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Reconstruction of deep fluid chemical constituents for estimation of geothermal reservoir temperature using chemical geothermometers

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Abstract: This paper elaborates the chemical constituent change principles of deep geothermal fluid during the process of upward movement. It summarizes research methods of hydrochemistry, isotope and numerical modelling technique for the physiochemical processes such as decreasing temperature, shallow groundwater infusion, and degassing. The multi-component chemical geothermometry methods including gas geochemical method are discussed. High-temperature geothermal fields in China are mostly located in the southwest with frequent new tectonic movements, especially in Tibet high-temperature geothermal areas. Therefore the paper also focuses the status of high-temperature geothermal fluid research. At last, it's pointed out in the paper that in the future we can start from typical high-temperature geothermal zones and geothermal fields to explore optimization of the multi-component geothermometry method and use it in the reconstruction and analogue of the formation mechanism and internal relevancy of regional geothermal systems.

Keywords: High temperature geothermal fluid; Multi-component geothermometer; optimization; Geothermal gases; Analogue

Introduction

As an important clean energy, geothermal energy has been included in the China National "13th Five-Year Plan" development plan (2015). Exploiting geothermal resources and identifing the reserve and distribution of the resources are the primary task of today's development of geothermal energy. According to engineering practices, drilling is the most direct and effective way to exploit geothermal resources. However, due to its high cost and risks, we have to leverage indirect ways to analyze deep geothermal fluid features and geothermal reservoir temperature before geothermal resource evaluation and geothermal exploitation and drilling. For example, one can collect chemical information including thermal water, geothermal gases, ground water and sinter to reconstruct geothermal fluid chemical constituents (Reed M H and Spycher N F, 1984; ZHAO Qing-sheng, 1988; ZHAO Ping *et al.* 2002; PANG Zhong-he *et al.* 2013; Spycher N *et al.* 2014).

Reconstructing deep geothermal fluid constituents is of important significance to determining geothermal reservoir temperature based on the equilibriated temperature of chemical reaction, under the prerequisite that assuming temperature decrease during the process of upward movement of geothermal fluid has not caused constituent changes. However, this is not the reality. The upward movement of geothermal fluid goes together with temperature decrease, shallow water infusion and degassing, which will break the original chemical equilibrium, change fluid chemical constituents and lead to big uncertainties in temperature calculation by the chemical geothermometry. Therefore, if deep geothermal fluid chemical constituents can be restructured, it will be possible to precisely calculate the temperature of geothermal reservoir, and thus provide precise and important parameters for researches of geothermal resource formation, type

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*Corresponding author: E-mail: guilingw@163.com classification, resource potential evaluation and geothermal development and use conditions in the exploitation and utilization. Besides, deep geothermal fluid chemical constituents also carry rich information including geothermal fluid sources, reserve, formation and evolution, so this is of important significance to the research on the geothermal formation mechanism.

High-temperature geothermal fields in China are mostly located in areas with frequent new tectonic movements in the southwest, of which, high-temperature geothermal activities are most frequent in Tibet in particular. This paper will brief the status of geothermal research in Tibet based on the chemical geothermometry method.

The paper summarizes the application of hydrochemistry method, isotope technique and numerical modelling technique in the deep geothermal fluid chemical constituents, which is an economic and fast way frequently used in geothermal resource exploitation in order to induce more value for researches on the geothermal formation mechanism.

1 Traditional chemical geothermometry

Before drilling is used to reveal thermal reservoir stratum, fluid chemical geothermometer is used to determine the reservoir temperature, which is an indirect and yet effective way to inspect regional thermal reservoir and is widely used in geothermal resource exploitation (PANG Zhong-he *et al.* 2013). The mostly used geothermometry includes cation geothermometer (Fournier R O and Truesdell A H, 1973), silicon dioxide geothermometer (PANG Zhong-he, 1990; Fournier R O and Rowe J J, 1966), gas chemistry geothermometer (Giggenbach W F, 1991), isotope geothermometer (Lloyd R R, 1968), solute geothermometer (Pang Z H and Reed M H, 1998).

