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Geochemical Reservoir Damage due to Fluid-Mineral Interactions Induced by Drilling Fluid: An Inevitable Problem during Geothermal Drilling

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ABSTRACT

The target area for geothermal drilling is usually characterized by faulted as well as fractured structure, where the original cracks and fractures are beneficial to be functioned as flow channels during geothermal energy extraction. However, due to low formation pressure of geothermal reservoir, it is common for drilling fluid and its filtrate to invade into the surroundings of geothermal wells, and a series of fluid-mineral interactions along cracks and fractures will be induced under the promotion of high temperature. The results of fluid-mineral interactions are the characteristics evolution of cracks and fractures, which leads to permeability change inside rock, that is, geochemical reservoir damage. In this paper, a simulated fluid-mineral interaction experiment lasting 7 days was carried out between 1wt% KCl solution with a pH value of 10 referred to the drilling fluid used in geothermal wells and rock recovered from Gonghe Basin in Qinghai province. Further, electronic balance, XRD and XRF were conducted to identify the minerals and their mass as well as content changes. Based on the results above, the laws of geochemical reactions were analyzed. Finally, some design fundamentals of geothermal drilling fluid were proposed based on the laws of geochemical reactions, and hopefully will promote the development of geothermal drilling worldwide.

1. INTRODUCTION

There are three stages for geothermal exploitation containing drilling, completion and production when various fluids applied for engineering purposes will invade into the surrounding formation through cracks and fractures, and the invasion of fluids is liable to occur because of the low formation pressure of geothermal reservoir, which leads to formation damage through geochemical reactions, Zhao et al (2019), Gislason and Richter (2008).

It is common to find direct evidence of geochemical reactions in a formation proven by the trace of new minerals including alteration minerals, precipitation minerals as well as recrystallization minerals, and geothermal reservoirs are no exception because high temperature has a promotion effect on geochemical reactions, Zhang et al (2018), Clay et al (2018), Liu and Wang (1984). Fluid-mineral interactions as a form of geochemical reaction and its influence on physical and chemical characteristics of geothermal reservoirs such as weight, morphology, minerals as well as permeability has been studied experimentally, numerically and in situ, Charles and Bayhurst (1983), Richards et al (1992), Qiao et al (2019), Chen et al (2020).

However, fluid-mineral interactions during production and their mechanisms are highlighted more specially and are less studied during drilling. Compared with production process, although the duration for geothermal drilling is relatively short, a significant impact of geochemical reaction would be caused due to the chemical additives in drilling fluids and high temperature, Zhao et al (2019). Therefore, physical and chemical characteristics change induced by fluid-mineral interactions between drilling fluids and rock minerals should be further investigated.

In this paper, according to literature research, 1wt% KCl solution with a pH value of 10 was prepared to be reactive fluid containing caustic alkali and a type of salt which could play an inhibitory role in hydration swelling, Steven and Zhang (1982), Baumgärtner et al (2000). The rock for fluid-mineral interactions came from a geothermal well in Gonghe Basin, Qinghai Province. Considering the relatively short duration of geothermal drilling, a 7-day simulation experiment under 220°C, Zhang et al (2018), 24 MPa and 0.05ml/min conditions was carried out. Electronic balance was used to measure the mass of rock cuttings, and XRD, XRF was performed to identify the minerals and their content as well as chemical changes in rock cuttings.

The laws of fluid-mineral interactions including types of geochemical reactions and their influence were analyzed based on the results above, and provided some design fundamentals of drilling fluids to achieve minimum geochemical reservoir damage resulted from fluid-mineral interactions. Hopefully the conclusions will lead to a better understanding of geochemical reservoir damage mechanism and a method to construct a low damage drilling fluid system, which will promote the development of efficient geothermal drilling worldwide.

2. SIMULATED FLUID-MINERAL INTERACTIONS

2.1 Materials

2.1.1 Rock

The Rock sample was recovered from a geothermal well in Gonghe Basin, Qinghai Province, which belongs to granite with a depth of 2350m, and was crushed into around 5-mesh cuttings (about 2mm particle size). After crushing, the cuttings were dried in a drying oven for 24 hours at a temperature of 105°C in order to eliminate moisture content, Zhu et al (2020). Finally, the mass of dried cuttings was measured by a high precision electronic balance with four decimal places.

