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ANALYSIS OF NEUTRALIZATION REACTION IN A GEOTHERMAL WELL

Naoyuki Akazawa and Ryuichi Itoi

Dept. of Earth Resources Eng. Graduate School of Eng. Kyushu University
744 Motooka Nishi-ku Fukuoka 819-0394 Japan
e-mail: akazawa-n@mine.kyushu-u.ac.jp

ABSTRACT

Geothermal wells producing acidic fluid have been abandoned because of high corrosion potential on casing and surface pipeline even though their high productivity. Injecting alkaline solution into the wellbore at a depth of feedzone can be an effective method for neutralizing acid fluid. This study aims to develop a numerical wellbore model for simulating steam-water two-phase flow and equilibrium chemical reactions. The model consists of two-phase flow incorporated with mass action and mass balance equations with respect to chemical components.

The model was first verified by analyzing chemical data of the discharged fluid and steam-water flowrates measured at the wellhead of Well N13-SZ-4, and chemical conditions at the feedzone was estimated. Then, the effects of injecting sodium hydroxide (NaOH) solution quantitatively on neutralization of fluid and pH profiles along the wellbore were analyzed with the model.

INTRODUCTION

Producing acidic fluid may cause corrosion of well casing and surface pipeline, and consequently it damage wells and may lead to blowout of pipeline in the worst case. In order to make a use of the well producing acidic fluid, casing material need to be of unti-corrosion nature. However, acid fluid production is hard to predict as it is closely related to subsurface geological and geochemical conditions. Once the well started production, it is very difficult to replace casing with unti-corrosive one. For these reasons, they have been abandoned or converted to injection wells in spite of their sufficient productivity.

In the Miravalles geothermal field, Costa Rica, there are geothermal wells producing acidic fluid. For these wells, an acid neutralization system has been introduced and operated successfully (Moya and Sanchez 2002). This system employs injection of NaOH (NaOH) solution with tubing at a specified depth of wells. This system has enabled two wells, PGM-7 and PGM-19, to supply steam for power generation economically since October 2001 and February 2000, respectively.

In spite of these advantages, this system has some problems in practical operation. For example, deposition of minerals such as anhydrite ($CaSO_4$) may occur in the wellbore when neutralized geothermal water is oversaturated with respect this mineral. Plugging of wellbore leads to decrease of fluid production, then workover for cleaning well will be required by shutting the wellbore. Furthermore, insufficient neutralization caused by improper amount or concentration of NaOH solution will be resulted in damages of casing.

In this study, we have developed a wellbore flow model coupled with chemical equilibrium reaction. Measured data for steam and water flow rate as well as chemical analysis of fluid from Well N13-SZ-4, the Shiramizugoe geothermal field, was analyzed with the model. Then, effects of NaOH concentration and its injection amount on neutralization of the fluid at feedzone of the well have been evaluated. Profiles of temperature and pressure in the wellbore as well as chemical components were also analyzed.

WELLBORE MODEL

Figure 1 illustrates the neutralization system in the acidic wells. The model consists of two parts. Firstly, wellbore model simulate the vertical steam and water two phase flow in a vertical well. It can analyze pressure and temperature profiles and volume fraction of steam and hot water. Secondly, chemical model simulate the chemical reactions and concentration of the chemical species dissolved in geothermal fluid on the basis of equilibrium reactions. Analysis of neutralization reaction is also included. It is assumed that NaOH solution is injected at a depth of feed point through injection pipe or chubing. This model is based on the mass balance and mass action equations with respect to chemical species and also includes temperature effects on these equations.

By combining these two models, a developed model can be used to analyze a pH profile along wellbore as well as to evaluate saturation degree with respect to minerals.

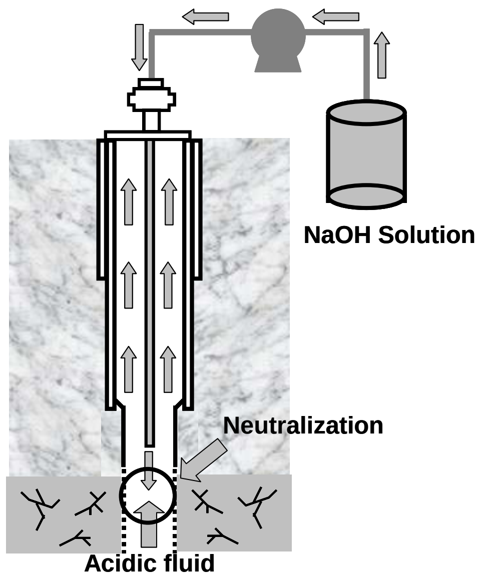

Fig.1: Neutralization system

NUMERICAL MODEL

Component species and complex species

In order to calculate the pH distribution in a well, it is important to estimate the variety of chemical species and their concentration in geothermal fluid. Geochemical models are available such as WATCH for geochemical calculation. However, we developed a chemical model by selecting main and important chemical species in the water for component and complex species, then developed a chemical model for simplicity.

