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CHEMICAL CHARACTERISTICS AND THE FORMATION CONDITIONS OF GEOTHERMAL FLUIDS IN REYKIR AT REYKJABRAUT, N-ICELAND

Li Yuanyuan Tianjin Geothermal Exploration and Development Institute No.189 Weiguo Road Hedong District, 300250 Tianjin P.R. CHINA tj lvy@126.com

ABSTRACT

The Reykir low-temperature geothermal area at Reykjabraut is on the edge of the Vatnsdalur extinct central volcano. Eight deep geothermal wells have been drilled in the area, with a depth of 234 to 1676 m. Wells RR-05 and RR-12 are the major production wells. Based on the consideration of fluid and mineral compositions, geothermometers, mineral-solution equilibrium, mixing, isotopes, and water-rock interaction, this paper mainly describes the chemical characteristics of the thermal fluids, their origins, and the recharge and discharge of fluids. Finally, the process of water-rock interaction was modelled with PHREEQC. Some of the results were compared to the nearby Laugarbakki field in order to discuss different formation conditions of geothermal fluids.

1. INTRODUCTION

During every stage of geothermal exploration and utilisation, geochemical research plays an important role. The main goal of geochemistry in geothermal research is to predict subsurface temperatures, to identify the origins of geothermal fluids, and to quantify the processes that govern their compositions and the associated chemical and mineralogical transformations of the rocks with which the fluids interact. In geothermal exploration, during drilling and later assessment of the reservoir, geochemical research produces data on the chemical properties of the discharged fluid, and the steam to water ratio in the reservoir. The origin, recharge and transport research can determine where from and how the geothermal fluids originate, and which minerals mainly dominate the composition of the fluids. This contributes to the overall understanding of the production characteristics of geothermal reservoirs, necessary in order to determine the types of utilisation, such as space heating, power plants, fish farming, etc. It can quantify scaling and corrosion tendencies. After production has been initiated, geochemical monitoring is one of the most useful tools in recording the response of the reservoir to the production load, including recharge, pressure drawdown and possible enhanced boiling. It is necessary to establish geochemical laboratory facilities at geothermal application units in order to strengthen monitoring studies that contribute data pertinent to optimizing the economy of exploitation (Arnórsson, 2000).

The objective of this project was to investigate the formation conditions of the geothermal fluids in Reykir at Reykjabraut low-temperature geothermal area in North Iceland by analysing the chemical characteristics and comparing the findings with the different formation conditions of nearby Laugarbakki geothermal field. The geology of the geothermal areas is described briefly as well as the classification of the thermal waters. Possible changes of the chemical composition and properties of the thermal fluids with time are described. With the aid of geothermometers and the WATCH speciation program (Bjarnason, 2010), reservoir temperatures and equilibrium conditions were obtained. Further, mixing processes on geothermal fluids were evaluated for the two fields. On the basis of these investigations, the origin, recharge and transport of the geothermal waters are discussed. The geothermal waters at Reykir at Reykjabraut are influenced by water-rock interaction. Initial data of both the original water and minerals were obtained to model the process of water-rock interaction with the geochemical modelling program PHREEQC to determine the possible reactions and products in the reservoir. Finally, with the results of the modelling, the possible formation conditions are discussed.

2. GEOLOGICAL BACKGROUND

The geology of Iceland has been described e.g. by Saemundsson (1979); the following summary is based on his work. The north and northwest parts of Iceland are composed almost entirely of the socalled Tertiary basalt formation, characterised by high mountains, deep fjords and valleys. Geological studies show that most of the formation consists of a number of tholeiitic lava flows of various thicknesses interbedded with soil layers, several sedimentary horizons and a number of extinct volcanic systems. In the area close to Reykir at Reykjabraut (Figure 1), the regional dip of the lava pile is to the west and southwest whereas the dip is to the east at Laugarbakki geothermal field.

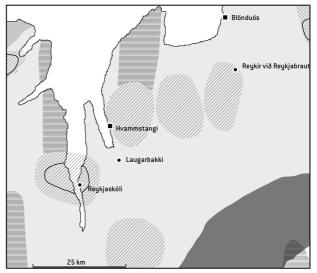


FIGURE 1: Location map of Reykir at Reykjabraut, and Laugarbakki area, NW-Iceland (from Orkustofnun, 2010)

The surface thermal activities in this part of Iceland are characterised as low-temperature

activity with temperatures ranging from 10 up to 100°C; it is believed that water penetration and circulation to depths of \sim 1-3 km can account for the thermal activity.

It has been postulated that the low-temperature systems in Iceland may have been developed by at least the four following processes or any combination of them (Arnórsson, 1995):

- 1) Deep flow of groundwater from highland to lowland areas through permeable structures driven by the hydraulic gradient;
- 2) Convection in young fractures formed by tectonic movements in old and relatively impermeable bedrock;
- 3) Drift of high-temperature geothermal systems out of the active volcanic belts in conjunction with their cooling and the extinction of the magma heat source; and
- 4) Magma intrusion, into the Quaternary or Tertiary formations that are adjacent to the active volcanic belts.

The basaltic lava pile in the Reykir and Laugarbakki area is of upper Miocene and late Pliocene age (3.3-8.5 My).

2.1 The Reykir at Reykjabraut geothermal area

The Reykir at Reykjabraut geothermal area is situated at the edge of the extinct Vatnsdalur central volcano. Prior to drilling, several springs were identified there with a combined flow estimated at about 2 l/s at a temperature of 68°C. Since 1967, six wells have been drilled and sufficient hot water obtained to pump 30-35 l/s of about 75°C hot water. The water was used for space heating of schools and farms in the neighbourhood and the township of Blönduós, a population of about 1000 people about 15 km to the north of Reykir. The flow declined with time and, in 1996, five shallow exploration wells were drilled (Björnsson and Fridleifsson, 1996); the results were used to locate well RR-12, drilled in late 1996, which yields about 50 l/s of water at 75°C. In 2006 further exploration took place at Reykir and 8 shallow gradient wells were drilled before a new production well, RR-21, was located and drilled to a depth of 1400 m. The well yields approximately 45 l/s at 73°C.

2.2 The Laugarbakki geothermal field

The Laugarbakki area is some distance south of one of the extinct central volcanoes (Figure 1). Hot springs with small flow have been known about for a long time at this site. In 1959, the temperature of one of them was found to be 88°C (Diaries of Jón Sólmundsson). The first well (LB-1) was drilled in 1964, and deepened in 1971. Another well was drilled in 1971 and 1972 and a third one in 1977. Well 2 and later well 3 were utilised for space heating of farms in the vicinity and the nearby village of Hvammstangi, with a population of about 700, since 1973. The water temperature is about 97°C and the wells yield 30-35 l/s upon pumping.

2.3 Reservoir rocks and alteration

The original rock in this area is basalt (Figure 2: blue colour area). But within the central volcanoes, acidic rocks are also found (Figure 2: yellow colour area). According to the drill cuttings from the main production wells at Reykir at Reykjabraut, RR-12 and RR-21, the major secondary minerals in the reservoir are: calcite, zeolites (laumontite, heulandites and stilbite), chalcedony, quartz, and pyrite (Gudbrandsson et al., 2007). The depth of RR-21 is 1400 m whereas well RR-12 is just 438 m.

Figure 3 shows the lithology of well RR-21. The lithology of the layer associated with the main aquifer in well RR-21 shows it to be fine-grained and partially fractured basalt formation. Calcite, quartz, zeolites and pyrite are the main alteration minerals. The aquifer is located close to an intrusion at 687 m and yields approximately 43 l/s.