2.1.2 Reactive Fluid

1wt% KCl solution with a pH value of 10 was prepared as reactive fluid based on geothermal drilling fluid used during geothermal drilling, Steven and Zhang (1982), Baumgärtner et al (2000). NaOH was added to adjust the pH value. There was 800ml of reactive fluid added into the hydraulic cylinder, which would be interacted with cuttings during simulated experiment.

Table 1: Experimental materials.

No.	Reagent	Functions	Content	Provider organization/company
1	Granite cuttings	Reactant	Minerals	China Geological Survey (CGS)
2	NaOH	pH buffer	⩾96.0% (AR)	Shanghai Aladdin Chemical Co., Ltd
3	KCl	Chemical and physical compatibility	⩾99.5% (AR)	Beijing Sinopharm Group Co., Ltd

2.2 Methods

2.2.1 XRD

X-ray diffraction (XRD) analyses of cuttings before and after reaction were performed at Beida Zhihui Microstructure Analysis and Testing Center on a Bruker D-8 Advance XRD system. The minerals and their mass percent inside cuttings were identified through XRD analyses.

2.2.2 XRF

Cuttings before and after reaction were tested using Bruker S2 PUMA X-ray fluorescence (XRF) spectrometer at Beida Zhihui Microstructure Analysis and Testing Center. XRF was conducted to quantitatively determine the chemical compound contents and the elements mass percent of chemical compounds.

2.3 Experiments

A simulated fluid-mineral interaction experiment lasting 7 days under high temperature (220°C), high pressure (24MPa) and seepage conditions (0.05ml/min) was carried out on the flow HTHP reaction equipment as is shown in figure 1.

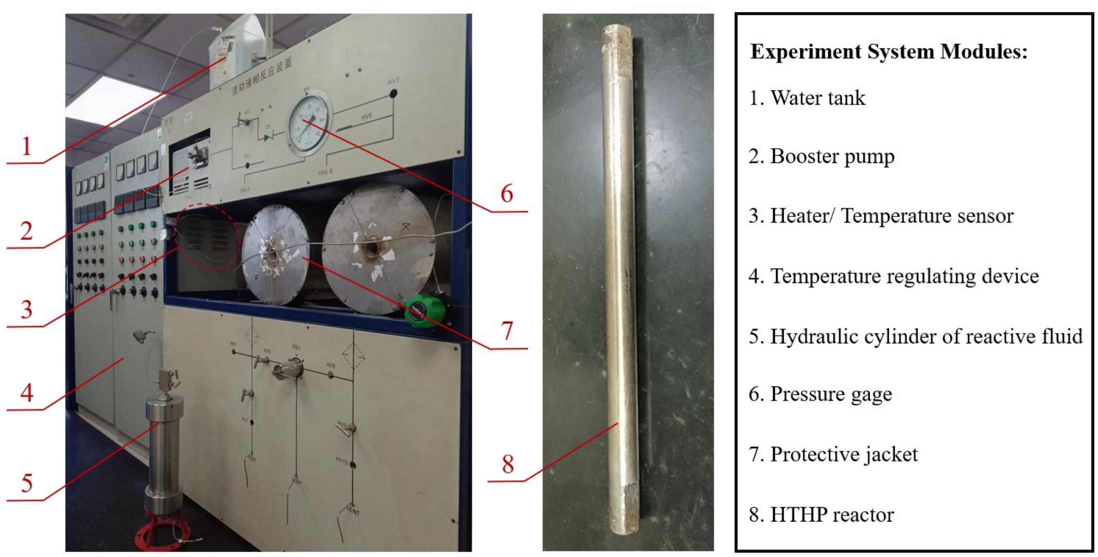

Figure 2: Image of flow HTHP reaction equipment and its main modules.