Table 1 shows the classification of chemical species of Well N13-SZ-4.

Table 1: Classification of chemical species
$^{\circ}$ represents hydrate

Component Species	Complex Species
H^+	OH^-
Cl^-	$NaCl^{\circ}$ KCl°
SO_4^{2-}	$H_2SO_4^{\circ}$ HSO_4^-
$H_4SiO_4^{\circ}$	$H_3SiO_4^-$ $H_2SiO_4^{2-}$
	$NaHSiO_4^{\circ}$
Na^+	$NaSO_4^-$
K^+	KSO_4^-
Ca^{2+}	$CaSO_4^{\circ}$ $CaOH^+$
Mg^{2+}	$MgSO_4^{\circ}$ $MgOH^+$
Fe^{2+}	$Fe(OH)_2^{\circ}$ $FeOH^+$
Al^{3+}	$Al(OH)_4^-$ $Al(OH)_3^{\circ}$
	$Al(OH)_2^+$ $AlOH^{2+}$

Component species consist of the minimal kind of chemical species which represent all the chemical species in the geothermal fluid with mass action and mass balance equations. Complex species consist of the chemical species which are calculated with concentration of component species and equilibrium constants. As shown in Table 1, 10 component species and 20 complex species are supposed to exist in the geothermal fluid. Concentration of each chemical species depends on each chemical reaction and equilibrium constants.

Then, concentration of chemical species were calculated with mass action and mass equations.

Chemical reactions

Table 1 summarizes 30 kinds of chemical species that are subject to be included in the model. For these chemical species, 20 chemical reactions shown in Table 2 are considered in this study.

Table 2: Chemical reaction in geothermal fluid

Chemical species	Reaction
1. H^+	$H_2O \Leftrightarrow H^+ + OH^-$
2. NaCl	$NaCl^{\circ} \Leftrightarrow Na^+ + Cl^-$
3. KCl	$KCl^{\circ} \Leftrightarrow K^+ + Cl^-$
4. H_2SO_4	$H_2SO_4^{\circ} \Leftrightarrow H^+ + HSO_4^-$
5. HSO_4^-	$HSO_4^- \Leftrightarrow H^+ + SO_4^{2-}$
6. $CaSO_4$	$CaSO_4^{\circ} \Leftrightarrow Ca^{2+} + SO_4^{2-}$
7. $CaOH^+$	$CaOH^+ \Leftrightarrow Ca^{2+} + OH^-$
8. $MgSO_4$	$MgSO_4^{\circ} \Leftrightarrow Mg^{2+} + SO_4^{2-}$
9. $MgOH^+$	$MgOH^+ \Leftrightarrow Mg^{2+} + OH^-$
10. $NaSO_4^-$	$NaSO_4^- \Leftrightarrow Na^+ + SO_4^{2-}$
11. KSO_4^-	$KSO_4^- \Leftrightarrow K^+ + SO_4^{2-}$
12. H_4SiO_4	$H_4SiO_4^{\circ} \Leftrightarrow H^+ + H_3SiO_4^-$
13. NaH_3SiO_4	$NaH_3SiO_4 \Leftrightarrow Na^+ + H_3SiO_4^-$
14. $H_3SiO_4^-$	$H_3SiO_4^- \Leftrightarrow H^+ + H_2SiO_4^-$
15. $Fe(OH)_2$	$Fe(OH)_2^{\circ} \Leftrightarrow FeOH^+ + OH^-$

Chemical species	Reaction
16. $FeOH^{+}$	$FeOH^{+} \Leftrightarrow Fe^{2+} + OH^{-}$
17. $Al(OH)_4^{-}$	$Al(OH)_4^{-} \Leftrightarrow Al(OH)_3 + OH^{-}$
18. $Al(OH)_3$	$Al(OH)_3^{\circ} \Leftrightarrow Al(OH)_2^{+} + OH^{-}$
19. $Al(OH)_2^{+}$	$Al(OH)_2^{+} \Leftrightarrow AlOH^{2+} + OH^{-}$
20. $AlOH^{2+}$	$AlOH^{2+} \Leftrightarrow Al^{3+} + OH^{-}$

Equilibrium constant

Equilibrium constant for chemical reactions which occur in geothermal fluid depends on temperature. The relationship between the equilibrium constant and temperature by Arnorsson and Sigurdsson (1982) was used in this study.