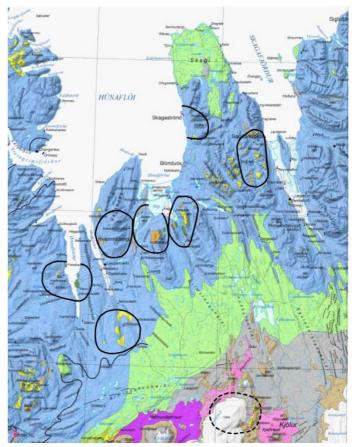


FIGURE 2: Geological map of the western part of North Iceland (from Jóhannesson and Saemundsson, 2009)

Lithology RR-21 Reykir - Reykjabraut

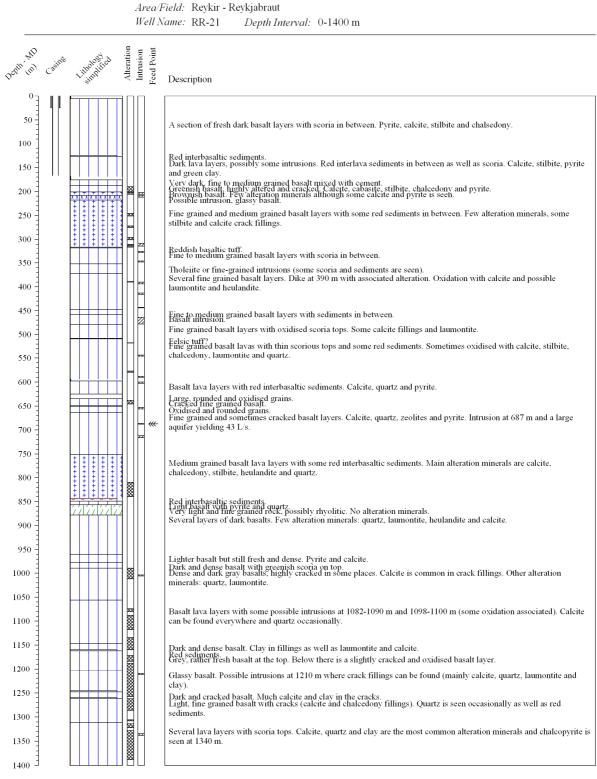


FIGURE 3: Lithological section of well RR-21 in the Reykir at Reykjabraut field (from Gudbrandsson et al., 2007)

3. CHEMICAL CHARACTERISTICS OF THE THERMAL FLUIDS

3.1 Classification of fluids

The Piper diagram classifies fluids on the basis of major ions. Based on it, the type of thermal water samples from Reykir at Reykjabraut is Na-SO₄-HCO₃, while at Laugarbakki it is Na-Cl-SO₄. The cold groundwater type in this area is Ca-Na-HCO₃ and the water from the warm spring is Na-SO₄ (Figure 4).

The Cl-SO₄-HCO₃ triangular diagram (Giggenbach, 1991) is commonly used for classifying waters of different origin. It not only filters out unsuitable samples for geochemical calculations, but also can provide an initial indication of mixing relationships or geographic groupings.

The source of Cl is seawater and rock dissolution, while for SO_4 , a small amount comes from seawater, whereas, a larger amount is usually derived from the oxidation of sulphide in volcanic steam, such as SO_2 and H_2S . Carbonates are obtained by the degassing of volcanic formations. In cold water, carbonates originate from biological sources.

Figure 5 shows that the thermal waters of Reykir at Reykjabraut are rich in SO_{4} , possibly influenced by some sulphide rich rocks. As mentioned above, the geothermal field is close to an extinct central volcano. The possibility of deepseated intrusions influencing the chemical composition of the thermal water cannot be ruled out.

At the Laugarbakki geothermal field, the concentration of Cl is higher than at Reykir at Reykjabraut. According to the location of the field, the fluids at Laugarbakki may have been influenced by seawater, which would account for an increase in Cl.

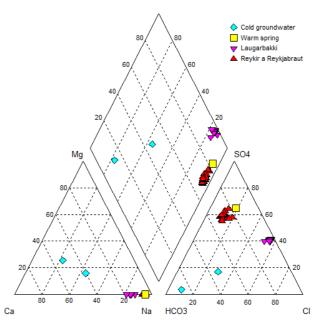


FIGURE 4: Piper diagram showing samples from Reykir at Reykjabraut and Laugarbakki area

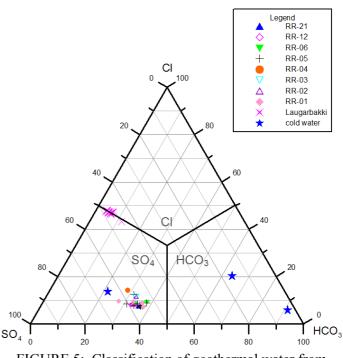


FIGURE 5: Classification of geothermal water from Reykir at Reykjabraut (compared with groundwater and geothermal water from Laugarbakki area)

The samples of cold groundwater are rich in HCO_3 . They are fresh waters with a composition close to precipitation in the area. But the warm groundwater (approx.40°C) may be mixed with geothermal water, explaining the increased temperature and its chemical composition being so close to that of geothermal water.

TABLE 1: Chemical composition of waters from Reykir at Reykjabraut in Iceland composition in mg/l

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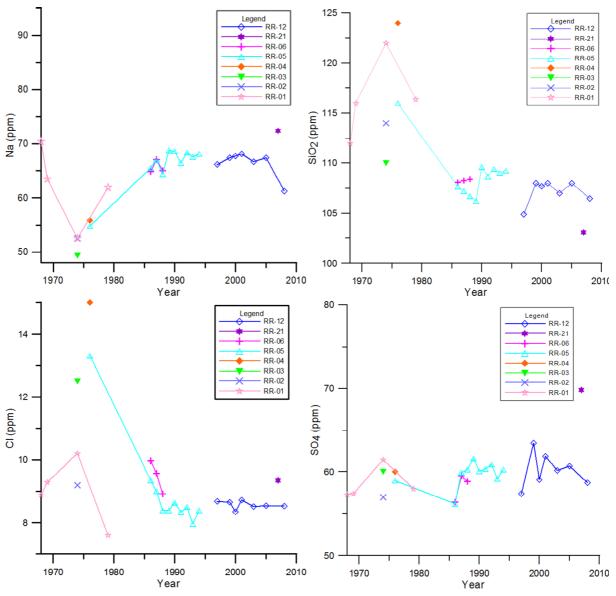
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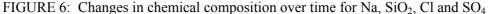
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3.2 Changes during utilisation

3.2.1 Chemical changes

In the Reykir at Reykjabraut geothermal field, wells RR-05 and RR-12 are the main production wells. The water is used for heating houses in the town Blönduós and the Húnavellir school and a few farmhouses in the area. Figure 6 shows the changes in the concentrations of, sodium, silica, chloride and sulphate in water from different wells at Reykir. At first, when pumping started, the waters may not have reached equilibrium and they might have been influenced by cooling water from the drilling process. Therefore, the chemical composition of the first sample may not necessarily represent the composition of the water from that particular well. In the following years, a new equilibrium was formed; the concentration became stable with little changes, especially in the main production wells (RR-05, RR-12) where most of the samples were collected. During utilisation, some changes in the chemical composition have been noticed, even a slight decrease. However, when analysing changes in the concentrations of different elements in water from a particular well, it is necessary to also study the discharge from the well at the time of sampling. The concentrations may, to some extent, depend on discharge due to influences from different aquifers in the well.