Cuttings was put into the HTHP reactor which would be located at empty column region in the center of the protective jacket, while reactive fluid was in upper part of the hydraulic cylinder. Distilled water was added to the water tank and was pumped into the bottom part of hydraulic cylinder, where the high pressure set by booster pump was transferred to reactive fluid. The heater and temperature sensor monitored by heating and temperature controlling system were placed in the interlayer between HTHP reactor and protective jacket, confirming a steady high temperature during fluid-mineral interactions.

After the pre-set temperature and pressure was reached, a simulated experiment continued for 7 days with a flow velocity of 0.05ml/min, and then the heating as well as pressurizing was stopped. Finally, cuttings after reaction were taken out under room temperature and atmospheric pressure, washed, and dried for 3 hours at a temperature of 130°C in order to remove free water and bound water.

2.4 Results

Rock before crushing as well as rock cuttings before and after reaction are shown in figure 3. Through lithological observation of the original drilled granite core, there are microcline in red meat, albite in white, quartz in translucent milky white as well as trace of biotite in black, and the rock belongs to biotite monzonitic granite. It is apparent that the cuttings after reaction are smoother than before.

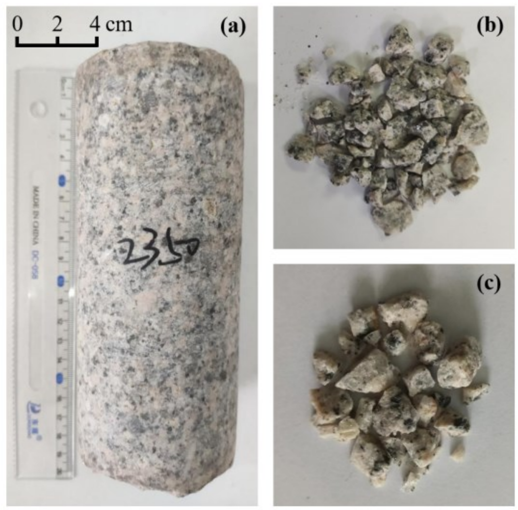

Figure 3: Pictures of rock cuttings. (a): Original drilled granite core. (b): Cuttings before reaction. (c): Cuttings after reaction.

After fluid-mineral interactions, mass of quartz decreases due to the dissolution of quartz. Three main types of feldspar including albite, microcline and anorthite are identified. In terms of feldspar, mass of albite has a large decrease while mass of microcline has a significant increase, and anorthite only dissolved a small quantity. Biotite is relatively stable without noticeable mass difference, and clinochlore the pre-existing clay mineral in cuttings has dissolved thoroughly. Under the experimental conditions, there are new clay minerals containing illite and kaolinite formed, indicating the alteration of minerals has occurred.

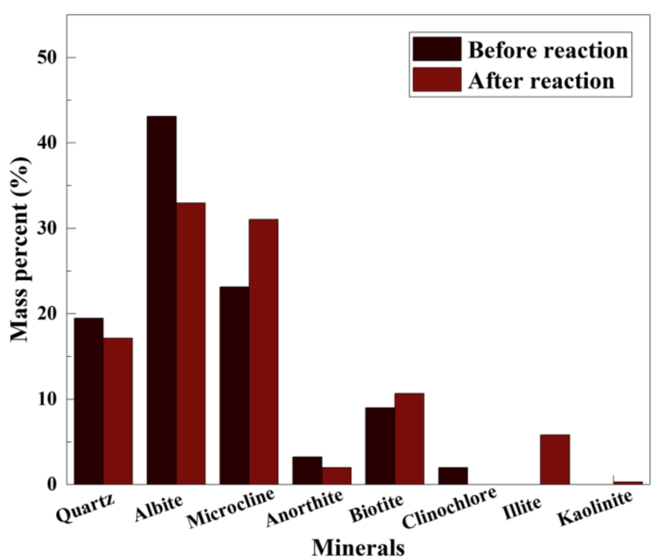

Figure 4: Minerals and their mass percent before and after reaction using XRD analysis.

Main chemical compounds and elements of minerals as well as their mass percent changes are presented in figure 6.