Mass action and mass balance equations

The activities of complex species can be estimated with the mass action law. This law defines the relationship between activity and equilibrium constant. Table 3 presents the mass action equations for each chemical reaction where $K_i(-)$ is the equilibrium constant of 'i' chemical species.

Table 3: Mass action equation

Chemical Species	Mass action equation
1. OH^{-}	$K_{H_2O} = a_{H^+} a_{OH^-}$
2. NaCl	$K_{NaCl} = \frac{a_{Na^+} a_{Cl^-}}{a_{NaCl}}$
3. KCl	$K_{KCl} = \frac{a_{K^+} a_{Cl^-}}{a_{KCl}}$
4. H_2SO_4	$K_{H_2SO_4} = \frac{a_{H^+} a_{HSO_4^-}}{a_{H_2SO_4}}$
5. HSO_4^{-}	$K_{HSO_4^-} = \frac{a_{H^+} a_{SO_4^{2-}}}{a_{HSO_4^-}}$
6. $CaSO_4$	$K_{CaSO_4} = \frac{a_{Ca^{2+}} a_{So_4^{2-}}}{a_{CaSO_4}}$
7. $CaOH^{+}$	$K_{CaOH^+} = \frac{a_{Ca^{2+}} a_{OH^-}}{a_{CaOH}}$
8. $MgSO_4$	$K_{MgSO_4} = \frac{a_{Mg} a_{SO_4^{2-}}}{a_{MgSO_4}}$
9. $MgOH^{+}$	$K_{MgOH^+} = \frac{a_{Mg^{2+}} a_{OH^-}}{a_{MgOH}}$
10. $NaSO_4^{-}$	$K_{NaSO_4^-} = \frac{a_{Na^+} a_{SO_4^{2-}}}{a_{NaSO_4^-}}$
11. KSO_4^{-}	$K_{KSO_4^-} = \frac{a_{K^+} a_{SO_4^-}}{a_{KSO_4^-}}$
12. H_4SiO_4	$K_{H_4SiO_4} = \frac{a_{H^+} a_{H_3SiO_4^-}}{a_{H_4SiO_4}}$
13. NaH_3SiO_4	$K_{NaH_3SiO_4} = \frac{a_{Na^+} a_{H_3SiO_4}}{a_{NaH_3SiO_4}}$
14. $H_3SiO_4^{-}$	$K_{H_3SiO_4^-} = \frac{a_{H+} a_{H2SiO4-}}{a_{H3SiO4}}$
15. $Fe(OH)_2$	$K_{Fe(OH)_2} = \frac{a_{Fe^{2+}} a_{FeOH^+}}{a_{Fe(OH)_2}}$
16. $FeOH^{+}$	$K_{FeOH^+} = \frac{a_{Fe^{2+}} a_{OH^-}}{a_{FeOH}}$
17. $Al(OH)_4^{-}$	$K_{Al(OH)_4^-} = \frac{a_{Al(OH)_3} a_{OH^-}}{a_{Al(OH)4-}}$
18. $Al(OH)_3$	$K_{Al(OH)_3} = \frac{a_{Al(OH)^{2+}} a_{OH^-}}{a_{Al(OH)_3}}$

Chemical species	Mass action equation
19. $Al(OH)_2^+$	$K_{Al(OH)_2^+} = \dfrac{a_{AlOH^{2+}} a_{OH^-}}{a_{Al(OH)_2^+}}$
20. $AlOH^{2+}$	$K_{AlOH^{2+}} = \dfrac{a_{Al^{3+}} a_{OH^-}}{a_{AlOH^{2+}}}$

On the other hand, mass balance equations express that total concentration of a chemical species is equivalent to the analysis value of its concentration. A mass balance equation corresponds to one component species shown in Table 1. Equations (1) through (11) represent the mass balance equation for each component species.