The data from well RR-12 shows that the biggest change happened between the years 1997 and 1999. If the first data points are ignored and a look taken at the data from 1999 to 2008, silica dropped from 108 to 106.5 mg/l, sodium changed from 67.4 to 61.3 mg/l, sulphate changed from 63.5 to 58.7 mg/l and chloride from 8.66 to 8.53 mg/l. All these changes are relatively small and some of the fluctuations may be caused by errors in the chemical analysis. The accuracy of the silica analysis in the ISOR lab is e.g. $\pm 2\%$.

3.2.2 Temperature changes

Figure 7 shows how the measured temperature at the wellhead at the time of water sampling had changed with time. As mentioned above, wells RR-05 and RR-12 are the major production wells and data showing temperature changes with time are available from them. If the first sample is ignored for each well, the data shows that the temperature has been fairly stable over the years, with even a slight increase. To see if this also applies to the reservoir temperature, we need to use indirect methods. such as silica geothermometers, Na-K geothermometers, Na-K-Mg triangular diagram and mixing models, to make estimates.

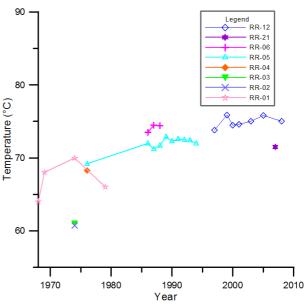


FIGURE 7: Measured temperature changes during the utilisation at Reykir at Reykjabraut

3.3 Geothermometers

Geothermometers are used to estimate subsurface temperatures and predict possible cooling in the reservoir during production. They have been classified into three groups, water or solution geothermometers, steam or gas geothermometers and isotope geothermometers. Water and steam geothermometers are collectively termed chemical geothermometers. On using geothermometers to estimate surface or aquifer temperature, there is a basic assumption that temperature-dependent chemical or isotope equilibrium prevails in the source aquifer. Further, that the chemical and isotopic reactions do not significantly modify the composition of the fluid as it ascends from the source aquifer to the point of sampling, whether it is a thermal spring, fumarole or wellhead (Arnórsson, 2000).

The most important geothermometers are the silica (quartz and chalcedony), Na/K and Na-K-Ca geothermometers. Different geothermometers are suitable for different reservoir situations. The main judgments are equilibrium between fluids and rocks, and mixing or degassing during up-flow. In order to get reasonable results, the validity of the assumptions of specific solution/mineral equilibrium and some of the physical and chemical processes involved should be taken into account.

WATCH is a computer program which can calculate reservoir temperature for a given geothermometer, compute the composition of aquifer fluids and the composition of the sample after cooling or boiling (Arnórsson and Bjarnason, 1993). From the WATCH results, we can also obtain the minerals' log Q and log K (Q-reaction quotient, K-equilibrium constant) which are used to calculate the saturation index to judge the mineral-solution equilibrium. The water geothermometers used in the WATCH program and in this report are listed here below.

Chalcedony geothermometer (Arnórsson et al., 1983):

$$t^{\circ}C = \frac{1112}{4.91 - \log S} - 273.15 \tag{1}$$

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Quartz geothermometer, 20-330°C (Fournier and Potter, 1982):

$$t^{\circ}C = \frac{1309}{5.19 - \log S} - 273.15 \tag{2}$$

Na/K geothermometer, 25-250°C (Arnórsson et al., 1983):

$$t^{\circ}C = \frac{933}{0.993 + \log\left(\frac{Na}{K}\right)} - 273.15 \tag{3}$$

Chalcedony is a very fine-grained variety of quartz composed of aggregates of tiny crystals. The individual quartz grains are so small that relatively large surfaces are energised compared to "normal" quartz and this results in an increase in solubility. Chalcedony is unstable in contact with water at temperatures above about 120-180°C because the smallest sized crystals completely dissolve relatively quickly and larger sized crystals grow large enough so that the surface energy is no longer a factor. Temperature, time, fluid composition, and prior history, all affect the size attained by quartz crystals. So a chalcedony geothermometer is more suitable for low-temperature geothermal fields while a quartz geothermometer is better used for high-temperature geothermal fields (Fournier, 1991).

The WATCH computer program (version 2.4) was used for calculations of silica (quartz and chalcedony) and Na-K temperatures (Bjarnason, 2010). The measured temperatures of the samples and results for the solute geothermometer (calculated subsurface temperatures) are summarized in Figure 8 and Table 2. Comparing the results from Table 2, we can see that the subsurface temperatures calculated by the chalcedony and Na-K geothermometers are quite similar while those taken by guartz geothermometers show much higher temperatures. According to the well temperature logs, the results from chalcedony and Na-K are much more reliable than the quartz temperatures.

The results from chalcedony and Na-K geothermometers, shown on the diagram in Figure 8, indicate changes in subsurface temperature (reservoir temperature) with time.

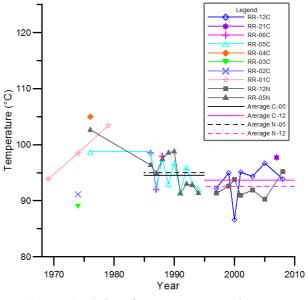


FIGURE 8: Subsurface temperature changes at Reykir at Reykjabraut field

In the legend, the well numbers with C indicate chalcedony temperatures and N Na-K temperatures. For well RR-05, the trend lines of the temperatures from the Na-K geothermometer and chalcedony geothermometer are similar. For both geothermometers of each major production well, the average subsurface temperature is comparable. Ignoring the bad samples, the average aquifer temperature for RR-05 is 94.5°C (chalcedony) and 95.0°C (Na-K). For RR-12, the average temperature is 93.6°C (chalcedony) and 92.5°C (Na-K). As in the previous diagrams, data from the two major production wells were chosen to describe this process. The curves fluctuate within a relatively large range, especially for well RR-05. Based on logs from well RR-12, it is believed that the well has two aquifers and the fluctuations may be due to different silica concentrations and pH and to a variable amount of water at different pumping rates. In other words, the chalcedony and Na-K geothermometer may fluctuate following these changes. The third sample from well RR-12 has an unusually high pH value, much higher than other samples from the same well. This may be an analysis mistake; it reveals a much lower chalcedony temperature than all the other samples. However, if we ignore the bad sample, we can see the tendency of a decrease in the subsurface temperature in RR-05 is almost 7°C, while the temperature of RR-12 has only decreased 1.1°C.

