H_2S Scrubbing from Noncondensable Gas in After-Condensers

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ABSTRACT

At Unit 12 in The Geysers, Calpine operates a Burner/Scrubber system to remove H_2S from the noncondensable gas. The operating rate of this unit has been declining over the years, and the noncondensable gas flow rate and H_2S concentration are about 10% or less of the original design. The partitioning of H_2S to the vent gas in the main condenser is low, and the absorbed H_2S in the cooling tower water systems is mostly naturally oxidized without the need for chemical addition. Additionally, the heating value of the noncondensable gas is now much lower than design, resulting in the need to add supplemental fuel to the burner. As a result of the low current H_2S and gas rates, if the Burner/Scrubber system is shut down, only a portion of the H_2S in the gas will have to be removed in order to comply with the permit limit. The after-condenser was considered for use in the bulk removal of H_2S from the gas. A study, based on process simulations and data taken at other Calpine facilities, was conducted to evaluate H_2S removal in the Unit 12 after-condenser i) in its current spray-tower arrangement and also ii) assuming the addition of a packed section. The methods used to predict H_2S removal in the after-condenser with and without the addition of a packed bed are discussed. Ideally only condensate would be used to absorb the H_2S, with the absorbed H_2S being naturally oxidized in the cooling tower loop. Some caustic addition could also be required to enhance H_2S removal, and also to control cooling water pH. The effect of operating conditions and packing characteristics on the performance of the after-condenser as an H_2S scrubber are also discussed.

1. Introduction

Geysers Cobb Creek Unit 12 began operation in 1979 generating 110 GMW (gross megawatts) with 1.9 million pounds per hour of dry steam at 100 psig inlet pressure. Due to reservoir decline at The Geysers, Unit 12 presently generates about 55 GMW, with an inlet steam pressure of 70 psig. Unit 12 was the first Geysers plant to be constructed with an integral H_2S abatement system, while all other previous Geysers plants were being retrofitted with H_2S abatement systems in order to meet Air Regulatory emission limits that began to be imposed in the late 1970's. The H_2S abatement system, deemed Iron –Caustic – Peroxide (ICP), scrubbed all of the incoming H_2S into the condensate, where it reacted with iron to form elemental sulfur. In this scheme, the plant direct-contact condensers are co-opted as H_2S scrubbers, by adding caustic soda to the circulating water. As shown in Figure 1, the caustic is added at the water inlet to the after-condenser, therefore the after-condenser acts as a final gas polishing step to maintain the H_2S concentration in the treated gas to less than 1000 ppmv, a regulatory limit. Once scrubbed into the condensate the H_2S is removed by reaction with ferric iron, in the form of an iron chelate, by the following reaction producing solid elemental sulfur.

$$H_2S + 2Fe^{+3}\text{ chelate} \rightarrow S^0\downarrow + 2Fe^{+2}\text{ chelate} + 2H^+ \quad (1)$$

The elemental sulfur produced in the condensate by the ICP-H_2S-abatement-coated heat exchanger surfaces, reduced plant cooling efficiency, and, over the course of 3-4 years, clogged injection wells. The sludge required a solids removal (clarifier) system that produced a hazardous waste and resulted in expensive transportation and disposal in a hazardous waste landfill.