$$\sum H^+ = [H^+] + [HSO_4^-] + [H_2SO_4] + [H_3SiO_4^-] + [H_4SiO_4] \quad (1)$$

$$\sum Cl^- = [KCl] + [NaCl] + [Cl^-] \quad (2)$$

$$\sum SO_4^{2-} = [H_2SO_4] + [HSO_4^-] + [SO_4^{2-}] \quad (3)$$

$$\sum SiO_2 = [H_4SiO_4] + [H_3SiO_4^-] + [NaH_3SiO_4] + [H_2SiO_4^{2-}] \quad (4)$$

$$\sum Na^+ = [Na^+] + [NaSO_4^-] + [NaCl] + [NaH_3SiO_4] \quad (5)$$

$$\sum K^+ = [K^+] + [KSO_4^-] + [KCl] \quad (6)$$

$$\sum Ca^{2+} = [Ca^{2+}] + [CaSO_4] + [CaOH] \quad (7)$$

$$\sum Mg^{2+} = [Mg^{2+}] + [MgSO_4] + [MgOH] \quad (8)$$

$$\sum Fe^{2+} = [Fe^{2+}] + [FeOH^+] + [Fe(OH)_2] \quad (9)$$

$$\sum Al^{3+} = [Al^{3+}] + [AlOH^{2+}] + [Al(OH)_2^+] + [Al(OH)_3] \quad (10)$$

where, [X] represents the concentration of the chemical species of X in mol/l.

As mentioned above, a mass balance equation corresponds to one component species, and a mass action equation corresponds to one complex species.

Hence, regarding the concentration and activity of each chemical species as the variables, the number of variable will be the same as that of equations. Consequently, it is possible to calculate the concentration and activities of chemical species shown in Table 1 if analysis values of them and each equilibrium constant are obtained. Chemical analysis data have obtained by the sampling of geothermal fluid and, the values of equilibrium constants can be also obtained by Arnorsson and Sigurdsson (1982). Therefore, combined equations of mass balance and reactions leads to a set of non linear equation that is numerically solved by applying Newton-Raphson mothod and calculate the concentration of chemical species at each temperature in a well. As a result, pH profile in a well can be obtained with the activity of H^+ and the equation below.

$$pH = -\log a_{H^+} \quad (11)$$

Neutralization reaction

Acidic fluid is to be neutralized by the injection of NaOH solution at feed zone depth.

It is assumed that a hydroxide ion (OH^-) reacts with a hydrogen ion (H^+). Thus injection of NaOH solution into acidic fluid decreases the concentration of hydroxide ion and pH value would rise.

Figure2 illustrates the neutralization scheme of acidic fluid by adding NaOH solution.

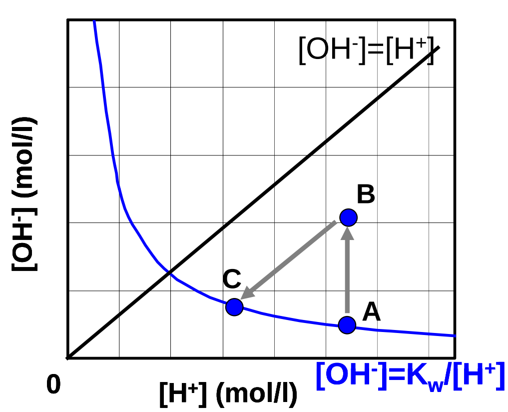

Fig.2: Neutralization reaction model

The procedure from A to B in Fig.2 represents an increase in hydroxide ion due to injection of NaOH solution. And that from B to C represents the neutralization reaction. In this procedure, a hydroxide ion reacts with a hydrogen ion as the reaction shown below.

$$H^{+}+OH^{-}\Leftrightarrow H_2O \qquad (12)$$

Therefore, these ions decrease concentration from B to C. The point C is on the hyperbolic line ($[OH^{-}]=K_{H2O}/[H^{+}]$) This is because the relationship between hydroxide and hydrogen ion in the solution is always represented with Eq.(12).In this way, neutralization reaction proceeds from point A to point C in the Fig 2.

Saturation index

In a geothermal well producing the acidic fluid, decrease in temperature of geothermal fluid or increase in pH value can causes the precipitation of anhydrite into a well. Therefore, it is important to estimate it. Mineral precipitation can be estimated by calculating Saturation Index (S.I.). In case of anhydrite, S.I. is shown below (Chiba, 1990).

$$\log(S.I.) = \log\left(\frac{a_{Ca^{2+}} \cdot a_{SO_4^{2-}}}{K_{CaSO_4}}\right) \qquad (13)$$

Positive log(S.I.) indicates over saturated with respect to anhydrite. Possibility of anhydrite precipitation was examined by calculating S.I along a wellbore.