Well	Sample date	T _{measured}	T _{quartz}	Tchalcedony	T _{Na/K}
RR-21	2007.10.23	71.5	127.5	97.7	97.2
RR-12	2008.10.20	75	125.1	93.9	95.2
RR-12	2005.10.04	75.8	127.3	96.7	90.3
RR-12	2003.11.10	75	125.4	94.3	91.9
RR-12	2001.11.09	74.6	126.1	95.1	91.0
RR-12	2000.10.03	74.5	119.9	86.6	93.8
RR-12	1999.11.25	75.9	126.0	95.0	92.6
RR-12	1997.11.20	73.8	123.7	92.2	91.4
RR-06	1988.10.21	74.4	128.2	97.9	97.1
RR-06	1987.10.13	74.5	123.8	92.0	92.0
RR-06	1986.10.15	73.5	128.7	98.6	95.0
RR-05	1994.11.17	72	123.9	91.9	91.4
RR-05	1993.10.16	72.4	124.6	93.0	92.8
RR-05	1992.11.06	72.5	126.9	96.0	93.0
RR-05	1991.10.28	72.6	123.5	91.5	91.3
RR-05	1990.11.21	72.3	127.5	96.8	98.8
RR-05	1989.10.10	72.9	124.3	92.9	98.6
RR-05	1988.10.21	71.7	127.8	97.5	97.5
RR-05	1987.10.13	71.2	124.1	92.5	94.9
RR-05	1986.10.15	72	128.7	98.7	96.4
RR-05	1976.11.18	69.2	129.5	98.7	102.6
RR-04	1976.11.18	68.3	134.9	105.0	93.6
RR-03	1974.11.07	61	121.8	88.9	79.0
RR-02	1974.11.07	60.7	123.7	91.1	89.3
RR-01	1979.07.20	66	132.9	103.4	106.1
RR-01	1974.11.07	70	129.9	98.5	97.7
RR-01	1969.09.17	68	126.0	93.9	96.2
RR-01	1968.08.15	64	77.0	20.3	76.8

 TABLE 2: Temperature of different geothermometers for the thermal waters from the Reykir at Reykjabraut wells (°C)

3.4 Na-K-Mg equilibrium diagram

Another method for estimating subsurface temperature is the Na-K-Mg triangular diagram of Arnórsson et al. (1998). It also can indicate the equilibrium temperatures of minerals and the equilibrium state between the geothermal fluids and rocks. Giggenbach (1988) suggested that a triangular diagram with Na/1000, K/100 and Mg^{0.5} at the apices could be used to classify water as being in full equilibrium, partial equilibrium or immature (dissolution of rock with little or no chemical equilibrium). The triangular diagram is employed to determine whether the fluid has equilibrated with hydrothermal minerals as well as to predict the equilibration temperatures. It is based on the temperature dependence of the three following reactions:

$$K\text{-feldspar} + Na^{+} = Na\text{-feldspar} + K^{+} \tag{4}$$

 $2albit + 0.8K - feldspar + 1.6water + Mg^{2+} = 0.8muscovite + 0.2clinochlore + 5.4silica + 2Na^{+}$ (5)

$$2.8K-feldspar + 1.6water + Mg^{2+} = 0.8muscovite + 0.2clinochlore + 5.4silica + 2K^{+}$$
(6)

All involve minerals of the full equilibrium assemblage that are expected to form after isochemical recrystallization of average crustal rock under geothermal conditions. The Na, K and Mg contents of water in equilibrium with this assemblage are accessible to rigorous evolution (Giggenbach, 1991).

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The theoretical temperature dependence of the corresponding concentration quotients may then be used to find:

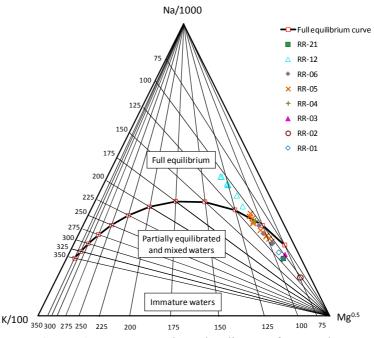
$$t_{kn} = 1390/(1.75 - L_{kn}) - 273$$
 and $t_{km} = 1390/(1.75 - L_{km}) - 273$ (7)

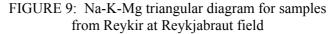
where $L_{kn} = \log (CK/CNa)$, and $L_{km} = \log (CK_2/CMg)$, Ci in mg/kg.

Individual application of Equation 7 frequently leads to significantly different apparent temperatures of equilibration. This observation is readily explained in terms of differing rates of readjustment of the two reactions to changes in the physical environment encountered by the rising water.

After plotting all of the samples from Revkir at Revkjabraut on the diagram, it is evident that most of the samples, except RR-12, are located in the partial equilibrium area which means that these waters have not reached full equilibrium between water and rock (Figure 9). The reason may be that the geothermal waters here have mixed with new recharge water and have not reached a new equilibrium with the rock. It must be an open hydrothermal system with adequate recharge water. Figure 9 also shows that the calculated t_{kn} varies within the range of 95-120°C while t_{km} changes in a wider range of approx. 75-133°C. However, cation geothermometer results can only be considered tentative since most of these waters are not in equilibrium with the reservoir rocks. It may also be possible that some mixing reactions take place in the reservoir. Therefore, temperatures obtained from the chalcedony geothermometer are accepted as being more reliable than those of the other cation geothermometers.

Figure 10 shows the Na-K-Mg triangular diagram for waters from Laugarbakki geothermal field, and it shows that almost all of the samples plot in the full equilibrium area.





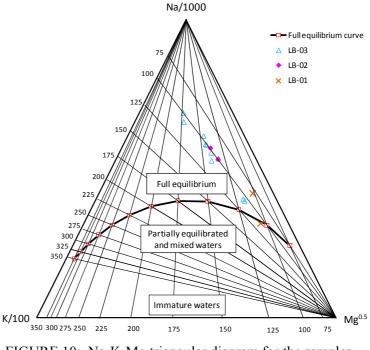


FIGURE 10: Na-K-Mg triangular diagram for the samples from Laugarbakki field

The reason could be that the system is a closed system, with influence from the recharge water due to

its slower recharge rate. Another reason is the higher temperature in the reservoir, which leads to a faster reaction rate between water and rock. Figure 10 shows that t_{kn} varies between 95 and 107°C.

3.5 Mineral-solution equilibrium

The mineral equilibrium approach is one method which estimates subsurface temperature in geothermal systems. It can reveal at which temperature equilibrium exists between the water and minerals and also can distinguish between equilibrated and non-equilibrated waters, and which minerals may precipitate during extraction and use of the water. It was proposed by Reed and Spycher (1984). It can be used to show temperature equilibrium between the water and minerals by giving a set of minerals over a range of temperatures.

The saturation index of water is widely used in hydro-geochemical studies; it is the study of mineral saturation in the groundwater: when SI = 0, the minerals in the water are at equilibrium; when SI < 0, it shows that the minerals in the solution have not reached saturation and mineral dissolution will occur; when SI > 0, the minerals in the water are said to be saturated and mineral precipitation will occur. The saturation index (SI) of the minerals is defined as: $SI = \log (IAP / K)$; for the mineral dissolution reactions, IAP is related to the ion activity product, and its value from the ion activity coefficient r and the concentration of the components are obtained by the product of m; K is a temperature balance constant. A saturation index of zero indicates that thermodynamic equilibrium exists with the solid phase of the relevant mineral. A negative (-) or a positive (+) index indicates under saturation and oversaturation, respectively.

When choosing the minerals for the SI diagram, we should take into account which hydrothermal minerals are present in the geothermal system. If most of the minerals indicate the same approximate equilibrium temperature, the average of the equilibrium temperature can be considered the best estimate of the subsurface temperature. In this report, the minerals chosen were chalcedony, quartz, anhydrite, laumontite, albite (low), fluorite, and calcite.