The application of fluid chemical geothermometry is based on relatively ideal assumptions. These geothermometers assume that chemical equilibrium of water-rock reaction is reached between fluids and minerals. For example, Na-K geothermometer assumes a chemical equilibrium between geothermal fluids and minerals albite and K-feldspar; K-Mg geothermometer is based on the chemical equilibrium between muscovite, clinochlore and K-feldspar (Giggenbach W F, 1991). Therefore, geothermometry method only works under specific conditions. When assumptions do not fit to actual conditions, these geothermometers will not work accurately. For example, in a system with low temperature, the ratio of K-Mg may be controlled by chlorite, which will make the calculated temperature invalid. While, in fact, geothermal fields are much more complicated than ideal conditions: The upward movement of geothermal fluids to the ground surface will undergo gas loss, mixture, dilution, which will distort their features of deep geochemistry, thus leading to geothermometers invalid.

XU Wan-cai (1992) used SI-T and SI-PH curves to estimate the infusion proportion of cold water during the upward movement of geothermal water. YAO Zu-jin and CHEN Zong-yu (1995) analyzed water-rock chemical reaction through water-rock reactor in the lab. ZHENG Xi-lai and GUO Jian-qing (1996) studied treatment on silicon dioxide geothermometer and related problems, and pointed out that the prerequisite of using geothermometer is to be clear about various physical and chemical reactions during the upward movement of geothermal water. Pang and Reed (1998) pointed out that under the condition of equlibrated multi-components and heterogeneous chemical equilibrium of the geothermal system, since minerals contain some chemical constituents, the equilibrium of aluminosilicate is mutually reliant. He also proposed the Fix-Al method, which has laid a solid foundation for deep fluid chemical equilibrium reconstruction. Palandri J and Reed M H (2001) put forward Si-enthalpy correction method and mixture method. ZHANG Zhan-shi *et al.* (2004) calculated the geothermal reservoir temperature of a set of hot springs in Jiangxi through the multi-component chemical geothermometry.

2 Multi-component geothermometry and reconstruction of chemical composition

Based on these researches, I start to study the reconstruction of geothermal fluid chemical equilibrium features in order to make up for the limitation of the above-mentioned geochemical applications. Michard G *et al.* (1981) put forward the multi-component chemical geothermometry method, which uses the complete water chemical composition analysis data to calculate saturation index (log（Q/K）) of possible minerals of thermal reservoir within a certain temperature scope (for example 25-300 ℃). Log (Q/K) curve of all minerals could indicate the temperature of the thermal reservoir when the value is close to 0 ℃, and temperature value of each mineral could be different. Within this temperature scope, if there is a temperature value that makes the saturation index of as many minerals as possible to be close to 0, then this temperature value is considered as the temperature of the reservoir (Fig. 1a, b). Compared with traditional chemical geothermometry, the multi-component geothermometry applies to any geothermal systems theoretically because it is based on thermodynamics principles and calculation of reservoir temperatures through all aqueous components and solid minerals. This method avoids the limitation of the traditional geothermometry that only uses limited number of mineral saturation index to calculate reservoir temperature. However, the multi-component geothermometry requires multiple iteration calculations with great computation burden. Arnorsson S *et al.* (1982), and Reed M H (1982) used computer technology to develop multi-component geothermometry numerical model to decrease uncertainties in thermal reservoir temperature estimation, which has been developed further later (Arnorsson S *et al.* 1983; Michard G and Roekens E, 1983; Reed M H and Spycher, 1984; Pang and Reed, 1998; Palandri and Reed, 2001; Spycher, 2014). CHEN Chong-cheng and HUANG Zhen-guang (1997) did experiments for the Zhanjiang geothermal fields: They used the traditional geothermometry and multi-component geothermometry separately to calculate deep thermal reservoir equilibrium temperature. After comparative study, they stated that multi-component geothermometry could avoid impact of certain mineral imbalance because it reaches saturation in equilibriated temperature, so the method is stable and objective. ZHAO Ping *et al.* (1998a) used the multi-component geothermometry to calculate the temperature of geothermal reservoir in the Yangbajing area of Tibet. LIU Ying-chao *et al.* (2015) leveraged the geochemical characteristics of geothermal water in Beijing to analyze water evolution during the hot water transport. LI Jie-xiang *et al.* (2015) identified the temperature of geothermal fluid in thermal fields of geothermal seas, geothermal fields and geothermal springs based on multi-component geothermometry models and analyzed different temperature decrease processes after the upward movement. However, before saturation index calculation, one should calibrate fluid composition changes due to dilution, mixture and degassing, and then reconstruct deep fluid chemical components (Spycher N *et al.* 2014).