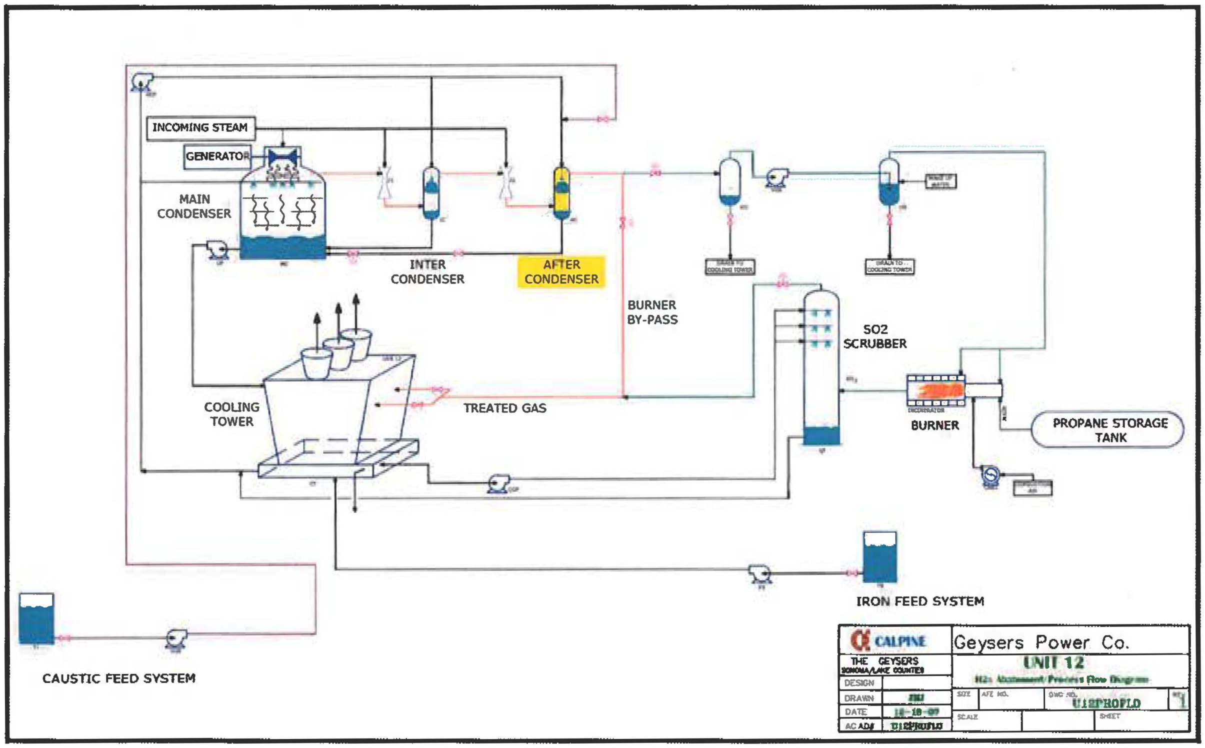

Figure 1: Unit 12 Process Flow Diagram

In 1983, Burner/Scrubber technology had been developed at The Geysers plants, as a means of eliminating circulating water sulfur solids, and the clarifier system. The Burner/Scrubber eliminates sulfur solids in the circulating water by solubilizing the elemental sulfur when the sulfurous acid produced in the scrubber is combined with the sulfur produced in the iron/H_2S reaction in the condensate to produce soluble thiosulfate, by the following reactions:

$$H_2S + 3/2O_2 \rightarrow SO_2 + H_2O \rightarrow HSO_3^{-1} + H^+ \text{ (Combustion and scrubbing)} \tag{2}$$

$$H_2S + 2Fe^{+3} \rightarrow S^0\downarrow + 2Fe^{+2} + 2H^+ \text{ (Aqueous chemical oxidation)} \tag{3}$$

$$HSO_3^{-1} + S^0 \rightarrow S_2O_3^{=} + H^+ \text{ (Sulfur dissolution)} \tag{4}$$

The decline of The Geysers reservoir that resulted in lower steam pressures was accompanied by higher noncondensable gas (NCG) and H_2S concentrations in the supplied steam. While the Unit 12 steam supply had previously been low in NCG, and therefore was not a candidate for Burner/Scrubber, by 1988 the NCG and H_2S levels had risen and justified a Burner/Scrubber system at Unit 12, designed with the capacity to handle as much as 316 lb/hr H_2S.

In the intervening years, treated wastewater injection was begun into the Geysers steam reservoir, and the NCG content of the steam has dropped, resulting in a greatly reduced H_2S concentration in the steam as shown in Figure 2. The NCG concentration has dropped considerably, to levels less than seen during the initial operation of the plant in 1979. Combined with a reduction in steam flow due to pressure decline, the overall NCG mass flow rate has declined to the point that adjunct fuel is needed in order to operate the Burner/Scrubber system. The H_2S contained in the steam to the plant has dropped to only about 90 lbs/hr. The high liquid/gas ratio that occurs in the Unit 12 direct contact condensers, coupled with elevated pH due to naturally occurring ammonia in the steam, results in low H_2S "partitioning" (% H_2S in vent gas vs condensate). Only about 20 lbs per hour of H_2S is currently treated by the Burner/Scrubber, or about 6% of the design Burner capacity.

Operating the Burner/Scrubber so far below its design capacity had resulted in added cost due to the need to add about 3 gallons per hour of propane to the feed gas. Operational difficulties also occurred at low gas rates, e.g. burner flame stability, vent gas burn-back, and combustion control issues. For these reasons, Calpine determined that operating without the Burner would be advantageous, as long as emission limitations could be met and circulating solids did not reduce operational efficiency of the power plant. Operational testing without the dissolution of sulfur from Burner/Scrubber indicated that the additional 20 lbs per hour of sulfur to the circulating water (achieved with caustic scrubbing in the after-condenser) had no detrimental effect on plant operation. Simply shutting down the Burner/Scrubber was not an option however, due to the high concentration of H_2S in the vent gas leaving the after-condenser (a regulatory limit). Caustic scrubbing normally utilized during non-Burner operation comes with a cost roughly equivalent to the use of propane in the Burner. It was reasoned though, that if adequate H_2S scrubbing in the after-condenser could be achieved with water only (and the possible addition of mass transfer packing), the overall H_2S mass emissions could be managed through condensate H_2S natural abatement in the circulating water, saving both the propane needed for Burner operation, and caustic needed for after-condenser scrubbing. An added advantage to non-Burner operation is the reduction in plant parasitic MW load, because shutting down the Burner eliminates pumps and blowers needed in its operation.

