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EVALUATION OF NaOH AND H_2SO_4 TREATMENT IN pH-MODIFICATION OF FLUIDS USING SOLVEQ PROGRAM (SOLVEQ-pH METHOD)

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ABSTRACT

A method (SOLVEQ-pH) was developed using SOLVEQ in evaluating the amount of base (NaOH) or acid (H_2SO_4) added to thermal fluids. In this method, pH is changed, as the charge balance is re-adjusted to compensate for the given pH. It is the adjustment of the charge balance, using a pre-selected ion, that makes this method useful in computing amount of a particular base (such as NaOH) or acid (such as HCl or H_2SO_4).

For addition of base, Na ion (assuming addition as NaOH = Na^+ + OH^-) is added as the charge balancer while for acid addition, Cl^- is added as the balancer (assuming addition as HCl = H^+ + Cl^-). For acid like H_2SO_4, one can also use Cl ion as the balancer, provided its adjusted mole is divided by two by the concept neutralization-equivalent-weight or simply the equivalent of H_2SO_4 to HCl: there are two equivalents per mole of H_2SO_4 per mole of HCl.

Based from the results, SOLVEQ-pH can evaluate not only the needed dosage of acid or base treatment, but also show the intricacy of the chemical equilibrium involved in the procedure by comparing the computed from actual data. To-date, although measurements of pH's at laboratory condition remain the only method of choice in evaluating required dosages, SOLVEQ-pH strengthens our understanding of the real conditions that affect the treatment: for NaOH, fluxes in condensate flow and the role of Eh are essential parameters to monitor effectiveness of the treatment, while for H_2SO_4, the line-temperature dictates the strength of acidity, hence the amount of dosage.

1.0 INTRODUCTION

Through the years, pH modification of geothermal waste fluids from fluid-collection-disposal-system has become a valuable industrial procedure in altering the fluid's chemical properties to attain a desired effect, such as corrosion control or prevent silica precipitation. Adjusting the pH of steam condensate by base addition (such as NaOH) to 8.00 (Lichti *et al.*, 1998; Villa *et al.*, 2003), acid corrosion is controlled, while lowering the pH of the brine by acid injection (such as H_2SO_4) to 5.50 (Garcia *et al.*, 1996), prevents deposition of amorphous silica. Although these procedures may sound like simple mixing of fluids, its application to industrial scale was proven to be a not so easy task.

The critical step in its implementation is the evaluation of the optimum dosing rate to attain a desired pH of acid or base given the chemistry and flow rate of the fluids. The dosing-rate alone dictates the scale and the cost of the procedure. To-date, its evaluation is determined by laboratory titration of the collected fluid and series of field trials for the right pH. These methods were proven useful in designing and optimizing the procedure. However, these steps were at times laborious and costly.

This paper presents a new method, namely the SOLVEQ-pH method, a computer-assisted calculation of amount acid or base to be injected to the fluids to attain a desired pH. Its inception came when there was a need to have a fast and reliable tool to determine the cost of the procedure for different types of power plants at PNOC-EDC's new projects. In absence of actual power plants and on-line fluids to conduct field trial, the SOLVEQ-pH method is considered as an alternative.

This was devised to evaluate the procedures' technical and financial feasibility given a set of power plant designs. In this way, the procedure's optimum conditions were calculated and its technical and financial viability were assessed, without the costly construction of the set-up to test them.

The term SOLVEQ-pH distinguishes it from laboratory or field pH. It means calculated pH using SOLVEQ program. Field trials, however, is today's tool in determining the optimum dosing-rate. Below is the demonstration of the method, its results, and comparison to actual field data.

2.0 THEORY

Arnorsson *et al.*, (1995) identified three chemical processes that affect pH of geothermal fluids. They include rock dissolution, the supply of acids to the water and precipitation of some secondary minerals from the water. The first process tends to increase the water pH whereas the other two processes tend to decrease it. For this paper, only the supply of acid or base will be discussed by changing the pH using SOLVEQ. Effect of dissolution or precipitation were demonstrated from other works using pH modification, such as silica prevention by Garcia *et al.* (1996) and corrosion control of Villa *et al.* (2003) and Sanchez *et al.* (2001).

