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MODELING OF FORMATION OF ACID WATER DISCHARGED FROM HIGH TEMPERATURE GEOTHERMAL RESERVOIR

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ABSTRACT

The Fe-rich acid geothermal waters (pH = 3-5) associated with large enthalpy fluids are discharged from the high temperature reservoirs (over 300°C) in the Fushime (Yamagawa) and Kakkonda geothermal systems. In contrast, the moderate temperature reservoirs in these geothermal systems produce neutral to slightly alkaline waters with a little or no excess enthalpies.

The geothermal water in Fushime is derived from seawater, while that in Kakkonda is believed to be of meteoric origin. In order to study the formation processes of these saline and dilute acid waters, numerical modeling based on chemical equilibrium for liquid-solid-gas phases was carried out. The models involve boiling in the reservoir caused by production-induced pressure decrease near the well and consequent reactions with wall rocks.

The calculated fluid compositions through the models closely agree with the compositions of the fluids discharged from the high temperature reservoirs in Fushime and Kakkonda. The calculation results indicate that the H^+ supply from sphalerite precipitation during boiling is the most probable cause of the observed acidity in the saline Fushime waters. On the other hand, the H^+ supply from the reactions with pyrite and magnetite and from H_2 redox reaction possibly contributes to the observed acidity in the dilute Kakkonda waters. The models demonstrate that production-induced boiling and consequent chemical reactions in the reservoirs are able to produce the observed acidity, without any contribution of acidic volatiles such as HCl and SO_2.

1. INTRODUCTION

As the geothermal energy exploitation reached to the high temperature and deep reservoirs, acid waters of pH 3 to 5 have been found in the Fushime (Yamagawa) and Kakkonda geothermal field, Japan (Akaku *et al*., 1991; Yanagiya *et al*., 1996). The geothermal water in the Fushime system is derived from seawater whereas that in Kakkonda is believed to be of meteoric origin. The higher temperature reservoirs in these fields (over 300°C) produce acid waters containing remarkable Fe^{2+}. Local near-well boiling caused by production-induced pressure decrease is common in these reservoirs. On the other hand, the moderate temperature reservoirs in these fields produce neutral to slightly alkaline waters containing much lower Fe^{2+}. The single liquid phase feed or a little excess enthalpy discharge is usual.

Since the acid waters sometimes introduce engineering problems related to corrosion and scale formation, it is important to know the source of acidity of these waters. This paper aims to report model calculations to investigate the effects of production-induced boiling in reservoir and consequent reaction processes with wall rocks.

2. OUTLINE OF THE GEOTHERMAL SYSTEMS

2.1 The Fushime geothermal system

The Fushime geothermal field is located close to the southeastern coast of the Satsuma Peninsula in Kyushu. The temperatures measured in some wells exceed 350°C at depths below 1,500 m. The wells penetrate thick dacitic to andesitic tuffs and lavas. The most productive reservoir is situated around the intrusive dacite found at depths below 1,600 m (*e.g.*, Okada *et al*., 2000).

The pH of the waters discharged from wells is in the range of 3.8-8.0 at room temperature, after steam separation. The acid waters (pH < 5) are discharged from higher temperature wells above 300°C, with large excess enthalpy. The temperatures at which the waters are in equilibrium with Na-feldspar and K-feldspar (330°-350°C) are higher than the quartz geothermometer temperatures (270°-325°C). The waters are almost exactly saturated with anhydrite at the quartz geothermometer temperatures (Akaku *et al*., 1991). It is possible that the slow readjustment of Na^+/K^+, relative to re-equilibration with quartz and anhydrite, in response to decreasing temperatures by production-induced reservoir boiling is a cause of the discrepancy. In contrast, the neutral waters are discharged from the lower temperature wells (usually less than 300°C), with small excess enthalpies.

As the chloride concentrations, Br/Cl ratios and δD in the high temperature reservoir waters are close to those of seawater, it is believed that the geothermal fluid originates from seawater. Various combinations of boiling of the seawater reacted with rocks at 350°C and dilution by groundwater are believed to occur in the geothermal system (Akaku *et al*., 1991, Okada *et al*., 2000). The chemical composition of the waters is characterized by depletion in Mg^{2+} and SO_4^{2-}, whereas enrichment in K^+, Ca^{2+}, Fe^{2+}, Mn^{2+}, Zn^{2+}, Pb^{2+}, SiO_2, *etc.* over those of seawater. This characteristic is common in both acid and neutral waters. However, it is noticeable that the acid waters contain much larger Fe^{2+} than the neutral waters.