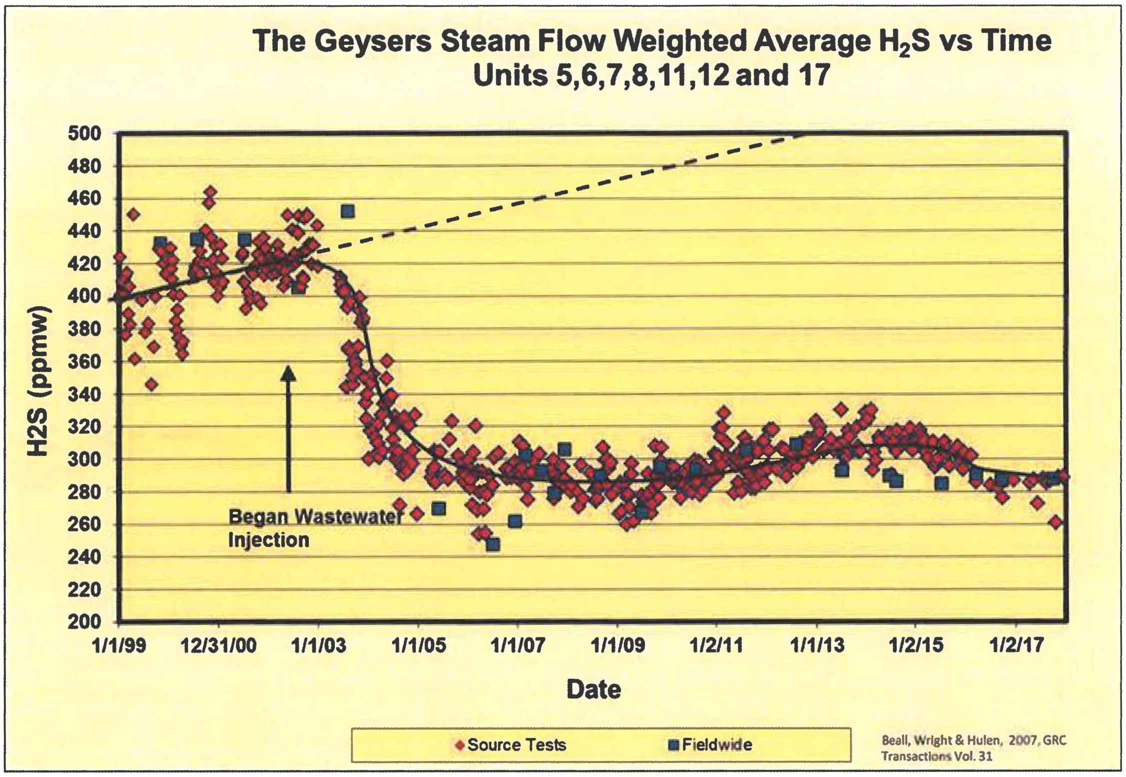

Figure 2: The Geysers Steam Flow Weighted Average H_2S vs Time (Units 5, 6, 7, 8, 11, 12 and 17)

In pursuit of a non-Burner and no caustic feed H_2S abatement scheme, Calpine sought assistance from Trimeric Corporation to model the Unit 12 after-condenser as a water only scrubber, with the possible addition of mass transfer packing to improve H_2S scrubbing without added caustic soda. Calpine also sought and received regulatory changes that allow increased treated vent gas H_2S concentrations (>1000 ppmv).

This paper first summarizes the design basis for the evaluation of the Unit 12 after-condenser as an H_2S scrubber. Then, the methods used to predict H_2S removal in the after-condenser in its current spray tower configuration and as a packed bed are also discussed. The use of only condensate to absorb H_2S versus caustic addition (to enhance H_2S removal and control pH) is also examined. The effects of operating conditions (temperature, water flow rate, and caustic addition) on the performance of the after-condenser as an H_2S scrubber are presented as well.

2. Design Basis

The design basis is shown in Table 1 for the Unit 12 after-condenser as an H_2S scrubber when the main Burner/Scrubber system is off-line.