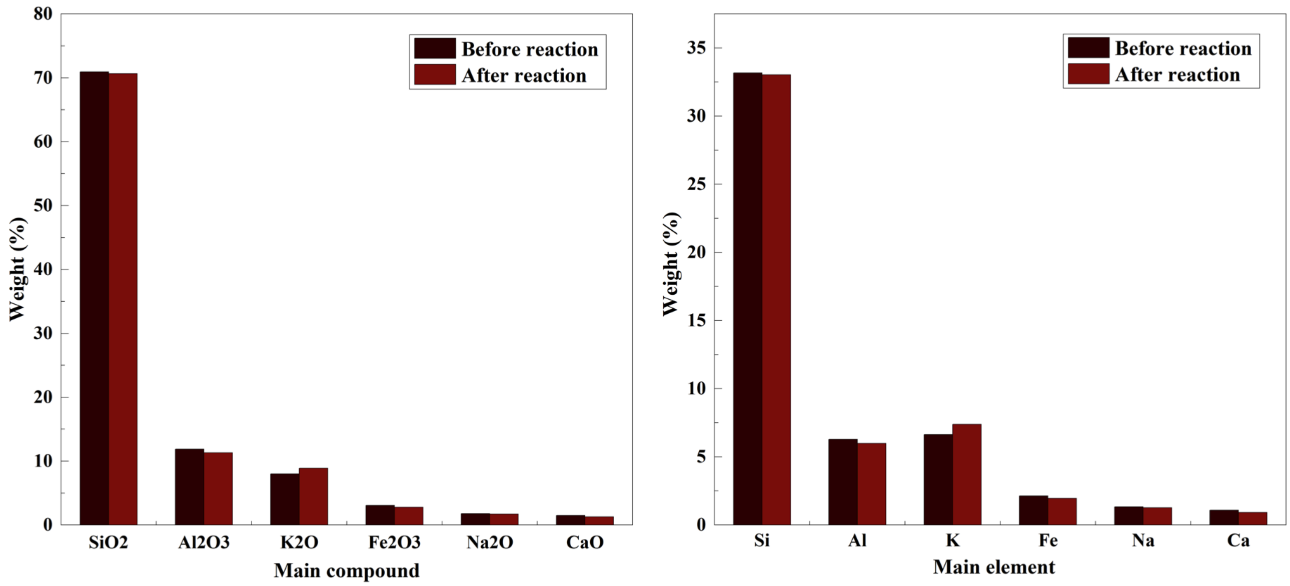

Figure 5: Main chemical content change of minerals before and after reaction using XRF analysis.

Silica and alumina are rich in granite cutting primarily consisting of aluminosilicate minerals and quartz, and hence silicon as well as aluminum are the main chemical elements. Biotite and clinochlore are iron-bearing minerals. After fluid-mineral interactions, aluminum and silicon have large decreases while iron, sodium and calcium have few decreases both due to the dissolution and alteration of minerals. Potassium basically occurs in microcline, biotite and illite, which is the main increased element in cuttings.

Table 6: Basic major elements of minerals.

No.	Minerals	Basic major elements (Expect hydrogen and oxygen)
1	Quartz	Si
2	Albite	Na, Si, Al
3	Microcline	K, Si, Al
4	Anorthite	Ca, Si, Al
5	Biotite	K, Fe, Mg, Si, Al
6	Clinochlore	K, Fe, Mg, Si, Al
7	Illite	K, Si, Al
8	Kaolinite	Si, Al

Mass of minerals or chemical compounds and elements is calculated by:

$$M = \text{wt}\% \times m_{\text{cuttings}} \quad (1)$$

where M, wt%, m_{cuttings} are mass and mass percent of minerals or chemical compounds and elements before or after reaction as well as mass of cuttings at corresponding reaction stage, respectively. wt% is obtained through XRD and XRF tests.

Combining the mass data of cuttings before and after fluid-mineral interactions weighed by electronic balance, the specific mass data containing mass change is given in table 7. Other major chemical elements with sufficient amount include Rb, Cl, Mn, Ti, Zr, Mg.

Table 7: Mass change of mineral chemical compounds as well as elements before and after reaction.