SOLVEQ is a FORTRAN computer program for computing homogeneous chemical equilibria in aqueous systems developed by Spycher and Reed (1990). It is used primarily as a work horse for processing water analyses of all types, but it is also useful for certain types of geochemical process modeling. For a given temperature, pH, and total composition of a homogeneous aqueous solution, SOLVEQ computes the activities of all aqueous species and the saturation indices of solids and gases' fugacity. It is the SOLVEQ's special ability to compute the homogeneous chemistry at various pH's that make this tool ideal in evaluating addition of acid or base to a particular solution.

The pH can be changed interactively when running SOLVEQ, just as for temperature. If pH is changed, the total molar amount of hydrogen ion is re-computed, and the charge balance is re-adjusted to compensate for the given pH. It is the adjustment of the charge balance, using a pre-selected ion that makes this program useful in computing amount of a particular base (such as NaOH) or acid (such as HCl).

For a base addition, Na ion (assuming addition as $NaOH = Na^+ + OH^-$) is added as the charge balancer while for acid addition, Cl^- is added as the balancer (assuming addition as $HCl = H^+ + Cl^-$). For acid like H_2SO_4, one can also use Cl ion as the balancer, provided its adjusted mole is divided by two. This is invoking the concept neutralization-equivalent-weight or simply the equivalent of H_2SO_4 to HCl: there are two equivalents per mole of H_2SO_4 per mole of HCl. Direct use of SO_4= (from H_2SO_4) as a charge balancer is not possible because this will interfere with the computation of redox potential: since the system is essentially in a reducing environment, SOLVEQ assumes a redox reaction of $HS^- + H_2O = 9H^+ + SO_4^= + 2e^-$ in its calculation of redox potential.

A note of caution, as calculating power of today's computers became faster and computer-time cheaper, geochemical simulations were at times taken for granted. There were times, simulations were mis-understood to be just too theoretical and hence unrealistic. At worst, data were carelessly used and particular assumptions were overlooked. To appreciate the use of these simulations, "theoretical" calculations are not mathematical axioms or abstract ideas to investigate a particular phenomenon. SOLVEQ (or any reliable geochemical programs) is a matrix of thermodynamic data from basic research experiments, integrated and packaged to a re-usable program. The quality of these data such as SOLTHERM, the data-matrix of SOLVEQ, is central to meaningful calculations. It is the primary goal of these calculations to validate similarities or differences from field data. If there are differences, actual conditions and assumptions must be validated to explain the disparity.

For this simulation, the assumptions are the following:

1. Line conditions, such as temperature, fluid flow, pressure, and volume, are constant
2. The given fluid chemistry represents the present fluids
3. Injection temperature of NaOH or H_2SO_4, usually at ambient condition, does not significantly change the initial temperatures and volume of the fluids
4. Complete dissociation of NaOH and H_2SO_4, (such as $NaOH = Na^+ + OH^-$ and $H_2SO_4 = SO_4^= + 2H^+$) is attained
5. Complete mixing of acid or base to the fluid

Table 1. Results and comparison of computed pH and chemistry of UM basin condensate chemistry in mg/kg except pH and Eh.