Table 1: Unit 12 After-Condenser Design Basis

Cooling Tower Water Feed to After-Condenser		
Flow rate	gpm	1,330 (max)
Temperature	F	75 (range: 55-90)
pH		6.8 – 7.7
NCG to After-Condenser: Conditions		
Flow rate (dry)	lb/h	2,600
Temperature	F	75
Pressure	psia	12.9
NCG to After-Condenser: Composition (dry basis)		
H_2S	mol%	0.45
CO_2	mol%	40.78
CH_4	mol%	2.01
N_2	mol%	35.86
O_2	mol%	10.95
H_2	mol%	9.94
H_2S Removal Requirement	%	50 (min)

Because of the high cost of operating the existing Burner/Scrubber NCG treatment system, Calpine sought an alternate solution for achieving the modest H_2S removal efficiency required (50%) that would allow them to shut down the Burner/Scrubber unit. Caustic (aqueous solution of NaOH) is a well-known chemical that can be used to scrub H_2S from gases (converting it to NaHS and Na_2S), but the presence of large amounts of CO_2 in the NCG complicates the use of caustic because CO_2 tends to be absorbed into caustic to a similar extent as H_2S. As a result of the high ratio of CO_2 to H_2S in the NCG, most of the caustic would be spent scrubbing CO_2, resulting in large caustic usage relative to the amount of H_2S removed from the NCG.

H_2S has a low solubility in water, but in some situations water can be an effective solvent to absorb H_2S from gases (Mamrosh, et al., 2014, McIntush, et al., 2017). Factors that favor consideration of water as a solvent for H_2S include:

- a large volume of water is available;
- the spent water must have a disposition route;
- the water cannot contain H_2S and would preferably not be acidic;
- the water should be cool or cold; and
- deep H_2S removal from the gas is not required.

The use of cooling tower water (originating as condensate from the main condenser) was considered for the scrubbing of the bulk of H_2S from the NCG entering the after-condenser at Unit 12. Additionally, it was not desired to add iron chelate to the cooling tower water because operating experience indicates that the majority of the H_2S absorbed into the water is oxidized naturally (to elemental sulfur and sulfur oxyanions) without the presence of the added iron, and little H_2S is stripped from the water into the air in the cooling tower. It was also assumed that there is no H_2S in the water feeding the after-condenser because of the natural oxidation. Some ammonia may also be present in the cooling tower water because it is absorbed from the inlet NCG in the main condenser, and may be held in the water (rather than stripped out into the air in the cooling tower) because it is associated with an acidic anion, and be present as ammonium carbonate and/or ammonium sulfate. Unit 12 operating data shows that the main salt species present in the cooling tower water is ammonium sulfate – $(NH_4)_2SO_4$ – which is present at a concentration of approximately 480 ppmw.

The NCG enters the after-condenser at a rate of 2,600 lb/hr at 75°F and a pressure of 12.9 psia. The NCG contains 4,500 ppmv H_2S with the significant amounts of CO_2, N_2, O_2, H_2, and CH_4 as shown in Table 1. The desired H_2S removal is approximately 50%.

As shown in Figure 3, the after-condenser has a 4' diameter with a height of about 20'. The gas enters the vessel from the discharge of the ejector and flows upwards through the vessel. Cooling tower water flows through a single spray nozzle (10" x 3" with internal vanes) mounted near the top of the vessel and sprays downward to counter-currently contact and further condense water from the gas. The spray water and condensate collect in the bottom of the after-condenser and flow to the cooling tower for temperature reduction and recycle through the Unit 12 condensers. Figure 3 shows that there is significant space in the after-condenser for the addition of packing if needed to achieve the desired H_2S removal.