The principal alteration minerals observed in the production zone are quartz, albite, K-feldspar, sericite, chlorite, epidote and anhydrite. An abundance of K-feldspar is the most distinctive feature around the feed points of the wells. There is no remarkable difference in alteration mineral assemblage between the high temperature (over 300°C) and the moderate temperature production zones (Yagi and Kai, 1990).

2.2 The Kakkonda geothermal system

The Kakkonda geothermal field is located inland in the northeastern part of Japan. The temperature profiles of the geothermal system have a steep increase at depths around 1,500 m, suggesting that two different convection cells exist. The upper and lower convection cells are called the shallow and deep reservoir respectively. The shallow reservoir in Miocene dacitic tuffs and shales, whose temperature is from 230° to 260°C, is very permeable. The deep reservoir in Pre-Tertiary formations and a neo-granitic pluton (the Kakkonda granite), whose temperature ranges from 300° to 350°C, is less permeable. Single-phase liquid feed is common in the shallow reservoir, while excess enthalpy conditions (two-phase feed) commonly develop during production in the deep reservoir (Hanano and Takanohashi, 1993).

Yanagiya *et al*. (1996) reported that the waters from the shallow reservoir are slightly alkaline (pH = 8-9, measured at room temperature, after steam separation), but those from the deep reservoir are acidic (pH = 3-5). The acid waters contain remarkable Fe^{2+} than the neutral waters. The Na/K geothermometry (*e.g*., Giggenbach, 1988) suggests that the deep waters are in equilibrium with Na/K-feldspars at about 300°C, while most of deep waters are supersaturated with respect to quartz at 300°C.

The origin of the both waters is probably meteoric based on the isotopic compositions. The chloride concentrations in the shallow reservoir waters were about 500 mg/kg before they were disturbed by reinjection of waste water. The deep reservoir waters have about 1,000 mg/kg of chloride, although there are some uncertainties caused by excess enthalpy discharge conditions (Yanagiya *et al*., 1996).

Yanagiya *et al*. (1996) interpreted that the acid waters from the deep reservoir have not fully attained to the equilibrium with the minerals in wall rocks. A possible sustained supply of magmatic volatiles such as HCl from a heat source is considered. The immature deep waters may be neutralized by the reactions with wall rocks during ascent.

The deepest well in Kakkonda, WD-1a, drilled by the New Energy and Industrial Technology Development Organization (NEDO) penetrated the Quaternary granite, a heat source of the geothermal system. The maximum temperature recorded in the well was over 500°C (*e.g.,* Doi *et al*., 1998). Emission of CO_2 and H_2S gas was experienced during drilling. Although the hypersaline Na-Fe-K-Mn-Ca-Cl type brine was obtained from the bottom of the well after the circulation of river water, its pH was 5.2. The sustained supply of magmatic volatiles from a heat source was not confirmed (*e.g.,* Kasai *et al*., 1998).

The secondary minerals in the Kakkonda system were formed through both of the contact metamorphism by the Quaternary granite and the hydrothermal alteration. Abundant and common hydrothermal minerals in the deep reservoir are quartz, albite, chlorite, muscovite, anhydrite and pyrite. K-feldspar, magnetite, pyrrhotite, hematite and actinolite-tremolite are also observed as hydrothermal alteration minerals in the deep reservoir (Doi *et al*., 1998). Acid alteration minerals such as alunite, sulfur, kaolinite and pyrophyllite are not observed in the deep reservoir and heat source, also arguing against the sustained supply of acidic volatiles.

3. MODEL CALCULATIONS

3.1 Calculation procedures

Numerical modeling is performed by using CHILLER (Reed, 1982; Spycher and Reed, 1992), a multi-component chemical equilibrium calculation program. For simulating effects of production-induced boiling in reservoir, we envision the following processes. The liquid water fully equilibrated with the alteration minerals in wall rocks begins to boil owing to production-induced pressure decrease near the well. The boiling in reservoir sometimes involves isothermal process due to heat transfer from wall rocks to the fluid. In response to the changes of physical conditions by boiling, the reactions of the fluid with the alteration minerals proceed to establish a new equilibrium until it enters the well. However, some reactions are too slow to attain equilibrium. In the well and surface equipment, boiling and consequent cooling are continued. They result in precipitation of scale.