Spycher N *et al.* (2014) made a set of objective standards to calculate temperature indicated by mineral saturation index. He also took into consideration the impact of non-equilibrium chemical reaction dynamics on temperature calculation of geothermometry. Objective methods and water composition analysis data with multiple water samples from the same sources were chosen, and a computing software GeoT^2 of multi-component geothermometry was developed (Reed M H and Spycher N F, 1984). The software is fully automatic, and adopts mathematical statistics to quantify estimated temperature and error, which makes great contribution to promoting the use of multi-component geothermometry.

Peiffer L *et al.* (2014) applied this method in the Dixie Valley geothermal system, treated several data sets and optimized numerical value of estimated temperature, and then proposed a new concept that this geothermal field probably is made up of two geothermal systems. He investigated the superiority of the multi-component geothermometry compared to the traditional geothermometry. Wanner C (2014a, b) built a concept model based on the Dixie Valley geothermal system and inspected the impact of the flow and heat transfer on the solute multi-component geothermometry calculation, and then put forward the mutual dependence of estimated temperature and the flow speed of geothermal fluid upward movement. Spycher N (2014) analyzed the impact of non-equilibrium situation on the temperature calculation based on lab experiment data and analog computation results. Peiffer L *et al.* (2014) pointed out that it’s easier for secondary mineral to reach chemical equilibrium with fluid based on analog computation. Therefore, before using the geothermometry, one must thoroughly study various geological, hydrogeological and geochemical information, analyze rock-water equilibrium in thermal reservoir and hot water-cold water mixing effects, namely temperature decrease, shallow water infusion and degassing during the upward movement of geothermal fluids. For high-temperature geothermal system partly or fully filled with geothermal gas, geothermal gas chemical geothermometry is an important method to study high-temperature geothermal system. If one can include gas chemical constituents into the multi-component chemical geothermometry program, then the result of the reservoir temperature calculation will be much more reliable.