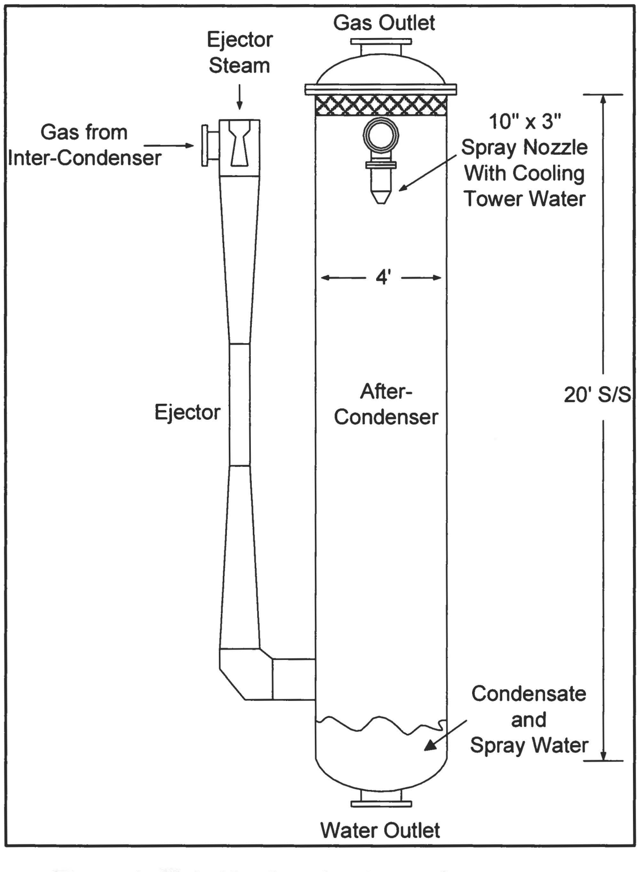

Figure 3: Unit 12 After-Condenser Spray Tower

The methods used to predict H_2S removal in the after-condenser using water with and without packing are described in the following sections.

3. Spray Tower Evaluation

Mass transfer in spray towers is not as well characterized in the literature as packed and tray towers, and absorption of H_2S into the water or caustic from gases in this mode is not common. Also, the use of water versus caustic to absorb H_2S gives very different results and different analysis methods may be used.

Calpine had previously collected test data for the Unit 11 after-condenser using caustic solution to characterize its use as a backup scrubber for H_2S removal when the burner was out of service. In addition to the caustic test data, some data without significant caustic addition was also collected (referred to as "water only" tests) at Unit 11 – very little caustic relative to the amount of H_2S in the NCG (only ~2% of the theoretical requirement) was added with varying water flow to the spray nozzle. Given the similarities of the Unit 11 and 12 after-condenser vessels, the low-caustic data from Unit 11 was applied to Unit 12 to estimate the mass transfer coefficients for the use of water to absorb H_2S in a spray tower.

3.1 Calculation Methods

The design details of the spray nozzle installed in the Unit 12 after-condenser were not known and there are limited standard methods for characterizing mass transfer in these systems. Because of these lack of design information it was not feasible to estimate some characteristics of the spray, such as the droplet size and number of droplets, a non-standard method was used to evaluate mass transfer in the spray tower. The method typically applied to packed towers was used to analyze the spray tower data. In this method, the height of the contacting section (h) is a function of the height of a transfer unit (HTU) and the number of transfer units (NTU):

$$h = NTU * HTU \tag{5}$$

$$HTU = \frac{G_m}{K_G a\ P} \tag{6}$$

$$NTU = \int_{y2}^{y1} \frac{dy}{y - y_e} \tag{7}$$

G_m is the gas superficial molar velocity, y is the mole fraction of H_2S in the gas, y_e is the mole fraction of H_2S in the gas at equilibrium with the liquid phase, K_Ga is the overall gas-phase mass transfer coefficient, P is the absolute pressure, and subscript *1* refers to the bottom of the column (gas inlet, liquid outlet) and subscript *2* refers to the top of the column.

Because the spray tower is not packed, there is no formal definition of the contacting section (h). In this study, it was assumed that h= 4 ft because in the Unit 11 work it was found that much of the water sprayed from the nozzle hits the wall at a distance from the nozzle of less than four feet, resulting in large losses of mass transfer area as the distance from the nozzle becomes greater; this mass transfer height was kept as a constant parameter.

For caustic scrubbers the term y_e is usually ignored, because when excess caustic is present the equilibrium partial pressure of H_2S is very small at most conditions. For that case, the NTU term simplifies to commonly used quantity: $\ln(y_1/y_2)$. However, for the scrubbing of H_2S with water only, y_e cannot be ignored due to the very limited solubility of H_2S in water.

When the amount of salts in water is very small and the pH is nearly neutral (such as is the case for Unit 12), as the H_2S is absorbed into water the pH quickly drops as a very small amount of H_2S is absorbed. Although a portion of the initial H_2S absorbed dissociates, the bulk of the H_2S absorbed is in its molecular form in water due to the lowered pH. This assumption results in the viability of analyzing the solubility of H_2S in water using Henry's Law only, and ignoring the acid-base chemistry. When Henry's Law is used, the integral term for NTU solves as shown in equation 8 (Treybal, 1980).

$$NTU = \frac{ln\left[\frac{y_1 - mx_2}{y_2 - mx_2}\left(1 - {}^1\!/_A\right) + {}^1\!/_A\right]}{1 - {}^1\!/_A} \tag{8}$$

Where m is the Henry's Law constant (converted to consistent units), and $A = L_m/mG_m$. L_m is the liquid superficial molar velocity and x_2 is the mole fraction of H_2S in the liquid at the top of the column.

