

How Operation of a Wet Flue Gas Desulfurization System Can Impact the Wastewater Quality Generated

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ABSTRACT

The treatment of wet flue gas desulfurization (FGD) wastewater streams for removal of trace metals and contaminants is a topic that is gathering a lot of attention in the industry. Understanding the operating complexities of wet FGD systems can further our understanding of the wastewater quality generated. The intent is to provide the audience with a better understanding of wet FGD operating parameters that may have an adverse impact on the quality of wastewater being generated.

INTRODUCTION

In order to address outdated Effluent Limitation Guidelines (ELG) standards, the Environmental Protection Agency (EPA) released new proposed ELGs on April 19th, 2013. The proposed ELG rule was officially published in the Federal Register on June 7th, 2013. The EPA intends on finalizing the ELG rule in 2014. Dischargers must meet the effluent limitations within the next permit cycle beginning July 1, 2017. Direct dischargers of wastewater streams (i.e., those discharging directly to surface waters) must comply with effluent limitations in their National Pollutant Discharge Elimination System (NPDES) permits. It is through the NPDES program that compliance with the proposed ELG limitations will be enforced. The proposed

ELG's include limits on the following waste streams that were not previously regulated as separate streams (i.e., they were lumped under the low volume waste classification):

- Flue gas desulfurization systems
- Flue gas mercury control systems
- Gasification processes

Table 1 provides a summary of the discharge limits in the proposed effluent limitation guidelines. The limits shown here are those applicable to flue gas desulfurization (FGD) system waste waters. Limits for pH, total suspended solids (TSS), and oil / grease are set at the same levels as previously established in the existing ELGs. New limits for arsenic, mercury, nitrate/nitrite, and selenium have been introduced.

Table 1: ELG Limits Applicable to FGD Wastewaters

Pollutant	Maximum, any 1-day	30-day Average
Arsenic, total	8 ug/L (ppb)	6 ug/L (ppb)
Mercury, total	242 ng/L (ppt)	119 ng/L (ppt)
Nitrate/Nitrite, as N	0.17 mg/L (ppm)	0.13 mg/L (ppm)
Oil and Grease	20 mg/L (ppm)	15 mg/L (ppm)
pH, standard units	6 – 9	6 - 9
Selenium, total	16 ug/L (ppb)	10 ug/L (ppb)
Total Suspended Solids	100 mg/L (ppm)	30 mg/L (ppm)

Source: 40 CFR Part 423 (June 7, 2013)

ppm – parts per million

ppb – parts per billion

ppt – parts per trillion

Table 2 provides a summary of wastewater characteristics from a number of reported sources for the constituents that will be regulated under the new ELGs. There is a wide range in the reported data and the reported data is fairly limited compared to the total installed base of FGD systems. Because the data for the typical FGD wastewater characteristics was not all

inclusive, there are likely facilities with concentrations that fall outside of ranges presented in Table 2. Utilities will need to sample their wastewater sources for these constituents under various operating conditions in order to determine the actual impact on their facility.

Table 2: Typical FGD Wastewater Characteristics

Pollutant	Minimum	Maximum
Arsenic, total	58 ug/L (ppb)	5,070 ug/L (ppb)
Mercury, total	7,500 ng/L (ppt)	872,000 ng/L (ppt)
Nitrate/Nitrite, as N	1 mg/L (ppm)	270 mg/L (ppm)
Selenium, total	40 ug/L (ppb)	21,700 ug/L (ppb)
Total Suspended Solids	1,000 mg/L (ppm)	170,000 mg/L (ppm)

Sources:

- EPA 821-R-09-008, Steam Electric Power Generating, Point Source Category: Final Detailed Study Report
- Electric Power Research Institute (EPRI) 1012549, Treatment Technology Summary for Critical Pollutants of Concern in Power Plant Wastewaters
- Actual project observations

This paper will provide an overview of the wet FGD system operating parameters that impact the quality of wastewater generated. Key operating issues will be discussed including chloride levels, operating pH level, forced oxidation, inhibited oxidation, natural oxidation, the use of organic additives such as DBA or adipic acid, the use of mercury re-emission control agents, and the type of solids dewatering employed.