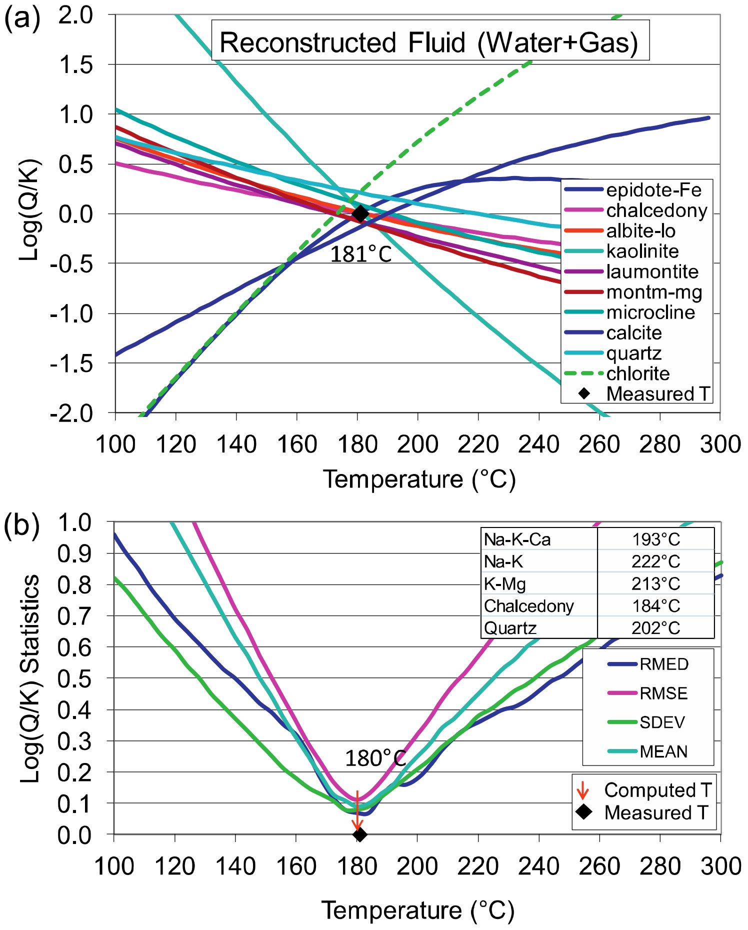

Fig. 1 Multi-component chemical geothermometry. Statistics information of (a) log(Q/K), (b) log(Q/K) computed by GeoT.

3 Example of chemical geothermometry method for a Tibet geothermal field

LIAO Zhi-jie (1982) pointed out that Tibet is one of the foremost Chinese geothermal regions, in which there are about 600 geothermal areas. The reconnaissance survey confirmed that an active geothermal belt, the Himalayan Geothermal Belt, extends about 2 000 km across southern Tibet along the India-Yalutzanbo Sture. DUO Ji (2003) studied the basic characteristics of the Yangbajing geothermal field. The deep thermal water is of sodium chlorite type, Carbon dioxide is a major non-condensate gas in the shallow and deep reservoir. According to the monograph “geothermal beneath Tibetan plateau” and the results of subsequent geothermal geological work, four geochemical geothermometers were used to calculate reservoir temperature. 131 geothermal spots whose reservoir is above 150 ℃ have been identified within Tibet, among which 8 geothermal spots’ reservoir is above 200 ℃.

AN Ke-shi *et al.* (1980) points out that the geothermal water in Yambajan geothermal field is blended with cold water as it moves upward, which leads to a relatively low estimation of the temperature of geothermal reservoirs. ZHAO Ping *et al.* (1998a), ZHU Li-xin (1989) and Shinichi Miyazaki *et al.* (2005) have carried out investigations on the hydro-geochemical features of deep-seated geothermal fluid in Yambajan, and calculated the temperature of geothermal reservoirs using chemical geothermometers, and made speculations about the sources of the fluid. ZHANG Meng *et al.* (2014) used hydro-geochemical methods to study the water-rock interaction of the high-temperature geothermal systems in Gulu Basin of Tibet. She pointed out that in some part of the ground, geothermal fluid and minerals have not reached the state of water-rock equilibrium. However, cationic geothermometers are not suitable for calculating the temperature of geothermal reservoirs.

CAI Zu-huang *et al.* (1985) pointed out that the piedmont fault of Nyenchen Tanglha Mountains only occurs inside the earth’s crust currently, and has not reached the mantle. No mantle substances have surfaced at the geosuture. ZHAO Ping *et al.* (2001) analyzed the sources of geothermal fluid in different reservoirs according to He isotopes in Yambajan geothermal gases, and perfected the formation mechanism of geothermal systems in Yambajan. HU Hong *et al.* (2003) pointed out that when temperature drops due to the slowing down of geothermal fluid movement or dilution of shallow underground water, equilibrium of chemical reactions will be broken, which gives birth to a new equilibrium. He also covers the applications of Na-K geothermometers under the new condition. Yokoyama T *et al.* (1999), Hoke L *et al.* (2000) have made comprehensive sampling and study on geothermal gases in the southern part of Tibet, indicating the apparent existence of mantle-derived helium. Arnórsson S *et al.* (2000) described the application of hydro-chemical and isotopic techniques during the exploration and development of geothermal resources. Some scholars have carried out investigations about hydro-geochemical processes in geothermal systems, sources of ions such as Cl, F, As and B, and solution equilibrium in high-temperature geothermal fluid (Armienta M A *et al.* 2014; Grassi S *et al.* 2014). LV Yuan-yuan *et al.* (2014) studied the sources and features of geothermal fluid with boron isotopes, and described the forming of the crust sources of underground water in Yunnan and Tibet geothermal areas.