The chloride concentration of the primary reservoir water (single-phase liquid) for the Fushime case is set to 19,000 ppm, being equal to that in seawater. For the Kakkonda case, it is set to 1,000 ppm. The CO_2 concentrations of the waters are set to 1,230 ppm and 250 ppm, based on the measured total discharge concentrations for the Fushime well SKG-16 and for the Kakkkonda well-19 (Akaku *et al*., 1991, Yanagiya *et al*., 1996). The compositions of the full equilibrium waters are computed by adding the principal alteration and scale minerals observed in the fields into these waters at 350°C, until the waters are saturated with them. The minerals used in the calculations are quartz, albite, K-feldspar, muscovite, clinochlore (Mg end-member of chlorite), anhydrite, clinozoisite (Ca end-member of epidote), pyrite, magnetite, sphalerite and galena.

The isoenthalpic reservoir boiling calculations from 350° through 250°C are conducted for the Fushime and Kakkonda case. Precipitation and dissolution of quartz, muscovite, clinochlore, anhydrite, pyrite, magnetite, sphalerite and galena are allowed, while albite, K-feldspar and clinozoisite are suppressed in the calculations. The isothermal reservoir boiling calculation with a stepwise increment of enthalpy at 300°C is tested only for the Kakkonda case. The liquid water fully equilibrated with the minerals mentioned above at 300°C is used as a starting solution. The precipitation and dissolution of clinochlore, anhydrite, pyrite, magnetite, sphalerite and galena are only allowed in the isothermal calculation. The redox reaction for H_2 is allowed in both of the isoenthalpic and isothermal models.

The fluids computed at 300^{o}C in the isoenthalpic models boil to 100^{o}C. Precipitation of scale minerals (sphalerite and galena, commonly observed in these fields) is only allowed, while other minerals are suppressed in the calculations. The redox reaction for H_2 is also disallowed for kinetic reasons. The same type of the calculations for the fluids computed at the enthalpies of 1,680 kJ/kg and 2,057 kJ/kg in the isothermal model are also carried out. The pH at 20^{o}C is calculated by cooling of the boiled waters (separated from gas phase) at 100^{o}C. Precipitation of any minerals is disallowed in the cooling calculations.

3.2 Results for the Fushime system

At the beginning of boiling, CO_2, H_2S and H_2 strongly partition into gas phase (Fig. 1a). The H_2S degassing drives dissolution of pyrite from wall rock and precipitation of magnetite (Fig. 1b). An increase in aqueous Fe^{2+} concentration is caused by the dissolution of pyrite (Fig. 1d), but most of Fe^{2+} dissolved from pyrite enters precipitating magnetite (reaction 1). At 270^{o}C, pyrite turns to precipitate, whereas magnetite dissolves. An increase in SO_4^{2-} concentration is caused by the combined effects of pyrite dissolution and H_2 redox reaction (reaction 2) at the early stage of boiling. However, dissolution of anhydrite supplies SO_4^{2-} at the later stage (Fig. 1b and 1d).

The continuous precipitation of sphalerite (reaction 3) and quartz is driven by temperature decrease. A small amount of galena also precipitates. The aqueous Mg^{2+} concentration increases with the dissolution of clinochlore, while the Al^{3+} concentration decreases with the precipitation of muscovite (Fig. 1b and 1d).

The water equilibrated with the principal alteration and scale minerals has a pH of 5.6 at 350^{o}C (Fig. 1c). The pH decreases to 5.1 at 300^{o}C and to 5.3 at 250^{o}C, as boiling and reactions proceed ("full reactions" in Fig. 1c). The pH decrease results from the excess supply of H^+ from the reactions 1, 2 and 3 over the H^+ consumption by CO_2 degassing (reaction 4). Therefore, the separate calculation in which all these reactions are disallowed shows the pH increase with decreasing temperatures ("w/o mineral and H_2 redox reac." in Fig. 1c). If sphalerite and galena are suppressed, a slight decrease in pH at high temperatures and a steep increase at low temperatures are caused by the reactions with pyrite and magnetite ("w/o sph & gal" in Fig. 1c). It is clearly demonstrated that the H^+ supply from sphalerite precipitation is the most effective cause of the pH decrease in the model.