3.2 Estimation of Unit 12 Water Spray Performance

The Unit 11 data (gas flow rate, liquid flow rate, H_2S in the feed and outlet gas) were used to calculate the NTU for several different water flow rates. With the assumed value of h (4 feet), the HTU could be calculated and then the K_Ga was estimated from the HTU. The K_Ga values calculated from this analysis are shown in the first row of data in Table 2.

Table 2: Calculation of H_2S in Treated Gas for Unit 12 Water Spray Scrubbing (Uses Unit 11 K_Ga Estimates)

Parameter	**Units**	**500 gpm water**	**950 gpm water**	**1,330 gpm water**
	Unit 11 K_Ga Data			
K_Ga (from Unit 11 data)	lbmol/h ft^3 atm	0.31	1.8	3.5
	Unit 12 Data and H_2S Removal Estimates			
NCG flow to Unit 12 after-condenser	lb/h (measured)	2,600	2,600	2,600
H_2S in	vol% (measured)	0.45	0.45	0.45
H_2S in	lb/h (calculated)	12.4	12.4	12.4
Temperature	F (water inlet temperature)	75	75	75
Henry's Constant	MPa/molfrac, derived from literature (Carroll, 1989)	53.2	53.2	53.2
Pressure (abs)	atm (assumed)	0.88	0.88	0.88
NTU	(calculated)	0.17	0.99	1.9
HTU	ft (calculated)	23.6	4.1	2.1
H_2S out	vol% (calculated)	0.40	0.27	0.18
H_2S removed	% (calculated)	12	40	59
H_2S in treated gas	lb/h (calculated)	10.9	7.4	5.1

The variation of the Unit 11 K_Ga with water feed rate shows the expected trend: increased K_Ga (i.e., better mass transfer efficiency) due to higher water flow rate. The higher water flow rate probably results in greater turbulence and more total mass transfer area. However, the magnitude of the K_Ga values appears to be a little higher than might be expected given that the system is non-reactive (physical-solubility-controlled scrubbing only). During the Unit 11 runs some caustic was added, albeit an insubstantial amount compared to the H_2S load. Additionally, a small amount of iron chelate was probably also present in the water, and the salt loads were probably substantial (possibly buffering the assumed pH drop with H_2S absorption). It is likely that the presence of these components resulted in enhancement of the mass transfer rate and changes to the equilibrium with the net result probably inflating the K_Ga.

Using the Unit 11 K_Ga values, the same methods were applied to Unit 12 to determine the H_2S concentration in the outlet gas expected as a function of water flow rate. The results for Unit 12 are also shown in Table 2.

The calculated data predict H_2S removal efficiencies that ranged from 12 to 59%, depending on the feed water flow rate. As the water feed rate to the after-condenser increased from 500 to 1,330 gpm, the H_2S removal increased by almost a factor of 5. Because of the fact that these predictions are based on mass transfer rates measured in Unit 11 when some caustic was added, and salts and iron chelate were present, it is likely that the H_2S removal for Unit 12 is somewhat overestimated. For reference, the Unit 11 spray tower H_2S removal during the "water only" tests ranged from 3 to 26%.

4. Packed Tower Evaluation

The design of the Unit 12 after-condenser is such that there is sufficient space between the water inlet nozzle and the gas inlet nozzle such that a packing could be added to enhance mass transfer between the two phases. It was, therefore, desired to estimate if the installation of a packing would enhance the predicted removal of H_2S from the NCG in the Unit 12 after-condenser. For the packed tower analysis, a commercial process simulation software (Aspen Plus) was used to estimate the achievable H_2S removal. The electrolyte non-random two-liquid (ENRTL) property package was used to represent the thermodynamics of the system in Aspen. The electrolyte model takes into account the vapor-liquid equilibrium, gas solubility, and electrolyte equilibrium of the ionic species. Aspen also has mass transfer models for common random packings of the type that would be used in the Unit 12 after-condenser. The H_2S removal modeling results and impacts of operating conditions and packing characteristics are discussed in the subsections below.

4.1 Estimation of Unit 12 Packed Tower Performance

Aspen Plus was used to create a simple model of the Unit 12 after-condenser for the conditions outlined in Table 1. For this analysis, it was assumed that 10 feet of a common random packing (#3.5 Jaeger Tri-Packs®) would be installed in the Unit 12 after-condenser. The results of this analysis are shown in Figure 4.