Numerical model for two phase flow analysis

As for analysis of two phase flow in a well, it is necessary to estimate the pressure, temperature and void ratio of fluid in a well, to calculate the pH profile. In this study, a numerical wellbore model for two phase flow by Akazawa et.al (2007) is used. This model is constructed on the basis of the assumptions below.

(1) Iso-enthalpy flow is assumed
(2) The effect of non-condensable gas is negligible
(3) The well is vertical

In this model, two phase flow is divided into 3types depending on the void ratio. First, the flow existing small bubbles in liquid phase is defined as Bubble flow. Second, the flow existing large bubble like a bullet in liquid phase is defined as Slug flow. Finally, the flow existing liquid phase like a mist in steam phase is defined as Mist flow. For these flow regimes, the pressure drop in a well is calculated with different equations. With this method, pressure and temperature profile in a well is calculated.

The input data for this model are the wellhead pressure, mass flux, enthalpy, depth at feed point and casing profile. With these data, it can simulate the temperature and void ratio at any depth in a well.

Construction of numerical model

By combining the numerical model for calculating the concentration of chemical species in the geothermal fluid with the model for two phases flow analysis, we can calculate the concentration of the chemical species and pH value in a well.

Figure3 illustrates the flow chart of the numerical analysis. At first, input the wellhead pressure, mass flux, total enthalpy, depth at feed point, casing profile, and concentration of chemical species at well head. Then, calculate the temperature profile in a well.

Secondly, to simulate the pH profile, calculate the concentration of chemical species in the geothermal fluid in a well. Consequently, calculate the pH value with Eq.(11). Next, to simulate the neutralization reaction with NaOH injection, input the concentration and mass flux of NaOHsolution.

Finally, simulate the neutralized pH profile

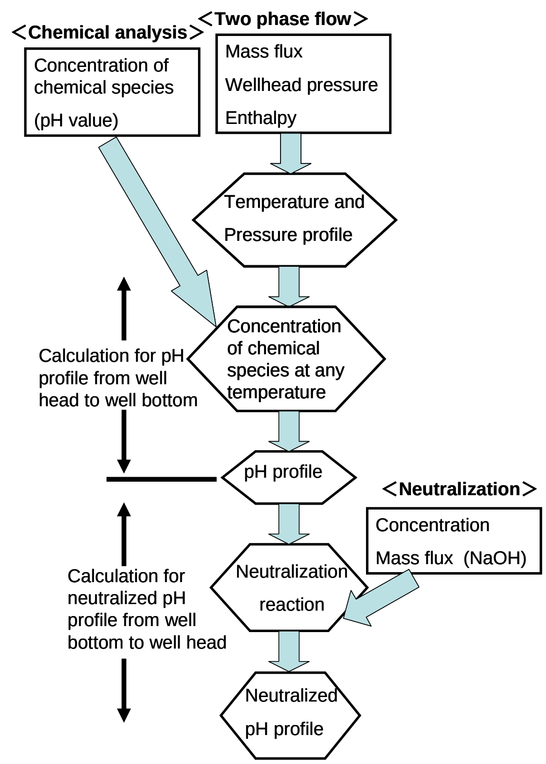

Fig.3: Flow chart of analysis

CALCULATION CONDITIONS

Well data

Well N13-SZ-4 was drilled in Shiramizugoe geothermal area in Japan. Table4 summarizes information of the well and Fig.4 illustrates the casing program.

Table4: Property of Well N13-SZ-4

Wellhead pressure (MPa)	1.45
Mass flux (t/h)	116
Enthalpy (kJ/kg)	1029
Feed point depth (m)	1758
Wellbottom depth (m)	2006

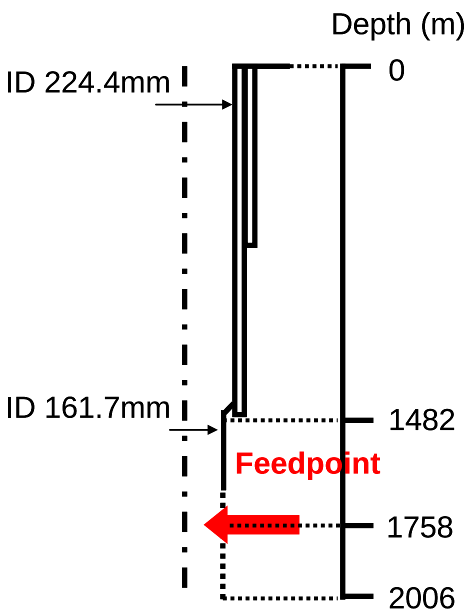

Fig.4: Casing program of Well N13-SZ-4

Table5: Chemical analysis data of Well N13-SZ-4l

Chemical species	Concentration	
	mg/l	mmol/l
H	2.51	2.51(pH=2.6)
Na	903	39.3
K	180	4.62
Li	5.61	0.81
Ca	8.41	0.841
Mg	15.0	0.625
Cl	1380	38.9
SO_4	625	6.51
HCO_3	N.D.	N.D.
T-SiO_2	961	16.0
Fe	44.4	0.794
Al	0.31	0.0115
H_2S	N.D	N.D.
T-CO_2	119	2.70