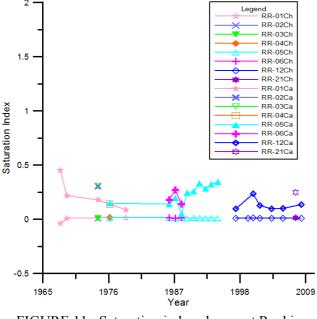
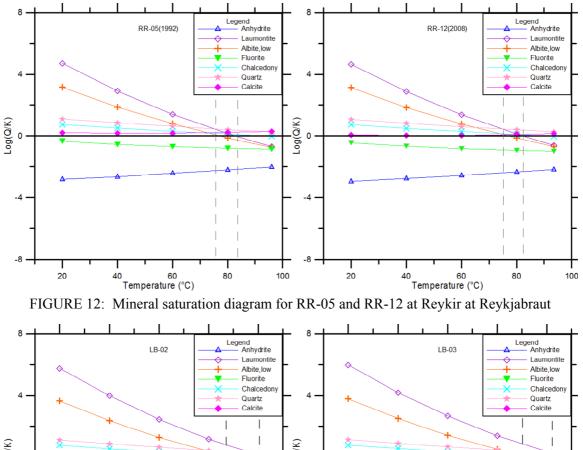


FIGURE 11: Saturation index change at Reykir at Reykjabraut over time

As mentioned before, the WATCH program can be used to calculate logQ and logK for geothermal water and the SI index can be calculated for every mineral. In order to see a change in the saturation index of chalcedony, the diagram was drawn, shown in Figure 11. It shows how the SI of chalcedony and calcite changes over time. In the legend, the well numbers with Ch represent the SI of chalcedony and the numbers with Ca those of calcite. The SI of both minerals is very low and slightly positive which means that these minerals should precipitate from the water. The super-saturation is, however, so little that it is not likely that these minerals will precipitate during utilisation of the water.

Figure 12 shows the SI-diagrams for two different wells, the main production wells, at Reykir at Reykjabraut, wells RR-05 and RR-12. Most of the minerals (except anhydrite and fluorite) cross the zero line at the temperature

range of approx. 78-88°C; that means that the water from the wells is close to saturation in this temperature range. Figure 13 shows the data from Laugarbakki geothermal field in a similar way and



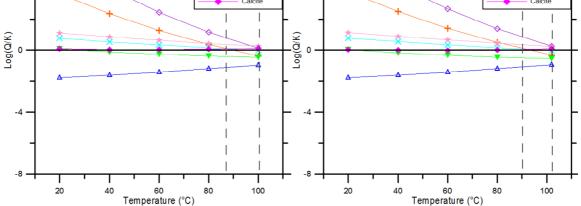


FIGURE 13: Mineral saturation diagram for LB-02 and LB-03 at Laugarbakki

shows that the subsurface temperature is in the range of 90-100°C.

Silica and calcite are the most common scaling minerals in the utilisation of geothermal systems. According to the diagrams above, the SI with respect to chalcedony, quartz and calcite indicates mostly oversaturation or a near equilibrium for geothermal waters of the two areas. It shows that there is little silica precipitation below the reservoir temperature while calcite is dissolved at any temperature. This indicates that silica scaling and carbonate scaling are not to be expected during production.

3.6 Mixing

Mixing models have been developed to allow for the estimation of the hot water component in mixed waters emerging in springs or discharged from shallow drill holes. There are essentially three kinds of mixing models. In this project, only the silica-enthalpy warm spring mixing model was studied.

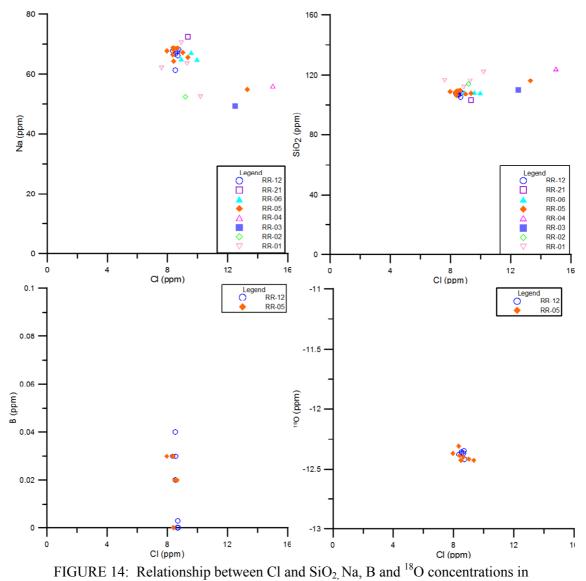
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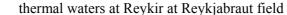
When hot water ascends from a geothermal reservoir, it may be cooled down by mixing in the up-flow with shallow, relatively cold waters, such as groundwater and seawater. The mixing may occur after a variable amount of conductive cooling of the hot water and before, after or during boiling. It also can upset chemical equilibrium between water and minerals, creating a tendency for the water to change composition after mixing with respect to reactive chemical components.

Mixing of geothermal and cold water as a mixing model can be applied to estimate subsurface temperatures in geothermal systems, where there are two solutions with quite different temperatures and where the concentrations of reactive components do not change much after mixing has occurred. Such a situation is likely to be confined to up-flow zones and such a model is not suitable for deep levels in geothermal reservoirs. So the application of geothermometers is more appropriate than mixing models for estimating subsurface temperatures.

3.6.1 Evidence of mixing

Using the geochemical data from Reykir at Reykjabraut and Laugarbakki, the evidence for mixing was investigated (Figure 14). The results are as follows:





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- 1) Evidence of mixing could not be found in downhole logs made at Reykir at Reykjabraut. The temperature is quite stable with a small decrease from the bottom of the wells to the surface.
- 2) Cl is a non-reactive component in the waters, because it only forms soluble minerals and its source of supply to the geothermal fluid is too limited to influence any mineral. After drawing diagrams of Cl-SiO₂, Cl-Na, Cl-B, Cl-¹⁸O based on the samples from Reykir at Reykjabraut, shown in Figure 14, it was concluded that there is no direct linear relationship indicating mixing; it is a more scattered correlation. This does not, however, rule out the possibility of mixing taking place.
- 3) However, for the Laugarbakki field, the concentration of Cl is higher in the geothermal water than at Reykir. The water may be influenced primarily by seawater. In the next section, the isotopic composition diagram addresses the issue of mixing in this field.

3.6.2 The silica-enthalpy mixing model

The dissolved silica-enthalpy diagram (Figure 15) demonstrates the procedure for calculating the initial enthalpy and, hence, the reservoir temperature, of a geothermal water that has mixed with a low-temperature water (Fournier, 1981). It can be used to correct silica concentrations for adiabatic cooling with single-stage steam loss at any temperature.

Enthalpy is a derived property which can be obtained from steam tables, provided that the temperature, pressure and salinity are known. It is useful to use enthalpy as a coordinate rather than temperature, because the combined heat contents of two waters at different temperatures are conserved when those waters are mixed, but the combined temperatures are not. It can handle non-boiled and boiled mixed waters separately. The temperature can be retrieved from the steam tables.

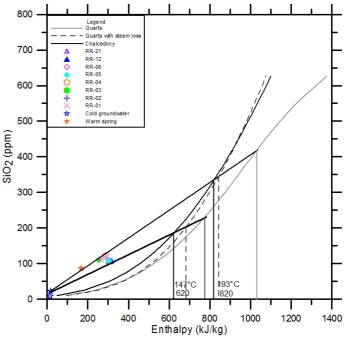


FIGURE 15: The silica-enthalpy mixing model for geothermal samples from Reykir at Reykjabraut

The dissolved silica concentration of mixed water and a silica-enthalpy diagram may be used to determine the temperature of the hot-water component, provided that silica has not precipitated before or after mixing, and that there has been no conductive cooling of the water. However, it is not always easy to determine that a given hot-spring water is a mixture of hotter and colder waters. It is even harder to verify that silica has not precipitated or that conductive cooling has not occurred.