CHLORIDES

The quantity of wastewater produced by wet FGD systems is directly related to the fuel characteristics and makeup water quality. Chloride concentration is the primary characteristic evaluated when selecting materials of construction for wet FGD systems. The concentration of chlorides in the fuel source contributes the majority of the chlorides present in the FGD wastewater stream. The concentration of chlorides in the makeup water source is usually insignificant by comparison to the chloride concentration attributable to the fuel source. The wastewater blow down rate from the wet FGD system is usually based on maintaining a chloride concentration in the recirculating slurry below a level that is suitable for the wet FGD system materials of construction.

CHLORIDE IMPACT ON MERCURY SPECIATION. The EPA finalized the Mercury and Air Toxics Standard (MATS) applicable to electric generating units in April 2012. The final MATS standard requires utilities to comply with reduced filterable particulate matter,

hydrochloric acid gases, and mercury emissions. Per the final MATS standard most units must comply with a 1.2 lb/TBtu mercury emission rate. Compliance with the new mercury emission rates can be accomplished through a couple of mechanisms depending upon the speciated form of mercury present in the flue gas.

Mercury exists in one of three forms in the flue gas following combustion: elemental mercury, Hg^0 , gaseous oxidized mercury, Hg^{+2} , and particulate bound mercury, Hg_p . Elemental mercury is very difficult to remove from the flue gas stream. Particulate bound mercury is readily removed by electrostatic precipitators or fabric filter systems. Oxidized mercury is easily removed from the flue gas by wet FGD systems or by dry sorbent injection systems.

There are two primary mechanisms for mercury removal from the flue gas. Sorbent injection systems can be used to adsorb the mercury present in the flue gas onto a solid that is subsequently removed by the particulate collection device. The second method is to convert the elemental mercury into an oxidized species that becomes readily absorbed in the FGD system.

The presence of elevated chloride levels in the fuel source result in higher concentrations of oxidized mercury present in the flue gas. This higher mercury oxidation rate in the boiler in turn results in a better mercury removal rate from the flue gas in a wet FGD system.

Table 3 shows the mercury speciation rates for various plant configurations and fuel types.