ZHAO Ping *et al.* (1998a, 1998b, 2001, 2002) did a study on the geochemistry of Yambajan gases, and pointed out that the major component is CO_2, accounting for 85% of the total contents. Other components include N_2, H_2S, H_2, CH_4 as well as the R/Ra value of helium isotopes. In his study, he described the different movement tracks and physico-chemical processes of the components and perfected the formation mechanism of the geothermal system in Yambajan. DUAN Chen-yang (2014) concluded the hydro-geochemical features of underground water in Bujiemu valley, and did an investigation on the sources of geothermal water and their movement tracks and discussed the forming mechanism of the geothermal system there. SUN Hong-li *et al* (2015) analyzed the water-rock interactions of deep-seated liquid in the twelve high-temperature geothermal fields in Tibet as well as the physio-chemical processes during the upflow of deep-seated high-temperature liquid.

Extensive research has been carried out on the atmospheric precipitation in Tibet and sources of water in geothermal fluid in Yambajan using the indication of deuterium and oxygen (YU Jin-sheng *et al.* 1980; WEI Ke-qin *et al.* 1983; ZHENG Xi-lai and GUO Jian-qing, 1983). The result shows that the major water sources in Yambajan area is atmospheric precipitation. TAN Hong-bing *et al.* (2014) estimated the ratio of different sources of deep-seated geothermal water quantitatively.

SHEN Li-cheng (2007) did respective investigations on CO_2 emission fluxes in Himalayas, the formation of geothermal fluid around the borders of Southern Tibet and the degassing processes of CO_2. SHEN Li-cheng *et al.* (2011) did a simulation on the partial pressure of CO_2 in the hot springs of Langjiu and Targejia geothermal fields, and did a quantitative evaluation on the degassing effect of thermal water that occurs as the water moves from the aquifers to the surface. FENG Jin-liang *et al.* (2014) made a systemic analysis on the Rare Earth Elements of sinters in some of the geothermal system in Yambajan, and Eastern Himalayan syntaxis, and pointed out that the REE components in geothermal water largely affect the REE components in sinters. According to their studies, the calcification in the geothermal water with CO_3^{2-} and HCO_3^- as the major components is abundant in heavy rare earth elements.

4 Discussions and conclusions

Above all, in terms of the study on deep fluid in high-temperature geothermal systems, we take methods concerning hydrochemistry, environmental isotopes and gas geochemistry to study the water sources and movement tracks of geothermal fluid, and thermal sources and formation mechanism of geothermal systems. The development of chemical geothermometer approaches is a process from simple components to complex components, from experience to theory, from qualitative analysis to quantitative estimation (PANG Zhong-he *et al.* 2013). Chemical thermodynamics theories, computer technologies and the development of experiments and database of mineral thermodynamics have important influence on the quantification of temperature of geothermal reservoirs, as well as the ratio changes of chemical components during the temperature drop, mixture with shallow groundwater and degassing that occur with the moving upward of deep-seated geothermal fluid. Previous findings have provided feasible approaches and have laid theoretical foundation for future research, for instance, the formation and evolution of geothermal fluid, forming mechanism of geothermal system and the database of mineral thermodynamics. However, further study on some field is still needed, including defects in the application of multi-component chemical geothermometer in high-temperature geothermal gases, reconstruction of geochemical components in deep-seated geothermal fluid and the transformation and evolution of geothermal fluid during its movement. From now on, we can start our research from typical high-temperature geothermal zones and geothermal fields, and explore an optimized approach of multi-component (containing geo-chemical components) chemical geothermometry. Such approach can be used in the reconstruction and comparative study of the chemical components of local and regional deep-seated geothermal fluid to explore the formation mechanism and internal relatedness of local and regional geothermal systems. It can provide guidance for future exploration of geothermal resources and will inspire further investigations on improving the formation mechanism of geothermal systems.

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