			Minerals and their mass change (g)								
	Reaction Temperature (℃)	Stage	cuttings mass (g)	Quartz	Albite	Microcline	Anorthite	Biotite	Clinochlore	Illite	Kaolinite
1wt%KCl + NaOH (pH=10)	220℃	Before reaction	39.7421	7.7457	17.1368	9.1963	1.2916	3.5768	0.7949	0.0000	0.0000
		After reaction	38.8559	6.6600	12.8108	12.0686	0.7771	4.1459	0.0000	2.2653	0.1282
		Change amount	-0.8862	-1.0857	-4.3260	+2.8723	-0.5145	+0.5691	-0.7949	+2.2653	+0.1282
			Main mineral compounds and elements as well as their mass change (g)								
				SiO_2	Al_2O_3	K_2O	Fe_2O_3		Na_2O	CaO	Others
		Before reaction	39.7421	28.2001	4.7214	3.1237	1.2161		0.7154	0.6041	1.1613
		After reaction	38.8559	27.4595	4.3985	3.5887	1.0841		0.6683	0.5012	1.1556
		Change amount	-0.8862	-0.7406	-0.3229	+0.4650	-0.1320		-0.0471	-0.1029	-0.0057
				Si	Al	K	Fe		Na	Ca	Others
		Before reaction	39.7421	13.1825	2.5000	2.5373	0.8505		0.5286	0.4332	19.7100
		After reaction	38.8559	12.8378	2.3275	2.9142	0.7577		0.4935	0.3590	19.1662
		Change amount	-0.8862	-0.3447	-0.1725	+0.3769	-0.0928		-0.0351	-0.0742	-0.5438

2.5 Discussions

2.5.1 Dominating Geochemical Mechanisms

The mass decrease of quartz is more than the mass decrease of silica, and the reason for that is because there are silica deposits after the release of silica due to quartz dissolution and alteration of feldspar.

For ions participating in geochemical processes, sodium and potassium cations are released from the active albite and microcline in cuttings under experimental conditions. The small ionic radius of sodium cations and proper ionic radius of potassium cations make them common interlayer cations, Zhao et al (2009). Under high temperature and high pressure, silicon in silica tetrahedron could be replaced by aluminum, and the negative charge caused leads to the introduction of cations between crystal layers. Therefore, sodium in cuttings has relatively little mass change after reaction despite albite has a drastic reduction.

In alkaline potassium chloride solution, ionization of potassium chloride provides potassium cations:

$$KCl \rightarrow K^{+} + Cl^{-} \tag{2}$$

In an environment of sufficient potassium cations and HTHP, processes including potassic feldspathization of albite and transformation of feldspar into illite are promoted, thus resulting in the significant formation of microcline and illite rich in potassium in cuttings.

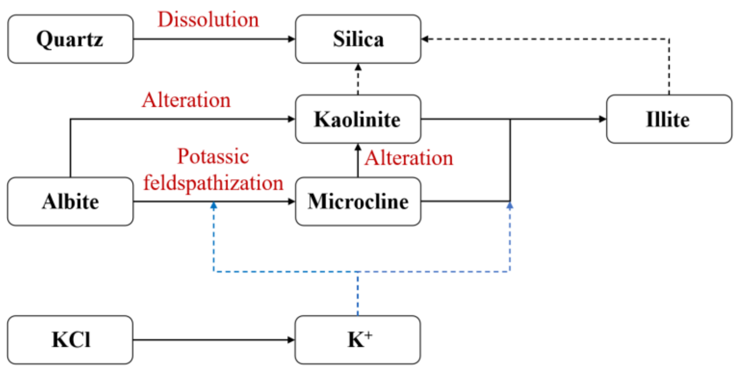

Figure 7: Schematic diagram of dominating geochemical mechanisms.