Feed zone of the well locates at a depth 1758 m, where acidic fluid is supposed to flow into the well. Table5 shows the chemical analysis data of the discharged fluid of the well. Fluid has low pH of 2.6 and is of sulfuric acidity. This acidity seems to be originated from sulfuric acid in the fluid. High concentration of chloride and sodium indicate that fluid is of NaCl type.

Procedure for the analysis

In order to calculate the pH profile and estimate the effect of the injection of NaOH solution, at first, input the property and chemical data into the numerical model. With this data, simulate the pH profile in a well.

Second, input the concentration and mass flux of NaOH solution. In this procedure, change the concentration and mass flux and estimate the neutralization reaction in each case.

Hirowatari (1992) pointed out that the corrosion of casing can be negligible when the pH value of the geothermal fluid is around 5.0. Hence, if the pH value rises above 5.0 in a well, regard the neutralization reaction as sufficient to prevent the corrosion.

Calculation conditions

In order to quantitatively evaluate the validity of neutralization operation, the base case was calculated by giving the parameter values shown in Table 6.

For parameter studies, concentration and mass of NaOH solution, and fluid temperature are varied as shown in Table 7.

Table 6: Parameter for base Case

Base Case	
Diameter of injection pipe	6.0(cm)
Mass flux of NaOH solution	0.2(kg/s)
Concentration of NaOH solution	0.01(mol/l)
Temp. of geothermal fluid	238(℃)
Mass flux of geothermal fluid	32(kg/s)

Table 7: Parameter for case A,B and C

Case A / Concentration of NaOH solution (mol/l)
0.005 0.0075 0.01 0.02
Case B / Mass flux of NaOH solution (kg/s)
0.1 0.15 0.2 0.25
Case C / Temp. of geothermal fluid (℃)
238 230 225

The concentration of NaOH solution is varied from 0.005 to 0.02 mol/l, the mass flux of NaOH solution from 0.1 to 0.25kg/s, and the temperature of geothermal fluid at the well bottom from 225 to 238℃

RESULTS OF ANALYSIS

Temperature and pH profiles

Figure 5 illustrates the simulated temperature and pH profiles of Well N13-SZ-4. The geothermal fluid flows into the well as the water single phase, whose temperature is about 238℃. Then, it starts to flash about 500m depth. Above the flashing point, void ratio of steam increases as the fluid flows upward to wellhead.

pH value indicates about 4.3 at well bottom and, it is constant from bottom to flash point. Above the flashing point, pH value starts to decrease as the volume of steam increase. This is because the chemical species remain in hot water phase when the geothermal fluid starts flashing and the concentration of hydrogen ion increases. As a result, pH value indicates 3.1 at the well head.

Finally, this profile shows that flashing and decrease in temperature cause the rise in pH value and acidity of the fluid at well head is higher than at the well bottom.

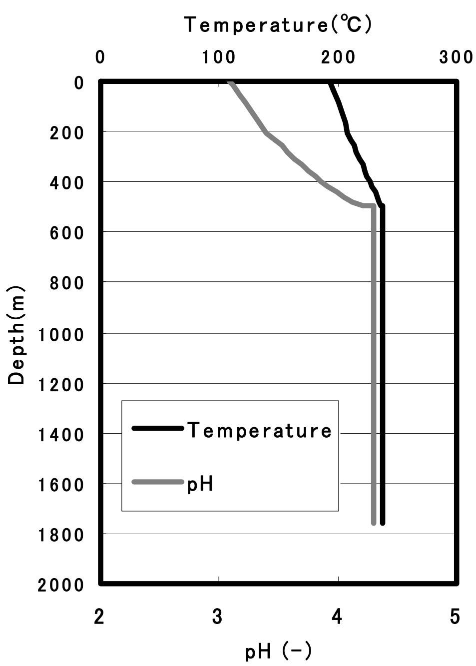

Fig.5:Simulated temperature and pH profiles in Well N13-SZ-4

The effect of neutralization reaction

Case A

Figure 6 illustrates the pH profiles for different concentration of NaOH solution with that of Figure 5.When the concentration of NaOH solution is given as 0.005, 0.0075, 0.01, 0.02 mol/l, pH value of geothermal fluid rise to 4.8, 5.8, 6.4, 7.1 at the well bottom, and 3.4, 4.4, 5.3, 6.1 at the well head respectively.