SAMPLE 1: UM CONDENSATE COLLECTED AT BASIN DATED 2003-8-17 *FLOW RATE: 290 KG/S*															
INITIAL CHEMISTRY OF UM CONDENSATE FROM BASIN															
DATE	PH/25C	DOSAGE LI/HR	Na	K	Ca	Mg	Fe	Cl	SO_4	HCO_3	B	NH_3	SiO_2	H_2S	TCO_2	Eh MV
8/17/2003	6.98	0.00	2.23	0.61	0.34	0.02	0.03	2.02	8.73	45.6	8.3	14.1	0.39	0.49	47.2	-113
COMPUTED CHEMISTRY AFTER DOSING 12.5 N (50%W/V) NAOH TO ATTAIN PH 8.0 AT LINE CONDITION															
DATE	PH/45C	DOSAGE LI/HR	Na	K	Ca	Mg	Fe	Cl	SO_4	HCO_3	B	NH_3	SiO_2	H_2S	TCO_2	Eh MV
8/17/2003	8.00	38.00	12.73	0.61	0.34	0.02	0.03	2.02	11.23	45.6	8.3	14.1	0.39	0.49	47.2	-344
ANALYZED CHEMISTRY AFTER DOSING 12.5 N (50%W/V) NAOH TO ATTAIN PH 8.0 AT LINE CONDITION															
DATE	PH/25C	DOSAGE LI/HR	Na	K	Ca	Mg	Fe	Cl	SO_4	HCO_3	B	NH_3	SiO_2	H_2S	TCO_2	Eh MV
8/17/2003	8.37	28.00	14.7	0.47	0.32	0.04	0.47	1.02	8.78	72.1	6.99	11.9	0.33	0.2	53.1	-111
SAMPLE 2: UM CONDENSATE COLLECTED AT BASIN DATED 2003-9-15 *FLOW RATE: 290 KG/S*															
INITIAL CHEMISTRY OF UM CONDENSATE FROM BASIN BEFORE DOSING															
DATE	PH/25C	DOSAGE LI/HR	Na	K	Ca	Mg	Fe	Cl	SO_4	HCO_3	B	NH_3	SiO_2	H_2S	TCO_2	Eh MV
9/15/2003	6.85	0.00	1.72	0.15	0.28	0.03	0.03	1.09	8.9	38.2	8.3	14.3	0.69	1.99	44.7	-152
COMPUTED CHEMISTRY AFTER DOSING 12.5 N (50%W/V) NAOH TO ATTAIN PH 8.0 AT LINE CONDITION															
DATE	PH/45C	DOSAGE LI/HR	Na	K	Ca	Mg	Fe	Cl	SO_4	HCO_3	B	NH_3	SiO_2	H_2S	TCO_2	Eh MV
9/15/2003	8.00	42.43	13.4	0.15	0.28	0.03	0.03	1.09	15.15	38.2	8.3	14.3	0.69	1.99	44.7	-341
ANALYZED CHEMISTRY AFTER DOSING 12.5 N (50%W/V) NAOH TO ATTAIN PH 8.0 AT LINE CONDITION															
DATE	PH	DOSAGE LI/HR	Na	K	Ca	Mg	Fe	Cl	SO_4	HCO_3	B	NH_3	SiO_2	H_2S	TCO_2	Eh MV
9/15/2003	8.18	28.00	16.8	0.47	0.28	0.03	0.03	1.01	7.18	70.9	9.66	14.8	0.68	2.42	52.2	-111

Table 2a. Representative chemistry (As Average) of MB brine for pH adjustment to 5.50 at line condition using 36N H_2SO_4.

Source	Treatment	Flow, kg/s	Temp, °C	Date	pH	Na	K	Ca	Mg	Fe	Cl	SO_4	HCO_3	B	NH_3	SiO_2	H_2S	TCO_2
MB Brine	H2SO4, 36N (18M)	0.18-0.22 (Pilot Test)	160	2003-06-19	6.97	5564	1157	238	0.21	0.39	10691	20.50	7.96	209	3.11	773	5.02	23.90
				2003-07-11	7.01	5553	1089	264	0.60	0.53	10574	27.00	6.42	213	2.68	713	5.36	18.40
				2003-08-11	7.04	5949	1428	259	0.31	0.35	10660	22.80	6.72	206	1.32	776	4.91	13.20
				AVERAGE	**7.01**	**5689**	**1225**	**254**	**0.37**	**0.42**	**10642**	**23.43**	**7.03**	**209**	**2.37**	**754**	**5.10**	**18.50**

Table 2b. Computation of dosing rate of 36N H_2SO_4 to adjust pH 5.5 at line condition from SOLVEQ/PHREEQC programs using average brine chemistry.