The silica-enthalpy mixing model is one of the commonly applied mixing models used to estimate subsurface temperature. When applying mixing models to estimate subsurface temperature, several simplifying assumptions are made. Conservation of mass and heat is always assumed, both during and after mixing. Thus, it is assumed that chemical reactions occurring after mixing are insignificant and do not modify water composition.

As shown in Figure 15, the best-fit line between the cold groundwater and the possibly mixed geothermal water from Reykir at Reykjabraut intersects the chalcedony solubility curve at a point of enthalpy equal to 620-820 kJ/kg, which indicates a reservoir temperature of 147-193°C which is much higher than the reservoir temperature obtained by the chalcedony geothermometer. The reason is the excess silica, which may originate from the acidic rocks of the extinct central volcano (70% of silica).

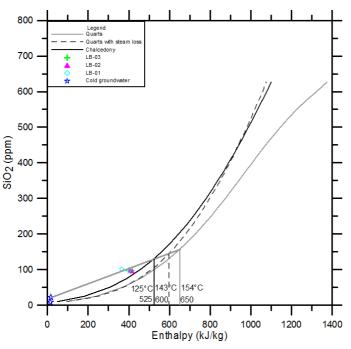


FIGURE 16: The silica-enthalpy mixing model for geothermal samples from Laugarbakki

That means the water-rock interaction influenced the geothermal water a lot and the mixing model is not applicable here. But in the Laugarbakki field (Figure 16), the silica-enthalpy diagram indicates a similar value of subsurface temperature as the geothermometers, approximately 125°C.

3.7 Summary

The geothermal fluids from Reykir at Reykjabraut are classified as $Na-SO_4$ - HCO_3 type, which is SO_4 -rich geothermal water. The reason may be the sulphide rich rocks because of the extinct volcano. But for Laugarbakki field, the geothermal fluids are a little enriched with Cl-rich water, indicating some seawater influence.

During utilisation, a slight decrease of chemical concentration occurred over

time. The measured temperature decreased slightly. The WATCH program was used to calculate different geothermometers and to get an idea of the subsurface temperature in the reservoir. The aquifer temperatures of different samples were obtained. They have a similar trend to that of measured temperature but fluctuated in a wider range. According to the results of the chalcedony geothermometer, the average temperature in the reservoir is 95° C.

The Na-K-Mg triangular diagram also shows the subsurface temperature but it is a little higher than the results from the WATCH program. This depends on which formula is chosen for the calculation. The diagrams can also indicate that the geothermal waters in the reservoir of Reykir at Reykjabraut have not reached equilibrium, which means they are in an open system and have adequate recharge.

The multiple mineral equilibrium approach reveals the reservoir temperatures of wells RR-03, RR-12, LB-02, LB-03. In Reykir at Reykjabraut, the temperature is in a range of approx. 78-88°C. In the Laugarbakki area, the reservoir temperature varies from 90 to 100°C. According to the diagram, it also indicates that some minerals precipitated or dissolved in the reservoir. Pyrite always precipitated while anhydrite, fluorite always dissolved. Silica and calcite dissolved at reservoir temperature and to a lesser extend at a lower temperature; that means there is no likelihood of silica and carbonate scaling during production.

There is uncertain evidence as to whether mixing occurs in the reservoir at Reykir at Reykjabraut. The silica-enthalpy mixing model is not well suited to estimate the subsurface temperature for the geothermal water because of the excess silica from acidic rocks. But for Laugarbakki area, the mixing model indicates a subsurface temperature of about 125°C.

4. ORIGIN, RECHARGE AND TRANSPORT OF FLUID

4.1 Origin of the geothermal water

Geothermal fluids originate mainly from meteoric water (Craig, 1963). An important aspect of

geothermal investigations is to determine the recharge to the geothermal systems. Craig (1963) established the isotopic characteristics (δD and $\delta^{l\delta}O$) of precipitation related to latitude and altitude as well as to continental effects. The values of δD and $\delta^{l\delta}O$ in precipitation were approximately related to the meteoric water line by:

$$\delta D = 8\delta^{18}O + 10\tag{8}$$

 δD can also indicate recharge and the relative contribution of magmatic fluids. Together with $\delta^{18}O$ it is a valuable indicator for mixing and vapour separation processes. Unlike $\delta^{18}O$, δD is hardly affected by exchange processes.

It is not easy to measure absolute isotopic ratios accurately, but differences in isotopic ratios between a particular sample and a standard can be measured accurately. For this reason isotopic concentrations are conveniently expressed in delta notation (δ) as parts per thousand, or

$$\delta = \frac{R_{sample} - R_{standard}}{R_{standard}} \times 1000 \tag{9}$$

 $R_{standard}$ stands for the isotopic ratio of the standard. The standard that was used for measuring delta values for the isotopes of oxygen and the stable isotopes of hydrogen is the Standard Mean Ocean Water (SMOW) (Arnórsson, 2000).

According to Figure 17, all the samples from Reykir at Reykjabraut are located around the Global Meteoric Line. This is strong evidence for proving that the origin of the geothermal water is mostly from precipitation. But for the Laugarbakki field, there seems to be a small oxygen shift. That is mainly because of its higher temperature (above 100°C). The warm spring water may have mixed with the geothermal water from another field with different origins.

4.2 Recharge, discharge and transport

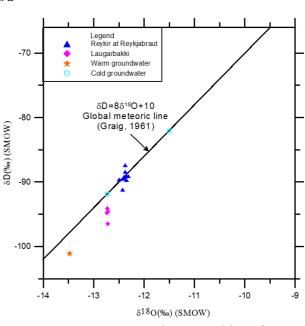


FIGURE 17: Isotopic composition of geothermal water and groundwater in the study area

Recharge elevation is a way to determine the recharge area of the geothermal fluids. As deuterium is not only a non-reactive component in water, but is also sensitively influenced by elevation and latitude, it can be considered a natural tracer. The relationship between δD and elevation is as follows (Pang Zhonghe et al., 1993):

$$gradD = \frac{\Delta\delta D}{\Delta H}$$
 or $H_{origin} = (\delta D_s - \delta D_p)/grad D + H_s$ (10)

where $\Delta \delta D = \delta D_s \cdot \delta D_p$; $\Delta H = H_0 \cdot H_s$; grad D = The decreasing gradient with a height of D (‰/100 m); $H_o =$ The elevation of the recharge area; $\delta D_s = \delta D$ of the recharge area the same as δD of samples; $\delta D_p = \delta D$ of the local precipitation; $H_s =$ The elevation of the aquifer location. The elevation of the well location can be obtained from the contour map. Then, using data from wells RR-12 and RR-21 to calculate the equation above, the elevation of the recharge area is found to range from 2705 to 4190 m. Based on the contour map, the highest place in this area are the two glaciers, Langjökull and Hofsjökull in Central Iceland. They have a height of 1300 and 1700 m, respectively. Maybe there is some deviation of the data, but when compared with other evidence such as deuterium contours (Figure 18), we can confirm that the recharge water is originally from the precipitation of these two glaciers.

The low deuterium content of geothermal waters in relation to local precipitation has been explained by distant source inland areas on higher ground where the deuterium content of the precipitation is lower (more negative) than that in the respective geothermal fields (Árnason, 1976). In his classic isotopic studies on natural waters in Iceland, Árnason extensively sampled the country waters from local streams and springs, considered to be representative of the average deuterium content of the local precipitation, to construct a map showing how the deuterium content of the local precipitation varied across the country. By drawing a line perpendicular to topographic contours from a particular geothermal field until the deuterium content of the geothermal water matched with the value of the precipitation, he defined what he considered to be the groundwater flow from the recharge area to that geothermal field.