$$\begin{array}{llllllll} 3FeS_2 & +6H_2O & \rightarrow & Fe_3O_4 & +0.5SO_4^{2-} & +5.5H_2S & +H^+ & (1). \\ \text{(pyrite)} & \text{(aq)} & & \text{(magnetite)} & \text{(aq)} & \text{(aq)} & \text{(aq)} & \end{array}$$

$$\begin{array}{llllll} H_2O+ & 0.25H_2S & \rightarrow & H_2 & +\ 0.25SO_4^{2-}+0.5H^+ & (2). \\ \text{(aq)} & \text{(aq)} & & \text{(gas)} & \text{(aq)} \quad\quad \text{(aq)} & \end{array}$$

$$\begin{array}{llllllll} ZnCl_2 & +\ H_2S & \rightarrow & ZnS & +\ 2H^+ & +\ 2Cl^- & & (3). \\ \text{(aq)} & \text{(aq)} & & \text{(sphalerite)} & \text{(aq)} & \text{(aq)} & & \end{array}$$

$$\begin{array}{llllll} HCO_3^- + & H^+ & \rightarrow & CO_2 & +\ H_2O & (4). \\ \text{(aq)} & \text{(aq)} & & \text{(gas)} & \text{(aq)} & \end{array}$$

The calculated pH at 20^{o}C is 3.3 when precipitation of sphalerite and galena during boiling from 300^{o}C to 100^{o}C is allowed (Fig. 2). If their precipitation is disallowed, a higher pH of 4.3 is obtained. These values are fairly close to the measured pH for the waters discharged from the Fushime wells (pH = 3.8-4.7). The calculated fluid composition also closely agrees with the observed compositions, except for the minor discrepancies in Fe^{2+} and Pb^{2+} concentrations.

3.3 Results for the Kakkonda system

The calculation results of isoenthalpic reservoir boiling for the Kakkonda case are shown in Fig. 3. The quantity of sphalerite precipitation from dilute Kakkonda type water is remarkably smaller than that from saline Fushime type water (Fig. 3b). Degassing of H_2S from aqueous phase drives dissolution of pyrite and precipitation of magnetite (reaction 1) from 350_{o} to 290^{o}C and its reverse reaction proceeds at low temperatures. The pH change during boiling is also different from the Fushime case (Fig. 3c). The pH decreases very steeply at higher temperatures, due to the combined effects of reactions 1 and 2 (from 6.4 at 350^{o}C to 5.3 at 320^{o}C). The reverse progress of reaction 1 causes the pH increase at lower temperatures ("full reactions" in Fig. 3c). Therefore, the separate calculation in which the reactions with pyrite and magnetite are disallowed causes a continuous pH decrease as boiling proceeds, owing to the H^+ supply from H_2 redox reaction ("w/o Fe minerals" in Fig. 3c). It is obvious that the pH of the dilute Kakkonda type water is scarcely influenced by the precipitation of sphalerite and galena because the quantity of their precipitation is small.

The calculated pH at 20^{o}C is on the range of the measured pH for the waters discharged from the Kakkonda wells (pH = 3.2-4.5). However, the calculated Fe^{2+} concentration is far apart from the observed concentrations (Fig. 4). The calculated concentrations of SO_4^{2-} and Al^{3+} are also lower than the observed concentrations. It is indicated that the isoenthalpic model cannot fully explain the processes occurred in the Kakkonda deep reservoir.

One possibility to explain the observed high concentrations of Fe^{2+} may be that isothermal boiling process in the reservoir leads to more vigorous degassing of H_2S from the liquid phase to cause the further dissolution of pyrite. Isothermal boiling results in continuous dissolution of pyrite and precipitation of magnetite from enthalpies of 1,345 kJ/kg (the enthalpy of the liquid water at 300^{o}C) through 2,057kJ/kg (Fig. 5b). The concentrations of H_2S gas and SO_4^{2-} increase with increasing enthalpies owing to the progress of reaction 1. The pH decrease is mainly due to the reactions with pyrite and magnetite, though some contribution of H_2 redox reaction can be seen in Fig. 5c. The concentrations of Fe^{2+}, Mg^{2+}, Al^{3+}, Zn^{2+} and Pb^{2+} increase owing to the dissolution of pyrite, clinochlore, sphalerite and galena. A small amount of anhydrite precipitation scarcely influences on the Ca^{2+} concentration (Fig. 5b and 5d).