Given the average brine chemistry in Table 2a and brine-line pH of 6.44 at 160°C

1) COMPUTED MOLES OF CL (AS HCL) TO ADJUST LINE PH TO 5.50 PER KG BRINE = **0.00177 MOLES**

2) VOLUME OF 36N H2SO4 TO ATTAIN PH 5.50 IN LITERS = 0.00177 / 2) / 36 N = **0.000150 LITERS**

3) DOSING RATE OF 36N H2SO4 (LITERS/HR) AT BRINE FLOW OF 0.18 KG/S = **0.097 LITERS/HR**

3) DOSING RATE OF 36N H2SO4 (LITERS/HR) AT BRINE FLOW OF 0.22 KG/S = **0.118 LITERS /HR**

Based from these computation, to adjust the brine-line pH from 6.44 to 5.50 :

The dosing rate of 36N H_2SO_4 ranges from **97 ml/hr to 118 ml/hr** or an ***Average Dosing*** of **108 ml/hr**

It is envisaged that the differences between actual and computed values are attributed to the violations of these assumptions. If there were similarities these assumptions were then attained.

3.0 METHODOLOGY

Two sets of data were used for this demonstration. These are the representative steam condensate and brine chemistry from Leyte geothermal project as shown in Tables 1 and 2, respectively. Below are the SOLVEQ methodologies of NaOH and H_2SO_4 treatment in pH modification of geothermal fluids:

3.1 Method for NaOH Treatment to Attain pH 8.00

Step 1. Get representative complete fluids chemistry as analyzed.

As much as possible the data to be used represents the usual untreated sample. This was ensured by getting a sample with a good analytical balance. Usually it has 1 to 5% ion balance difference. Sample with difference of higher than 5% is not recommended for this type of calculations.

Step 2. Using PHREEQC or Watchworks or SOLVEQ run data at desired line temperature (as data2).

This step is called ‘data polishing’. Polishing includes correcting the ion difference to zero at line temperature. This can be done by running the sample at line temperature using any reliable geochemical simulation codes (such as PHREEQC, Watchworks, or SOLVEQ). For Leyte sample, the condensate temperature is set at 45°C.

Step 3. Using SOLVEQ, run data2 at desired line temperature but vary pH to 8.00.

With a polished data, we are now ready to adjust the pH to 8.00 using Na ion as the charge balancer. Refer to SOLVEQ manual (Spycher and Reed, 1990) to interactively select Na as the balancer.

Step 4. Run SOLVEQ at pH 8.00 using Na as charge-balancer simulating dissociation of NaOH.

Step 5. Compute moles of NaOH (as Na^+) added per kg condensate.

To compute the amount of NaOH adder per kg of sample, get the amount of Na ion added or changed from the SOLVEQ's output file. This can be computed as illustrated below:

Sample Calculation

Find: Dosing rate of 12.5 N (50% w/v) NaOH needed per kg condensate to attain pH 8.000

Given: From SOLVEQ output file, it needed $0.446e^{-3}$ NaOH (as Na^+) equivalent per kg condensate at line condition to adjust pH to 8.00; Flow rate of condensate = 290 kg/s

Solution

Volume of 12.5N NaOH/kg sample
= $0.446e^{-3}$ equivalent / 12.5N
= $3.57e^{-5}$ liter/kg-condensate
At given flowrate, compute dosing rate:
Dosing rate = 290 kg-condensate/sec x 3.57 e^{-5} liter/kg-condensate x 3600 sec/hr
= 37 li/hr

3.2 For H_2SO_4 Treatment to Attain pH 5.50

Step 1. Get representative complete fluids chemistry as analyzed.

Step 2. Using SOLVEQ run data at desired line temperature (as data2).

Similar to NaOH treament, initial data has to be polished before any pH adjustment. For Leyte sample, the brine temperature is set at 160°C.

Step 3. Using SOLVEQ, run data2 at desired line temperature but vary pH to 5.50.