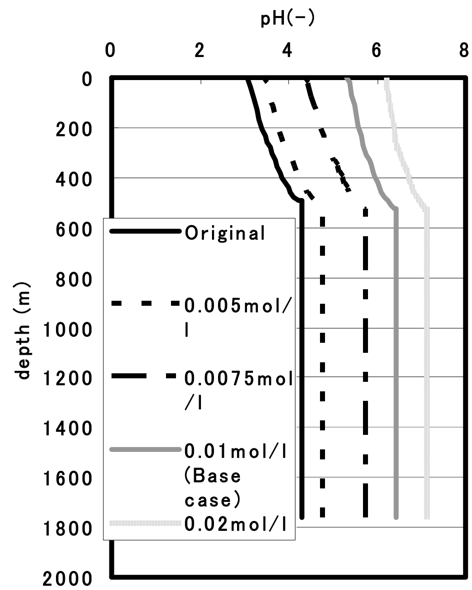

Fig.6:pH profile (Case A)

Case B

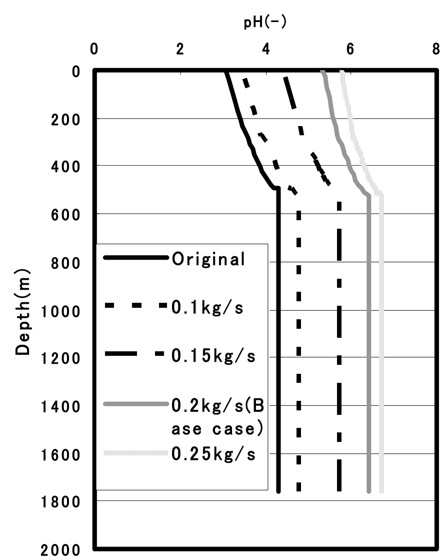

Fig.7:pH profile (Case B)

As a result, NaOH injection at the well bottom neutralizes the acidic geothermal fluid effectively. This result suggests that NaOH solution should be injected at higher concentration than 0.01 mol/l to avoid the risk of corrosion of casing.

Figure 7 illustrates the pH profiles of Case B and original profile.

When the mass flux of NaOH is 0.1, 0.15, 0.2, 0.25 kg/s, pH value of geothermal fluid at well head is elevated to 3.4, 4.4, 5.3, 5.7 respectively. As well as Case A, pH value at the well head indicates the minimum value in the well for each profile. That's because the decrease in temperature of geothermal fluid leads to increase in concentration of hydrogen ion.

Figure 8 illustrates the relationship between pH value at well head and concentration of NaOH.

Judging from the figure, in order to satisfy the criteria for avoiding any corrosion problem in the well, mass flux of NaOH solution should be more than 0.2 kg/s for this well.

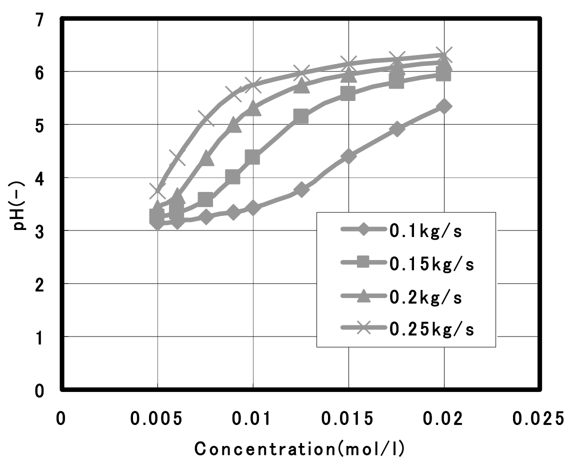

Fig.8: Relationship between pH at well head and concentration of NaOH solution (Case B)

Case C

Figure 9 illustrates the pH profiles of Case C. The effects of the geothermal fluid temperature at the well bottom on neutralization are examined.

When the temperature of geothermal fluid at well bottom is 238, 230, 225℃, pH value of well head indicates 5.3, 4.7, 4.1, respectively.