2.5.2 Main Geochemical Reaction Equations

Based on Huang et al (2009) and Qiao et al (2019), main geochemical reaction equations are given as follows.

$$NaAlSi_3O_8(\text{albite}) + K^{+} \rightarrow KAlSi_3O_8(\text{microcline}) + Na^{+} \tag{3}$$

$$2NaAlSi_3O_8(\text{albite}) + 2H^{+} + H_2O \rightarrow 2Na^{+} + 4SiO_2(\text{silica}) + Al_2Si_2O_5(OH)_4(\text{kaolinite}) \tag{4}$$

$$2KAlSi_3O_8(\text{microcline}) + 2H^{+} + H_2O \rightarrow 2K^{+} + 4SiO_2(\text{silica}) + Al_2Si_2O_5(OH)_4(\text{kaolinite}) \tag{5}$$

$$KAlSi_3O_8(\text{microcline}) + 4Al_2Si_2O_5(OH)_4(\text{kaolinite}) + 2K^{+} \rightarrow 3KAl_3Si_3O_{10}(OH)_2(\text{illite}) + 2SiO_2(\text{silica}) + 4H_2O + 2H^{+} \tag{6}$$

Based on Crundwell (2017), in terms of quartz dissolution in an alkaline environment, hydroxide attacks the silicon site ($\equiv SiO$) on quartz surface, and $SiO(OH)^{+}$ which later transforms into H_4SiO_4 as well as a vacancy site ($\equiv^{2-}$) which attracts cations are formed, also causing the introduction of sodium and potassium cations in cuttings:

$$\equiv SiO + 2OH^{-} + H_2O \rightarrow H_4SiO_4 + \equiv^{2-} \tag{7}$$

According to Rimstidt and Barnes (1980), silica released into fluid by other geochemical reactions goes through:

$$SiO_2 + 2H_2O \rightarrow H_4SiO_4 \tag{8}$$

And the precipitation of silica is described as:

$$H_4SiO_4 \rightarrow SiO_2 \rightarrow SiO_{2(s)}(\text{silica}) \tag{9}$$

Alkaline environment provides hydroxide and consumes H_4SiO_4, promoting quartz dissolution forward.

2.6 Expectations

Geothermal reservoir is characterized by high temperature, low formation pressure and fractured structure, Gislason and Richter (2008), Zhang et al (2018), and there are high possibilities for drilling fluid and its filtrate to invade into the surrounding formation, leading to reservoir damage problems. In this paper, geochemical reservoir damage is intently studied by figuring out the geochemical changes of minerals after fluid-mineral interactions, which further causes permeability change, thermal conductivity change and stress intensity factor change as is presented in figure 8, Yasuhara et al (2011), Yang et al (2010).

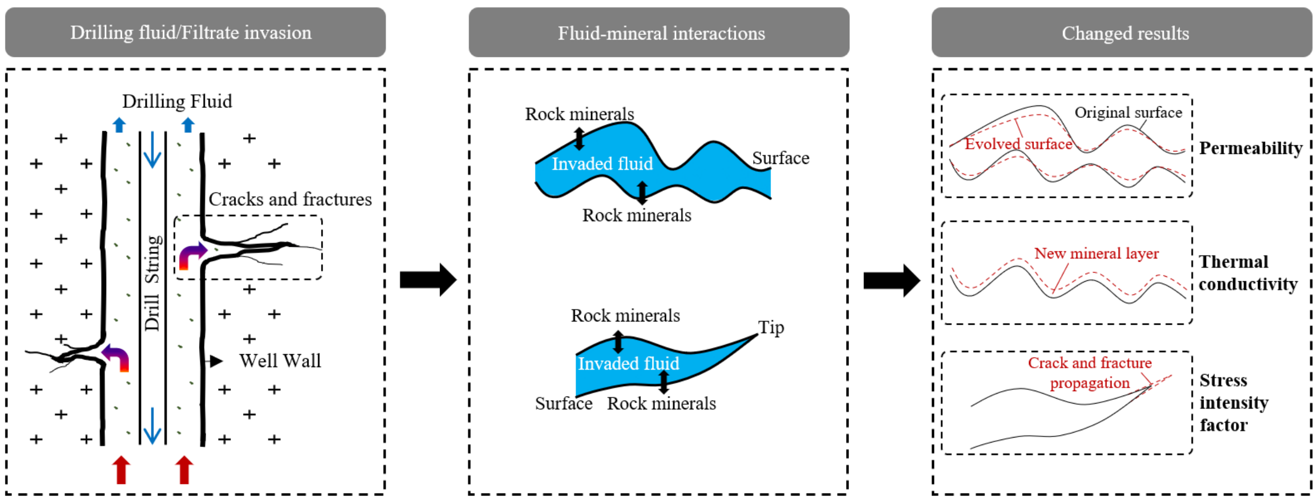

Figure 8: Schematic diagram of geochemical geothermal reservoir damage induced by drilling fluid.