With a polished data, we are now ready to adjust the pH to 5.50 using Cl ion as the charge balancer. Again refer to SOLVEQ’s manual (Spycher and Reed,1990) to interactively select Cl as the balancer.

Step 4. Run SOLVEQ at pH 5.50 using Cl as charge-balancer simulating dissociation of HCl.

Step 5. Compute moles of H_2SO_4 (as $Cl^-/2$) added per kg condensate.

From the SOLVEQ output file, get the moles of Cl ion from the SOLVEQ’s output file, and divide it by two. This can be computed as illustrated below:

Sample Calculation

Find: Dosing rate of 36 N H_2SO_4 needed per kg condensate to attain pH 5.50

Given: From SOLVEQ output file, it needed $0.1077e^{-1}$ / 2 H_2SO_4 equivalent (as HCl) per kg brine to adjust line pH to 5.50 at line condition; Flowrate of brine = 0.18 kg/s

Solution

Volume of 36 N H_2SO_4 sample
= $5.385e^{-3}$ equivalent / 36 N
= $1.496e^{-4}$ liter/kg-brine
At given flowrate, compute dosing rate:
Dosing rate = 0.18 kg-brine/sec x $1.49e^{-4}$ liter/kg-brine x 3600 sec/hr
= 0.097 li/hr

4.0 RESULTS AND DISCUSSION

The computed data from NaOH and H_2SO_4 methods are presented in Tables 1 and 2, respectively. Table 1 includes the actual untreated/treated chemistry of the fluid based

Table 3. Comparison of actual from computed data in NaOH treatment.

Sample	Field dosing rate li/ hr (Villa et al., 2003)	Computed dosing rate li/hr	Percent difference (computed – field)/field x 100
8-17-2003	28	38	36
9-15-2003	28	42	52
Average		40±3	

Table 4. Comparison of actual from computed data in H_2SO_4 treatment.

Comparison	Brine flow kg/s	Injection temp. °C	pH before H_2SO_4 injection	pH after H_2SO_4 injection	Dosing rate (ml/hr)
MB lab pH based from lab titration	0.10	25	6.90 measured at 25°C	5.60 measured at 25°C	4.80
MB pH-MOD pilot test trials (actual)	0.18 to 0.22	160	6.97 to 7.04 measured at 25°C	5.20 to 5.60 measured at 25°C	8.93
This study	0.18 to 0.22	160	6.44 computed at 160°C	5.50 computed at 160°C	97 to 118

from the field and its dosing rate in comparison with the computed data. However, in Table 2, only untreated brine is presented and the field dosing rate. For H_2SO_4 treatment there is a critical difference between the computed and the laboratory results.

4.1 About NaOH Treatment

Table 3 shows the percent difference in dosing rate of the 50% (w/v) NaOH between computed and field data from two samples.

Although there are significant differences of 36 to 52% between the computed and field dosing rates, the difference between resultant chemistry (except for Eh) is generally small. Example, the difference in increase in Na (from NaOH injection) between the computed and analysis varies by 1.0 to 3.0 mg/kg only, which for practical purposes, is essentially the same. The only major analytical difference is the Eh or the redox potential. Theoretically, Eh and pH are interdependent: as Eh decreases (or increase its negative value) pH increases. Since the analytical value remained unchanged, there must a possible error in its measurements, and consequently, its pH. In this case, the computed data are more reliable than the analytical values.

The disparity in Eh and pH may indicate that there must be something wrong with the on-line pH meter, hence NaOH dosage was suspect. It is also possible that condensate flow is actually larger and varies significantly than previously measured. In this case, it was recommended that a re-evaluation of the fluctuation in the condensate flow be established and check the on-line pH/Eh meter.

4.2 About H_2SO_4 treatment

The results of Table 2 were compared from the actual pH-modification system in Malitbog (MB) at Leyte (Alcober, 2003) as presented in Table 4.