The calculated pH at 20^{o}C for the fluid computed at the enthalpy of 1,680 kJ/kg in the isothermal model is 3.4, and it is on the range of the measured pH (from 3.2 to 4.5). The calculated composition closely agrees with the observed compositions, but the calculated Fe^{2+} concentration is still lower than the observed concentrations (Fig. 4). The fluid computed at the enthalpy of 2,057 kJ/kg contains much higher Fe^{2+}, being similar to the observed concentrations. However, the calculated pH at 20°C (2.9) is lower than the measured pH. In addition, the small disagreements with respect to Mg^{2+} and Al^{3+} concentrations can be seen in Fig. 4.

4. DISCUSSION

The fluid composition calculated through the isoenthalpic model for the Fushime type water is very similar to those of the fluids discharged from the wells, except for the minor disagreements in Fe^{2+} and Pb^{2+}. The reason for the disagreements is not clear, but it is possible that the isothermal boiling process reconciles the disagreement in Fe^{2+} as we describe in the Kakkonda case. It is also possible that the equilibrium with magnetite and galena is not fully achieved in the real system. Another possibility is that the waters discharged from wells may be enriched in Fe^{2+} by dissolution from steel equipment.

The calculation results indicate that the H^+ supply from sphalerite precipitation is the most probable cause to form acid waters in the Fushime system, as already demonstrated by Akaku et al., (1991, 1997). The H^+ supply from dissolution of pyrite and precipitation of magnetite occurred at high temperatures is also able to contribute to the acidity, but it is less effective than the sphalerite precipitation in the isoenthalpic boiling model.

The isoenthalpic model for the Kakkonda case forms acid water only at high temperatures, and fails to explain the observed high concentrations of Fe^+. In contrast, the isothermal boiling model shows the continuous pH decrease and the Fe^+ increase with increasing enthalpies. The calculated composition through the isothermal model fit the observed compositions more closely. This may suggest that the isothermal model is more appropriate for the Kakkonda case. The both models indicate that dissolution of pyrite and precipitation of magnetite driven by H_2S degassing is effective to form acid water. In addition, the H^+ supply from H_2 redox reaction is able to cause the pH decrease.

The possibility that acidic volatiles such as HCl and SO_2 contribute to form acid water discharged from the Fushime and Kakkonda wells was discussed in Reed (1991), Yanagiya *et al.* (1996) and Akaku *et al.*, (1997). However, the contribution of acidic volatiles is not necessary to explain the observed acidity in our models.

5. CONCLUSIONS

The model calculations indicate that temperature decrease and degassing caused by production-induced boiling in reservoir drive the reactions of fluid with minerals in wall rocks. The calculated compositions through the models closely agree with those of the fluids discharged from the high temperature reservoirs in the Fushime and Kakkonda geothermal systems. The models are able to explain the observed acidity, without taking account of acidic volatiles such as HCl and SO_2. The H^+ supply from sphalerite precipitation is the most probable cause of the observed acidity in the saline Fushime waters. On the other hand, the H^+ supply from dissolution of pyrite and precipitation of magnetite and from the H_2 redox reaction possibly contributes to the observed acidity in the dilute Kakkonda waters.

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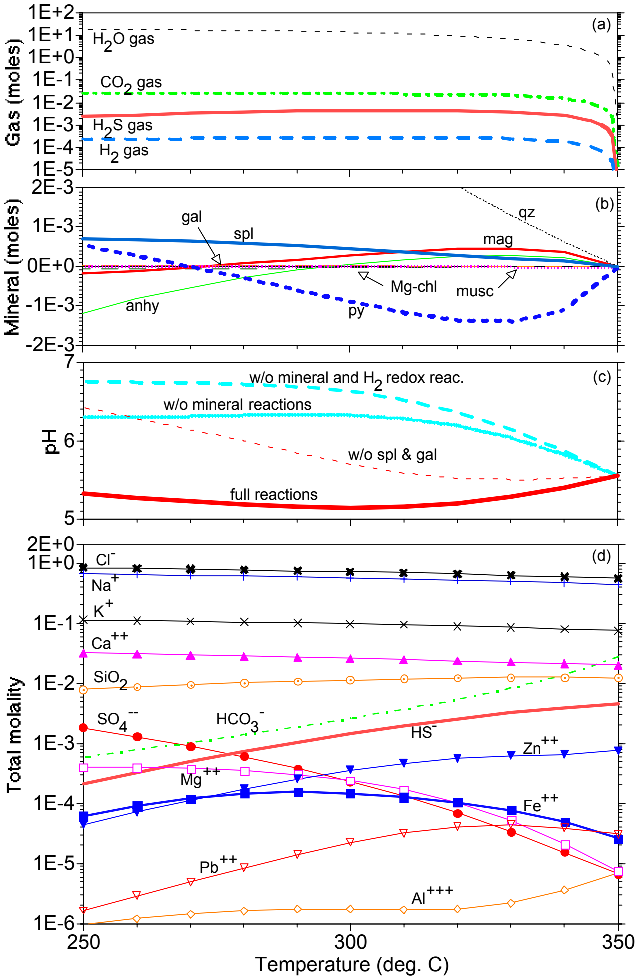