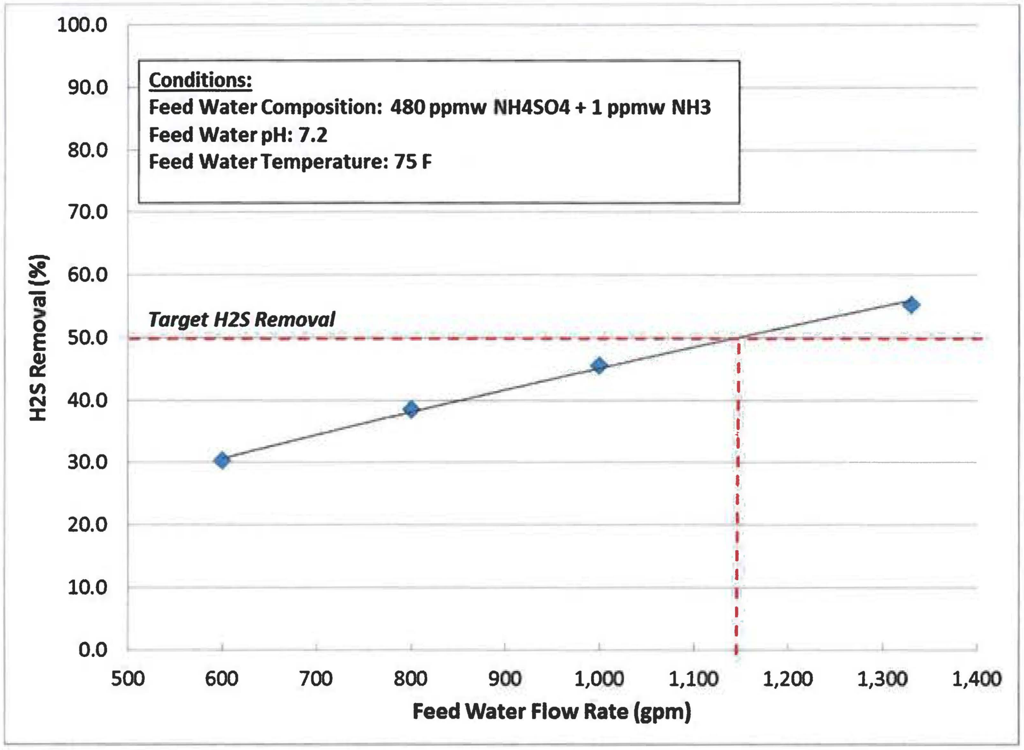

Figure 4: Predictions of H_2S in Treated Gas for Unit 12 Packed Tower Scrubbing (no caustic addition)

The figure shows that the H_2S removal efficiency ranges from 30 to 56% as the feed water flow rate to the after-condenser varied from 600 to 1,330 gpm, respectively. Water flow rates higher than 1,150 gpm would meet the 50% H_2S removal requirement necessitated by Calpine.

4.2 Impact of Water Temperature

The impacts of the feed water temperature on H_2S removal in the Unit 12 after-condenser were also evaluated using Aspen simulations. Figure 5 shows that the removal of H_2S is also a significant function of temperature. As the feed water temperature varies from 55 to 85°F, the H_2S removal efficiency decreases from 63 to 52%. This is because the solubility of H_2S in water decreases with increasing temperature.

The 50% H_2S removal specification can be met up to about 90°F at the conditions outlined in the figure. It is assumed that no significant temperature change occurs in the water in the after-condenser; however, in reality the water temperature will likely increase somewhat.

