram can be divided into three regions which represent fully equilibrated water, partially equilibrated water, and immature water, respectively. Its application is based on the following three temperature-dependent reactions:

$$K - feldspar + Na^+ = Na - feldspar + K^+$$

$$0.8\,muscotite + 0.2\,clinochlore + 5.4\,silica + 2Na^+ \\ = 2\,albite + 0.8\,K - feldspar + 1.6\,water + Mg^{2+}$$

$$0.8\,muscotite + 0.2\,clinochlore + 5.4\,silica + 2K^+ \\ = 2\,albite + 2.8\,K - feldspar + 1.6\,water + Mg^{2+}$$
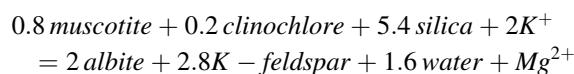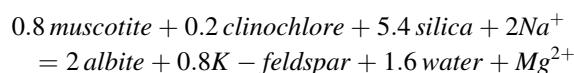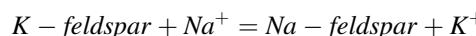
The advantage of this method is that it can be used to plot a large number of water samples on the same diagram
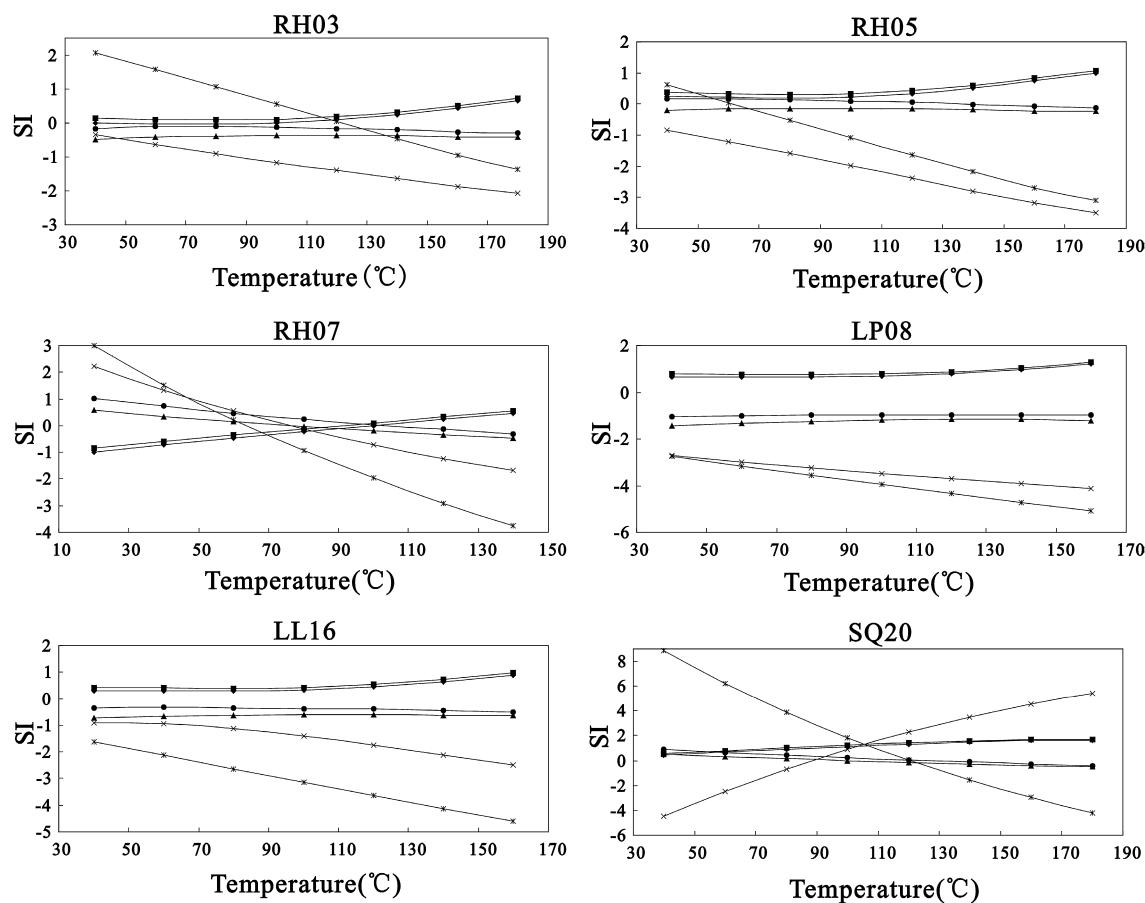
Fig. 5 Log (Q/K)—temperature diagrams for geothermal water samples in disequilibrium with selected minerals. The legend is the same as in Fig. 4
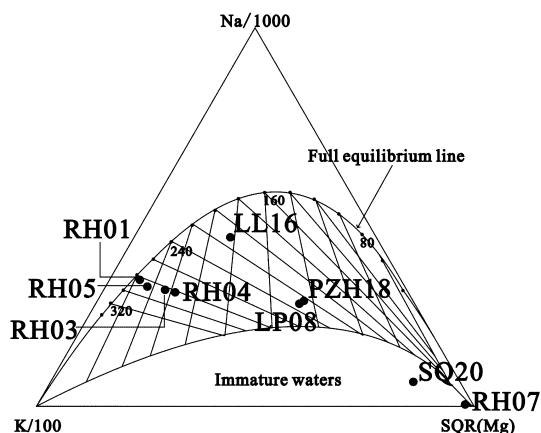
Fig. 6 Triangular Na–K–Mg diagram for geothermal water samples from Tengchong