Figure 1. Numerical reservoir boiling of the Fushime type water. Isoenthalpic boiling proceeds from the right to left side of the graphs. The starting solution (1kg) is fully equilibrated with the alteration and scale minerals (quartz, albite, K-feldspar, muscovite, clinochlore, anhydrite, clinozoisite, pyrite, magnetite, sphalerite and galena) at 350°C.

(a) Gas phase composition. (b) Mineral precipitates and dissolves. (c) pH. (d) Total aqueous molality of component species.

Mineral abbreviations: qz, quartz; musc, muscovite; py, pyrite; Mg-chl, clinochlore (Mg end member of chlorite); anhy, anhydrite; spl, sphalerite; gal, galena; mag, magnetite.

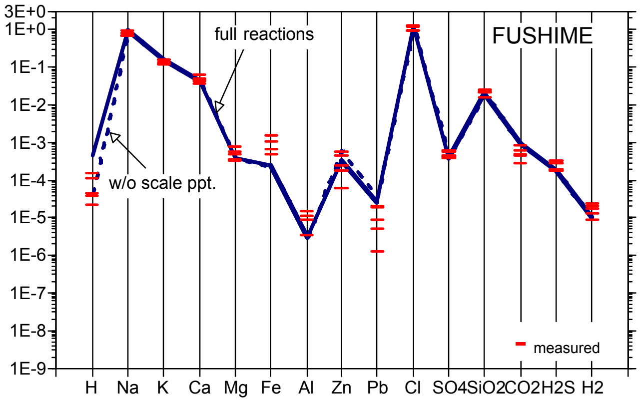

Figure 2. Comparison between calculated and measured compositions for the Fushime case. Dashed line indicates the composition when scale precipitation (sphalerite and galena) is disallowed. Activity of H^+ (10^{-pH}) at 20°C, aqueous total molality of Na^+, K^+, Ca^{2+}, Mg^{2+}, Fe^{2+}, Al^{3+}, Zn^{2+}, Pb^{2+}, Cl^-, SO_4^{2-} and SiO_2 at 100°C and mole fraction of CO_2, H_2S and H_2 in gas phase at 100°C.