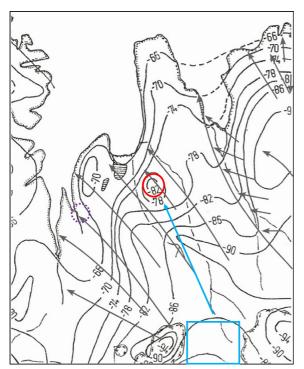


FIGURE 18: Deuterium in groundwater; arrows indicate recharge for thermal systems (from Árnason, 1976)

Figure 18 shows the subsurface flow of water from the recharge areas to the geothermal fields according to the interpretation of deuterium data in meteoric and geothermal waters by Árnason (1976). The arrowheads coincide with a particular geothermal field, and the tail of the same arrow to the recharge area for that geothermal field.

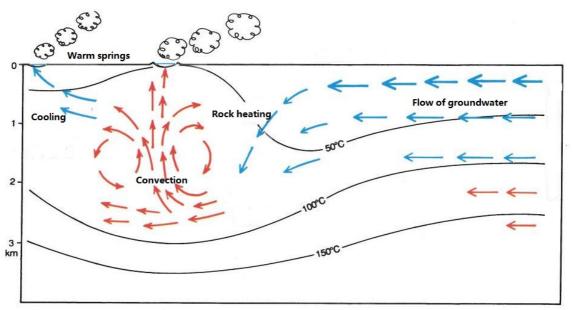
According to the map, the solid circle is Reykir at Reykjabraut. The arrow indicates that the recharge area is the rectangular area in the middle of the two glaciers. That is strong evidence to prove the recharge fluid comes from the two glaciers. The precipitation flowed down to this area and then along the fracture or some other channels, mixed with the groundwater, reacted with the rocks, and finally flowed into the reservoir (Figure 19). That theory is only hypothetical, but in the next step, it may be proven by the water-rock interaction model. The dotted circle is the Laugarbakki area which is recharged directly by the Langjökull glacier.

As mentioned above, the chemical composition of the warm spring in Reykir at Reykjabraut is similar to the geothermal water. Aside from geothermal

wells, another kind of discharge channel for the reservoir could be warm springs. The geothermal water flows along the fractures, is cooled down by the rocks, and finally reaches the surface and forms warm springs (Figure 19).

4.3 Discussion

The origin of the geothermal waters in both Reykir at Reykjabraut and Laugarbakki is from precipitation. But the waters from Laugarbakki area may be influenced by seawater. The recharge area of the two fields is the area between the two glaciers. The rain water flows down from the



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FIGURE 19: Schematic section of the geothermal system in Reykir at Reykjabraut (adopted from Björnsson et al., 1990)

glaciers and converges with groundwater; it is then transported into the reservoir through channels. On the way to the reservoir, it may mix with groundwater, react with rocks and be heated up by hot rocks. The discharge channels of the geothermal water are warm springs and fractures to the ocean. Because of a lack of ¹⁴C data, the age of the geothermal water cannot be estimated in this project; it needs to be analysed in the future by collecting special samples.

5. WATER-ROCK INTERACTION

5.1 General

Water-rock interaction is an important process which occurs in a hydrothermal system. Isotopic exchange, chemical evolution of geothermal fluids and alteration of rocks are closely related to water-rock interaction. Therefore, a study of the interaction between geothermal fluids and reservoir rocks will be helpful; not only to determine the origin of geothermal fluids, but also to explore the mechanism of many complicated geochemical reactions in a geothermal system and to provide useful information on the exploitation and utilisation of geothermal resources (Zhang Zhanzhi, 2001).

Water is a complicated solution, which can be simple ions, or a complex synthesis of different ions or complex molecules. These components take part in the complex physical and chemical reaction and form complicated components. So it is difficult for traditional geochemical methods to deal with such a complex process. Hydro-chemical modelling, based on thermodynamics, provides some solutions for these problems. There are two general approaches of water-rock interaction modelling. The first one is "forward modelling", which predicts water composition, and mass transfer that can result from hypothesized reactions. The second one is "inverse modelling", which defines mass transfer from observed chemical and isotopic data. Forward modelling begins with a defined initial water composition and simulates the chemical evolution of the water and rock for a set of specified reactions. Inverse modelling produces mineral mass transfers that can explain changes in the compositions observed in the hydrothermal system. A series of reaction programs, such as PHREEQC, NETPATH and CHILLER were developed to model natural water-rock interactions. In this report, the model was performed by PHREEQC that operates for geothermal systems. Using a mass-balance model, the dissolution and precipitation of minerals was calculated after the transport of the recharge fluids. However, most of the possible reactions which are defined in the program may

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not occur in natural systems. So it is necessary to compare the results from the computer models with their natural counterparts to identify similarities and differences. This type of analysis and comparison greatly enhances the understanding of natural processes (Reed, 1982).

PHREEQC version 2 is a computer program for simulating chemical reactions and transport processes in natural or polluted water. The program is based on equilibrium chemistry of aqueous solutions interacting with minerals, gases, solid solutions, exchangers, and sorption surfaces, but also includes the capability to model kinetic reactions with rate equations that are completely user-specified in the form of basic statements. Kinetic and equilibrium reactants can be interconnected, for example by linking the number of surface sites to the amount of a kinetic reactant that is consumed (or produced) during the course of a model period. (PHREEQC manual, 2010)

A mass-balance model is used to investigate the chemical reaction during the transport of groundwater. According to the reactions between water and rock, it can confirm the solution and mineral changes (dissolution or precipitation) of two different points quantitatively. If it is assumed that there are available two water analysis results from two points along the groundwater flow path; then the upstream sample can be considered as the initial water, while the downstream sample can be considered final water. An equation can then be obtained based on the principle of the mass-balance model:

$$Initial water + reactant = final water + products$$
(11)

5.2 Water-rock interaction modelling with PHREEQC

5.2.1 Solution composition and the reaction path

The purpose of water-rock interaction modelling is to reveal which geochemical reaction happened during the recharge of groundwater, and also to evaluate the strength of the reaction; in other words, how many minerals took part in the reaction and how many secondary minerals formed. As confirmed in Section 4, the origin of the geothermal fluids at Reykir at Reykjabraut is precipitation. The recharge area is in the southeast part of the geothermal field (Figure 18). If one chooses the arrow as the reaction path and samples from a cold spring in the recharge area as the original water, then the chemical composition of the water is HCO_3 -Na. The composition of the groundwater samples from the recharge area is listed below:

 TABLE 3: Chemical composition of a cold spring representative for Reykir at Reykjabraut (concentrations in mg/l)

T _{measured}	pН	SiO ₂	Na	K	Mg	Ca	F	Cl	SO ₄	HCO ₃
3.2	8.88	8.65	2.97	0.28	0.57	2.39	0.03	2.35	1.84	7.35

 $T_{measured} = Measured temperature (°C)$

5.2.2 Mineral composition

As already mentioned in the geological background, the original rock in this area is basalt, which is composed of silica, iron, calcium, aluminium, manganese etc. A representative analysis of basalt is shown in Table 4.

5.2.3 Reaction condition and the temperature setting

Data on the original solution (Table 3) and original rock (Table 4) were input into PHREEQC to make a model. The blocks which were used in this program are solution, phases, reaction, solution of the master species, solution of the species, and the reaction temperature.