and to distinguish immature waters from fully equilibrated waters and from those having experienced cooling and re-equilibrium.

As shown in Fig. 6, the samples RH01 and RH05 from Rehai are very close to the full equilibrium line, whereas the rest are located in the partial equilibrium area and the non-

equilibrium area. The samples that are not in full equilibrium may have undergone different forms of cooling or never been in full equilibrium with hostrocks as discussed above. Thus, cationic solute geothermometers cannot be used to estimate their corresponding reservoir temperatures.

7 Selection of geothermometers for reservoir temperature estimation

The quartz geothermometer can be expressed in two different forms (Crerar and Anderson 1971; Tong and Zhang 1989). One of them is suitable for geothermal waters that have no vapor loss during upflow to the surface; the other is applicable to the reverse situation. Put another way, distinct quartz geothermometers are designed for geothermal waters undergoing conductive cooling and adiabatic cooling. In this study, quartz geothermometer (b) was adopted to calculate the reservoir temperatures of samples RH01, RH05, and LP08 collected from boiling springs, while the other samples were subjected to quartz geothermometer (a).

RH01 is almost in full equilibrium with hostrock minerals in the reservoir, as indicated by its very close Na–K

and K–Mg temperatures. In contrast, RH03, RH04, RH05, and LL16 are located in the partial equilibrium area but very close to the full equilibrium line, revealing these samples have experienced one or more cooling processes, but their chemical compositions adjusted only slightly to new environmental temperatures. Thus, the Na–K geothermometer, re-equilibrating very slowly upon cooling, is suitable for these four samples. In contrast, the samples LP08 and PZH18 also plotted in the partially equilibrated area but far from the full equilibrium line, implying the geothermal water temperatures changed to a much greater extent during their re-equilibrium processes. Correspondingly, the differences between Na–K temperature and K–Mg temperature become larger. For these two samples, the K–Mg geothermometer is the more suitable one since it re-equilibrates much more quickly. Moreover, for some of the above samples, the reservoir temperatures estimated by quartz geothermometer are much lower than those by cationic solute geothermometers, which suggests the silica concentrations in these geothermal waters were controlled by other minerals such as chalcedony and amorphous SiO_2 instead of quartz.

In the Na–K–Mg diagram, there are two samples, RH07 and SQ20, located within the immature water area, wherein cationic solute geothermometers (both Na–K and K–Mg) are inapplicable. For example, the Na–K and K–Mg temperatures for the sample RH07 are 423 and 53 °C, respectively. Hence, for these two samples, the subsurface temperatures calculated using quartz geothermometer should be the closest to the actual values in the reservoirs.

Based on the above analyses, the reservoir temperatures corresponding to all samples are finally determined and given in Table 4.

8 Conclusions

The purpose of this study is to estimate the subsurface temperatures of several representative hydrothermal systems located in the Tengchong geothermal area using suitable geothermometers. The conclusions are as follows:

(1) If a geothermal water sample were in full equilibrium with hostrock minerals, its Na–K and K–Mg temperatures would theoretically be the same. For the geothermal waters very close to equilibrium with hostrocks, such as RH03, RH04, and LL16, the Na–K geothermometer is still the most applicable one for reservoir temperature estimation due to its slow re-equilibrium to lower temperature after a cooling process. In contrast, the K–Mg temperatures are more accurate for the samples LP08 and PZH18,

since they re-equilibrate their chemical compositions to a higher extent in shallow reservoirs.

(2) The quartz geothermometer in two different forms, suitable for geothermal water with and without vapor loss, are generally applicable to boiling springs having undergone an adiabatic process and to those springs with lower temperatures that have experienced conductive cooling.

(3) The samples RH07 and SQ20 are immature waters and cationic solute geothermometers are inapplicable to them. However, a quartz geothermometer can be used to calculate their corresponding subsurface temperatures.

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