In order to eliminate geochemical reservoir damage during geothermal drilling, it is important to understand the geochemical mechanisms of fluid-mineral interactions between drilling fluids and rock minerals. Based on conditions of geothermal reservoir, geochemical mechanisms can be artificially controlled by adjusting the chemical contents of drilling fluids.

2.6.1 Permeability Change

Permeability is a crucial factor of productivity of a geothermal well. Fluid-mineral interactions change minerals and their mass, and a direct expression is the evolution of crack and fracture surface, indicating permeability change, Yasuhara et al (2011). Dissolution of minerals is helpful of widening fracture aperture while precipitation causes permeability reduction. Besides, hydration swelling of clay minerals formed due to alteration leads to narrowing of cracks and fractures. As a result, dissolution should be promoted the same time hydration swelling of clay minerals and precipitation is inhibited.

2.6.2 Thermal Conductivity Change

The production efficiency of a geothermal well depends on thermal exchange between fluid and hot rock. Different minerals as well as their crystal axis represent different thermal conductivity, and hence the mineral change and crystal axis orientation change due to mineral precipitation and recrystallization induces thermal conductivity change inside cracks and fractures, indicating the change of production efficiency. According to Guo et al (2020), thermal conductivity of several minerals is listed in table 9.

Table 9: Thermal conductivity of diagenetic minerals.

No.	Minerals	Mean thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$)
1	Quartz	7.69
2	Albite	1.9~2.3
3	Microcline	2.4
4	Anorthite	1.7
5	Biotite	1.7~2.3 (2.09)
6	Clinochlore	5.17
7	Illite	1.9

8	Kaolinite	2.6

The higher thermal conductivity minerals have, the better thermal exchange ability of cracks and fractures is. In this paper, based on table 9, potassic feldspathization of albite transforming into microcline enhance thermal exchange inside cracks and fractures while formation of illite means the reduction of thermal conductivity. Therefore, it is capable to form new minerals with high thermal conductivity by controlling the geochemical mechanisms to improve thermal exchange ability.

2.6.3 Stress Intensity Factor Change

Partial cracks and fractures are more realistically existing in geothermal reservoirs. Propagation of cracks and fractures not only increases rock permeability by providing more seepage flow channels, but also improves thermal exchange ability because of a greater contact area between fluid and hot rock. Based on rock fracture mechanics, the initiation criterion of crack and fracture propagation depends on stress intensity factor. Fluid-mineral interactions change the stress intensity factor of cracks and fractures over tips, and the degree of change is affected by chemical contents including various types of fluids and minerals, Yang et al (2010).

3. CONCLUSIONS

In summary, the conclusions can be drawn as:

1) Under high temperature (220°C), high pressure (24MPa) and seepage (0.05L/min) conditions, geochemical processes including dissolution of quartz, precipitation of silica, potassic feldspathization of albite, alteration of feldspar and formation of illite from feldspar are promoted after fluid-mineral interactions lasting 7 days between 1wt% potassium chloride solution with a pH value of 10 and rock minerals in granite cutting recovered from Gonghe Basin, Qinghai Province.

2) In an environment of sufficient potassium cations, formation of potassium-rich minerals such as microcline and illite has occurred, and potassium cations in fluid enter into minerals as reactants and interlayer cations, increasing the potassium mass percent in cuttings.

3) Although albite has decreased significantly, sodium mass percent in cuttings is relatively stable with very little reduction, and the reason for that is because sodium cations enter minerals as interlayer cations due to their small ionic radius.

4) Understanding the geochemical mechanisms between drilling fluids and rock minerals under geothermal reservoir conditions is helpful of artificially eliminating geochemical reservoir damage from permeability, thermal conductivity as well as stress intensity factor three aspects.

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