Formula	wt%	mol%	Formula	wt%	mol%
SiO ₂	0.492	0.527	Fe ₂ O ₃	0.026	0.010
FeO	0.116	0.103	MnO	0.002	0.002
CaO	0.104	0.119	Na ₂ O	0.026	0.027
MgO	0.061	0.098	K ₂ O	0.005	0.003
TiO ₂	0.026	0.021	P_2O_5	0.002	0.001
Al_2O_3	0.14	0.088			

TABLE 4: Chemical composition and relative stoichiometry of basalt

Solution is used to define the original water, phase is used to define the original mineral, and reaction is used to define the possible reactions which may take place between water and rock (Appelo and Postma, 2007). The temperature of the waters (from groundwater to the reservoir, in the range of 20-100°C) is used to predict which solution and secondary minerals will form, and how much of each. This water-rock interaction system can be written as shown in Table 5.

 TABLE 5: Possible dissolving reactions in the hydrothermal system

Formula	Reaction equations	Formula	Reaction equations
SiO ₂	$SiO_2 + 2H_2O = H_4SiO_4$	Fe ₂ O ₃	$Fe_2O_3 + 6H^+ = 2Fe^{+3} + 3H_2O$
FeO	$FeO + 2H^+ = Fe^{+2} + H_2O$	MnO	$MnO + 2H^{+} = H_2O + Mn^{+2}$
CaO	$CaO+2H^{+} = Ca^{+2}+H_{2}O$	Na ₂ O	$Na_2O + 2H^+ = H_2O + 2Na^+$
MgO	$MgO+2H^{+} = Mg^{+2}+H_{2}O$	K ₂ O	$K_2O + 2H^+ = H_2O + K^+$
TiO ₂	$TiO_2 + 2H_2O = Ti(OH)_4$	P_2O_5	$P_2O_5 + 3H_2O = 2H_3PO_4$
Al_2O_3	$Al_2O_3 + 6H^+ = 2Al^{+3} + 3H_2O$		

5.2.4 Results of the model

The purpose of the mass-balance is not only to explain changes in the chemical composition in both the solution and rocks quantitatively, but also to explore the geochemical mechanism which caused the changes. Table 6 lists the simulation results for the geothermal water in Reykir at Reykjabraut.

TABLE 6: Simulation results for geothermal water at Reykir at Reykjabraut

pН	SiO ₂	Na	K	Mg	Ca	F	Cl	SO ₄	HCO ₃	Ti
8.13	8.65	0.71	0.28	0.01	1.59	0.0021	2.35	6.14	7.35	0.28

With the water-rock interaction, secondary minerals start forming. Table 7 shows the main species of the mineral types and the saturation index of each.

Minerals	Formula	Saturation index	Minerals	Formula	Saturation index
Anhydrite	$CaSO_4$	-2.95	Aragonite	CaCO ₃	0.87
Calcite	CaCO ₃	0.92	Boehmite	Alooh	2.31
Chalcedony	SiO ₂	2.34	Brucite	$Mg(OH)_2$	3.27
Chrysotile	Mg ₃ Si ₂ O ₅ (OH) ₄	22.18	Greenalite	Fe ₃ Si ₂ O ₅ (OH) ₄	21.14
Fluorite	CaF_2	-5.12	Hercynite	FeAl ₂ O ₄	15.87
Pyrite	FeS ₂	-19.40	Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄	10.32
Quartz	SiO ₂	2.70	Goethite	FeOOH	8.97
Gibbsite	Al(OH) ₃	1.84	Halloysite	Al ₂ Si ₂ O ₅ (OH) ₄	9.30

TABLE 7: Calculation of saturation index of secondary minerals

5.3 Summary of the simulation results

When comparing the solution samples taken from well RR-21 with modelled results, some compositions are similar others not. This is because influence from mixing was ignored in the model; the only concern being the effect of water-rock interaction. It has been shown that there are some sulphide minerals in the reservoir which influence the composition of the geothermal fluids. And no attention was paid to mixing or the sulphide minerals in the model. The result is, thus, based on an assumption for an ideal situation, where fluid composition only changes due to water-rock interaction.

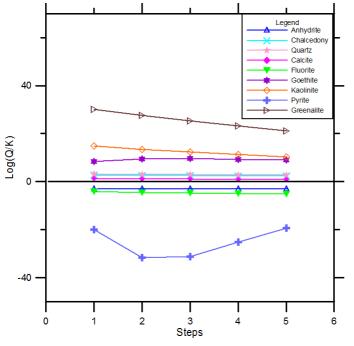


FIGURE 20: Mineral saturation diagram for simulated data from PHREEQC

Based on the cutting analysis of well RR-21, the major secondary minerals in this area are calcite, zeolites (laumontite, heulandites and stilbite), chalcedony, quartz, and pyrite, which are also included in Table 7 above. Most of the alteration minerals are identified both in actual field data and in the model.

In the simulation results, no zeolites or pyrite were seen in the reservoir. But chrysotile, hercynite, kaolinite, halloysite and greenalite, which were not found in the cutting analysis, were obtained in large amounts in the simulation results. However, as mentioned in Section 1, acidic rocks are the original rocks at Reykir at Reykjabraut. Furthermore, it has been proven that pyrite is one of the major secondary minerals in acidic rocks in Iceland (Björke, 2010). So the pyrite should mostly originate from water-rock

interaction between water and acidic rocks, and, hence, may have formed a long time ago due to the extinct central volcano.

6. CONCLUSIONS

- 1) The reservoir rocks of the geothermal fields are basalts of Tertiary age. According to drill cutting analyses of RR-21, the most abundant alteration minerals are calcite, zeolites, chalcedony, quartz and pyrite.
- 2) Some geochemical methods were applied to the low-temperature areas of Reykir at Reykjabraut and Laugarbakki in N-Iceland. The geothermal fluids from Reykir at Reykjabraut are classified as Na-SO₄-HCO₃ type, which is SO₄-rich geothermal waters. But for the nearby Laugarbakki field, the geothermal fluids are influenced by Cl-rich waters. During utilisation, there has been a slight decrease in chemical concentrations over time. The measured temperature has also decreased slightly over time. Using geothermometers and the program WATCH to calculate the subsurface temperature revealed similar trends but the temperatures had greater fluctuations. According to the results of the chalcedony geothermometer, the average temperature in the reservoir should be 95°C. The Na-K-Mg triangular diagram indicates that the geothermal waters in the reservoir of Reykir at Reykjabraut have not reached full equilibrium, which means they are in an open system and have adequate recharge. The multiple mineral equilibrium approach revealed that pyrite was precipitated while anhydrite and fluorite were dissolved. No silica or carbonate scaling are likely to occur during production.

- 3) The origin of the geothermal waters in both Reykir at Reykjabraut and Laugarbakki is meteoric. The recharge area of the two fields is believed to be the area between Langjökull and Hofsjökull glaciers. The rainwater flows down from the glaciers, merges with the groundwater, and then penetrates into the ground. On the way to the reservoir, the water may mix with other groundwaters, react with rocks and be heated up, finally forming the geothermal fluid.
- 4) Using PHREEQC, the possible evolution of the fluids was modelled. It suggested that most of the common cations found in the geothermal field originate from the dissolution of rocks. The higher SO₄ concentration in the samples is derived from sulphide minerals originating from the formation of the reservoir rocks. Water-rock interaction in the study area is not the only factor impacting fluid composition and alteration minerals. Influence from the acidic rocks associated with the extinct central volcano and, also, possible intrusions associated with it is also expected.

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