Figure 10 illustrates the relationship between pH value at the well head and concentration of NaOH solution for case C. If the temperature of geothermal fluid is low, the high concentration of NaOH solution is needed. From the results shown in Figure 9 and 10, 5℃ temperature drop requires 13% increase in concentration of NaOH solution in order to neutralize the geothermal fluid.

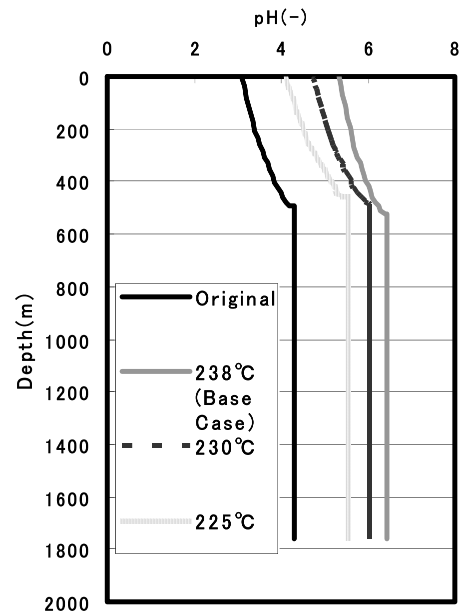

Fig.9: pH profile (Case C)

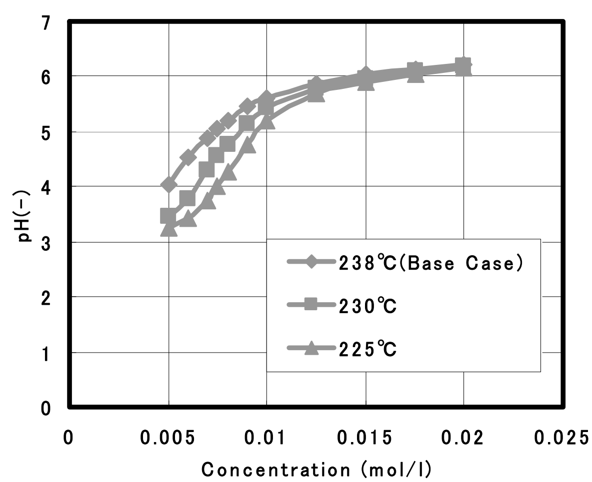

Fig.10: Relationship between pH at well head and concentration of NaOH solution (Case B)

Evaluation of S.I of anhydrite

The relationship between saturation index of anhydrite and depth are calculated to examine the precipitation of anhydrite in the well for Cases A, B and C. As shown in Figure 11, S.I remains constant in the water single flow. Figure 11 illustrate the S.I profiles of anhydrite for Case A. Other two cases show similar results. This is because equilibrium constant of anhydrite and activities of calcium and sulfate ion are constant under constant temperature. They also show that in the two phase flow, S.I. increases as the temperature decreases. This is because increase in vapor fraction causes the increase in concentration of chemical species resulted in a large activity product. Moreover, equilibrium constant of anhydrite increases with a decrease of temperature. These two reasons come an increase of S.I. with respect to anhydrite. Values of S.I. along the wellbore, however stays negative implying no possibility of anhydrite deposition.

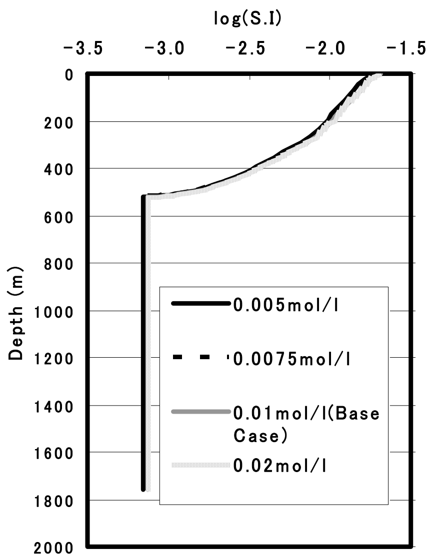

Fig.11: S.I profile of anhydrite (Case A)

CONCLUSIONS

A numerical wellbore model for simulating steam-water two phase flow coupled with chemical reaction model for the neutralization reaction in a geothermal well with NaOH solution was developed. The results are as follow,

1. The numerical model can simulate pH profile in a well which produces acidic fluid.
2. Conditions of NaOH solution injection at feed zone depth for neutralizing acidic fluid were quantitatively evaluated with the model for Well N13-SZ-4.
3. Saturation index with respect to anhydrite was calculated to be below 0 along the aellbore implying no precipitation in the well.

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