The difference between the computed and field dosing rates is calculated to be more than 10x or one log unit. The disparity is caused by the difference in temperature when the pH is computed. This hypothesis is tested by re-computing the dosing rate given the sample's average chemistry both at 25°C, and employing similar SOLVEQ methodology. If the computed dosing rate coincided or near the measured rate, then it is likely that the higher temperature (at 160°C) caused the difference. Table 5 shows the results of this re-computation:

Table 5. Re-computation of the H_2SO_4 treatment of brine at 25°C.

Sample	See Table 2a, use average brine chemistry
Equivalent of H_2SO_4 per kg brine at 25°C	$1.852e^{-4}$ equivalents
Volume of 36N H_2SO_4 per kg brine at 25°C	$5.165e^{-6}$ liter
Computed dosing rate at 0.18 kg/s brine flow	0.00333 li/hr or 3.33 ml/hr
Computed dosing rate at 0.22 kg/s brine flow	0.00409 li/hr or 4.09 ml/hr

The computed results, 3.33 to 4.08 ml/hr assuming dosing at 25°C, is now closer to the laboratory titration data. This is understandable since the simulation does not include the actual injection temperature of 160°C. Based from this re-computation, temperature difference affects the acid treatment of the brine.

Another major that caused this disparity is the violation of one of our assumptions: the complete dissociation of H_2SO_4 as $SO_4^=$ and H^+. This assumption, although valid in terms of chemical equivalence, is wanting at elevated temperature. At 25°C, H_2SO_4 acid is essentially a strong acid in terms of its first and second H^+ dissociation. However, at 160°C, its second dissociation as HSO_4^-, is weaker by 1.88 log unit than at 25°C. Table 6 shows the log K of HSO_4^- at 25 and 160°C based from Sillen and Martell (1964).

Table 6. Log K of H_2SO_4 at 25 and 160°C

Log K of HSO_4- at 25°C	-1.96
Log K of HSO_4- at 160°C	-3.84

Since H_2SO_4 acid (as HSO_4^-) had actually gone weaker at line temperature, brine needs higher dosage to attain pH 5.50. However, at 25°C where its acidity is stronger, then lesser dosage is required. Although the SOLVEQ program does compensate for this difference in log K of HSO_4^- making our computations valid, our initial assumptions of total dissociation is wanting. This explains the disparity between the values.

5.0 CONCLUSION

Using SOLVEQ, two sets of data from Leyte were used to demonstrate it's special capability to compute the homogeneous chemistry at various pH's. The pH can be changed interactively when running SOLVEQ, just as for temperature. If pH is changed, the total molar amount of hydrogen ion is re-computed, and the charge balance is re-adjusted to compensate for the given pH. It is the adjustment of the charge balance, using a pre-selected ion that makes this program useful in computing amount of a particular base (such as NaOH) or acid (such as HCl or H_2SO_4).

For base addition, Na ion (assuming addition as $NaOH = Na^+ + OH^-$) is added as the charge balancer while for acid addition, Cl^- is added as the balancer (assumimg addition as $HCl = H^+ + Cl^-$). For acid like H_2SO_4, one can also use Cl ion as the balancer, provided its adjusted mole is divided by two. This is invoking the concept neutralization-equivalent-weight or simply the equivalent of H_2SO_4 to HCl: there are two equivalents per mole of H_2SO_4 per mole of HCl. Direct use of SO_4= (from H_2SO_4) as a charge balancer is not possible because this will interfere with the computation of redox potential: since the system is essentially in a reducing environment, SOLVEQ assumes a redox reaction of $HS^- + H_2O = 9H^+ + SO_4^= + 2e^-$ in its calculation of redox potential.

Based from the results of the SOLVEQ-pH method, it can evaluate not only the needed dosage of acid or base treatment, but also the intricacy of the chemical equilibrium involved in the procedure. To-date, although measurements of pH's at laboratory condition remain the only method of choice in evaluating required dosages, evaluation through simulation strengthen our understanding of the real conditions that affect the treatment. For NaOH, fluxes in condensate flow and the role of Eh are essential parameters to monitor effectiveness of the treatment, while for H_2SO_4, the line-temperature dictates the strength of acidity, hence the amount of dosage.

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