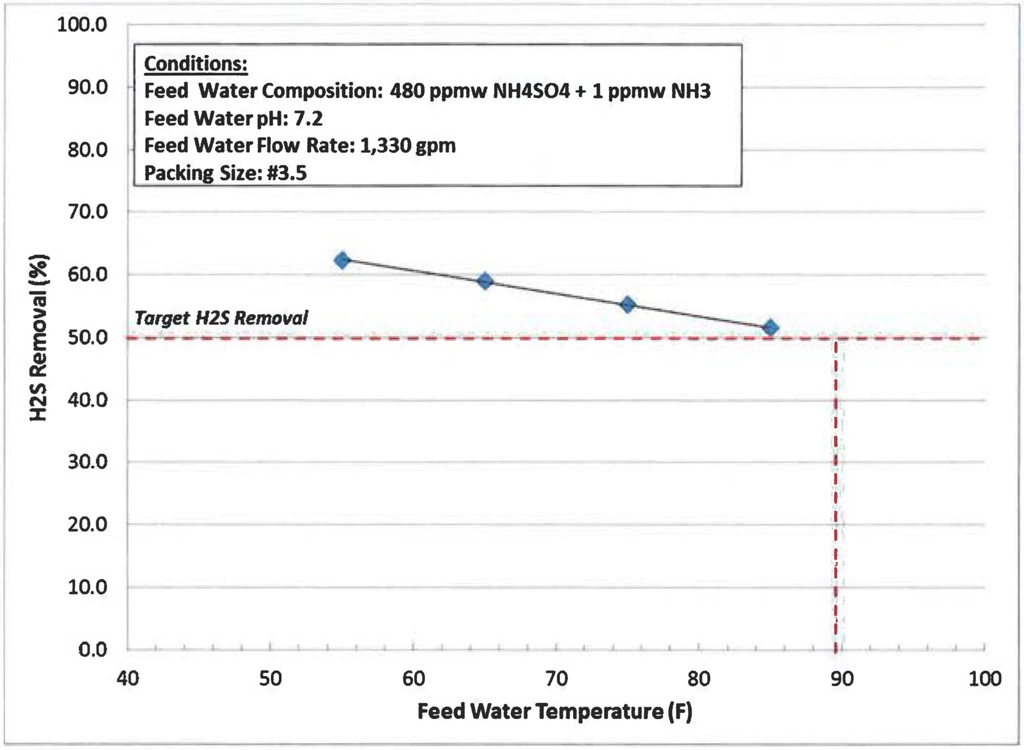

Figure 5: Impact of Feed Water Temperature on H_2S Removal (no caustic addition)

4.3 Impact of Packing Characteristics

The size of the packing can impact H_2S removal. Smaller size packing usually provides more surface area and better mass transfer, resulting in higher removal efficiencies. This is also demonstrated with the H_2S removal for this application with packing sizes ranging from #1 to #3.5. With a packing size of #3.5, the predicted H_2S removal efficiency is 55% while with a size of #1 the removal increases to 63%.

The downside of smaller packing, however, is that it can result in higher pressure drop through the after-condenser and it typically plugs with solids more easily. Significant pressure drop in the after-condenser is not desired since it could negatively affect the power production of the geothermal plant. In this case, the pressure drop ranged from only 0.02 to 0.07 psi for the #3.5 and #1 size packing, respectively.

4.4 Caustic Addition

Caustic can be added, if necessary, to the Unit 12 after-condenser to aid in H_2S removal and pH maintenance. It was assumed that there would be no H_2S in the feed water to the Unit 12 after-condenser because plant operating data suggests that most of the H_2S in the water from the main condenser is naturally oxidized. It was also assumed that the pH of the water would be in the 6.8

to 7.7 range, with the ammonia absorbed from the gas into the water balancing out the acidic H_2S oxidation products. The natural oxidation of H_2S in the cooling tower system can produce a variety of products, including elemental sulfur, thiosulfate ($S_2O_3^{2-}$), sulfite (SO_3^{2-}) and sulfate (SO_4^{2-}). It is unclear however what the balance of products will be, although it appears that sulfate and elemental sulfur are the two primary products. The products of the oxidation of H_2S are important because the presence of acidic components in the water to the after-condenser will result in reduced H_2S removal, and the acidic species must ultimately be neutralized by the addition of a base. The ammonia that is absorbed from the steam serves as the primary base source, although the addition of a small amount of supplemental base (caustic) may be required.

5. Conclusions and Path Forward

The results of the study indicate that bulk H_2S removal – on the order of 50% – can be achieved with the Unit 12 after-condenser using cooling tower water without caustic or iron chelate in a packed bed arrangement at the conditions assumed in this study. High feed water flow rate (>~1,150 gpm), cool feed water temperature (<~90°F), and smaller packing size (<#3.5) increases the H_2S removal efficiency to the required level.

The spray tower H_2S removal efficiency was estimated to be about the same level as the packed tower arrangement; however, spray towers are typically known to be less efficient than packing. The similarity of the results is likely due to the different calculation approaches used for the two types of cases and assumptions used. The spray tower estimates were based on data taken at the Unit 11 after-condenser; the data taken at Unit 11 included a small amount of caustic addition and the presence of a substantial amounts of salts (potentially representing buffering capacity), but were treated as "pure water" scrubbing results. Because of this, there is significant possibility that the spray tower estimates for Unit 12 may over-estimate the removal of H_2S.

Other factors such as H_2S being present in the feed water or the pH of the water being lower than expected (due to natural oxidation to sulfates in the cooling tower) could reduce the H_2S removal efficiency of the after-condenser. Calpine is able to add caustic to the after-condenser to help offset the potential negative impacts of these parameters.

Calpine is currently modifying the after-condenser to include 8 feet of packing. The after-condenser modifications will be completed in June 2018. Unit 12 will then be tested for operation without the Burner/Scrubber system and using only water (no caustic or iron chelate) for bulk H_2S removal in the after-condenser. Caustic will be added only as necessary to achieve 50% H_2S removal in the after-condenser.

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