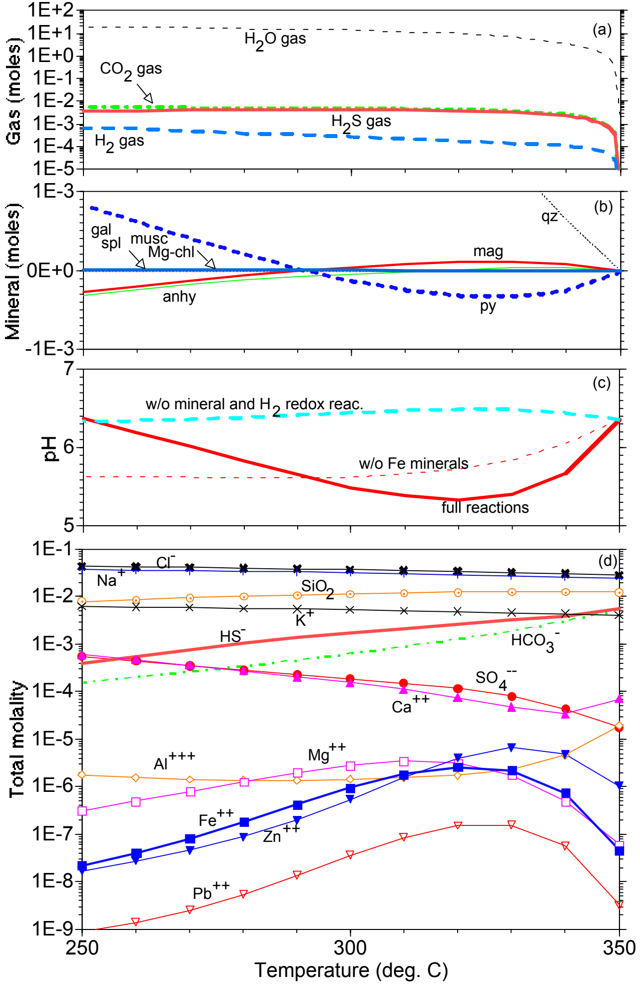

Figure 3 Isoenthalpic boiling of the Kakkonda type water. Boiling proceeds from the right to left side of the graphs. See caption to Fig. 1 for abbreviations.

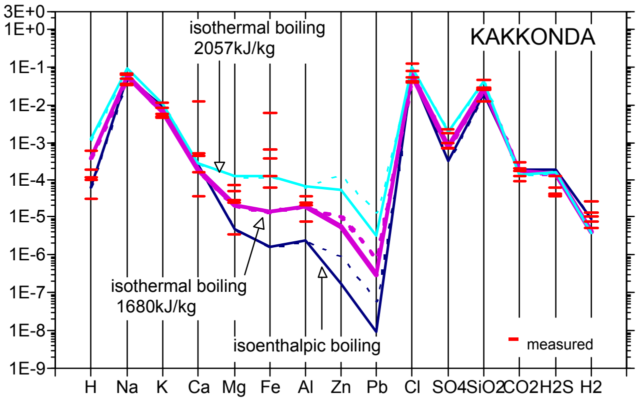

Figure 4. Comparison between calculated and measured compositions for the Kakkonda case. See caption to Fig. 2 for abbreviations. Dashed lines indicate the compositions when scale precipitation (sphalerite and galena) is disallowed.

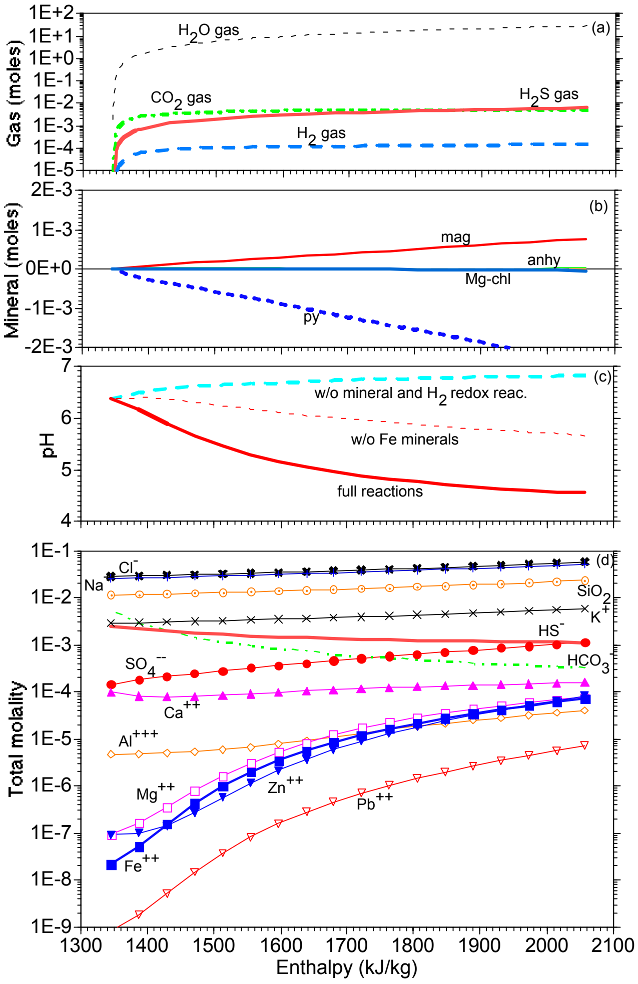

Figure 5. Isothermal boiling of the Kakkonda type water. Boiling proceeds from the left to right side of the graphs. The starting solution (1kg) is fully equilibrated with the alteration and scale minerals at 300°C.