Table 3: Typical Mercury Removal Rates and Percent Speciation

Bin Type	Fuel, Boiler, Emission control device(s)	Hg Removal	PERCENT SPECIATION		
			Particulate Hg	Oxidized Hg	Elemental Hg
0	Bituminous Coal, Coal Gasification	0.26%	0.51%	8.47%	91.02%
1	Bituminous Coal, PC Boiler with ESP-CS	29.13%	6.11%	68.20%	25.70%
2	Bituminous Coal and Pet. Coke, PC Boiler with ESP-CS	60.36%	1.17%	46.56%	52.27%
3	Bituminous Coal, PC Boiler with SNCR and ESP-CS	89.88%	20.32%	27.12%	52.56%
4	Bituminous Coal, PC Boiler with ESP-HS	10.65%	4.90%	57.84%	37.26%
5	Bituminous Coal, PC Boiler with PM Scrubber	12.07%	1.80%	19.51%	78.69%
6	Bituminous Coal, PC Boiler with Dry Sorbent Injection and ESP-CS	44.89%	0.16%	67.10%	32.74%
7	Bituminous Coal, PC Boiler with FF Baghouse	89.37%	3.98%	62.58%	33.44%
8	Bituminous Coal, PC Boiler with SDA/FF Baghouse	98.17%	9.17%	28.86%	61.97%
9	Bituminous Coal, PC Boiler with SCR and SDA/FF Baghouse	97.36%	5.06%	46.04%	48.90%
10	Bituminous Coal, PC Boiler with ESP-CS and Wet FGD	77.73%	0.22%	7.78%	92.00%
11	Bituminous Coal, PC Boiler with ESP-HS and Wet FGD	39.19%	0.63%	20.68%	78.70%
12	Bituminous Coal, PC Boiler with FF Baghouse and Wet FGD	97.39%	6.48%	33.00%	60.52%
13	Subbituminous Coal, PC Boiler with ESP-CS	2.65%	0.16%	30.83%	69.01%
14	Subbituminous Coal, PC Boiler with ESP-HS	0.00%	0.06%	12.52%	87.41%
15	Subbituminous Coal, PC Boiler with FF Baghouse	72.58%	1.49%	82.83%	15.68%
16	Subbituminous Coal, PC Boiler with PM Scrubber	0.00%	1.45%	5.11%	93.44%
17	Subbituminous Coal, PC Boiler with SDA/ESP	0.00%	0.32%	3.82%	95.86%
18	Subbituminous Coal, PC Boiler with SDA/FF Baghouse	23.30%	0.99%	4.35%	94.67%
19	Subbituminous Coal, PC Boiler with ESP-CS and Wet FGD	15.93%	0.43%	2.94%	96.63%
20	Subbituminous Coal, PC Boiler with ESP-HS and Wet FGD	8.03%	1.17%	4.46%	94.37%
21	Lignite Coal, PC Boiler with ESP-CS	0.00%	0.09%	3.62%	96.29%
22	Subbituminous Coal, Cyclone Boiler with PM Scrubber	22.15%	2.34%	5.75%	91.91%
23	Subbituminous Coal/Pet. Coke, Cyclone Boiler with ESP-HS	0.00%	0.93%	7.52%	91.55%
24	Lignite Coal Coal, Cyclone Boiler with ESP-CS	4.87%	0.04%	16.99%	82.97%
25	Bituminous Coal/Pet.Coke, Fluidized Bed Combustor with SNCR and FF Baghouse	91.82%	42.44%	27.87%	29.70%
26	Not Used				
27	Bituminous Waste, Fluidized Bed Combustor with FF Baghouse	99.89%	2.12%	38.81%	59.07%
28	Lignite Coal, Fluidized Bed Combustor with ESP-CS	40.36%	1.37%	11.64%	87.00%
29	Lignite Coal, Fluidized Bed Combustor with FF Baghouse	56.98%	0.42%	71.18%	28.40%
30	Antracite Waste, Fluidized Bed Combustor with FF Baghouse	99.75%	3.01%	37.30%	59.70%
31	Bituminous Coal, Stoker Boiler with SDA/FF Baghouse	93.66%	19.96%	17.94%	62.11%
32	Not Used				
33	Lignite Coal, PC Boiler with ESP-CS and FF Baghouse	0.00%	0.19%	64.49%	35.32%
34	Lignite Coal, PC Boiler with SDA/FF Baghouse	17.40%	0.36%	12.62%	87.02%
35	Lignite Coal, PC Boiler with PM Scrubber	32.77%	0.16%	2.98%	96.86%
36	Lignite Coal, PC Boiler with ESP-CS and Wet FGD	41.78%	0.82%	13.45%	85.74%
37	Bituminous Coal, Cyclone Boiler with Mechanical Collector	0.00%	18.75%	42.74%	38.51%
38	Bituminous Coal/Pet. Coke, Cyclone with ESP-CS and Wet FGD	50.48%	0.07%	11.30%	88.63%
39	Lignite Coal, Cyclone Boiler with SDA/FF Baghouse	38.24%	9.95%	17.07%	72.98%
40	Subbituminous Coal, Fluidized Bed Combustor with SNCR and FF Baghouse	52.52%	0.27%	3.42%	96.32%
41	Subbituminous Coal/Bituminous Coal, PC Boiler with ESP-CS	47.31%	0.88%	42.82%	56.30%
42	Subbituminous Coal/Bituminous Coal, PC Boiler with ESP-HS	34.03%	2.86%	49.11%	48.03%
43	Bituminous Coal/Pet. Coke, PC Boiler with FF Baghouse	0.00%	2.20%	78.41%	19.39%
44	Bituminous Coal/Subbituminous Coal, PC Boiler with FF Baghouse	68.22%	5.95%	42.10%	51.95%

Source: Bullock, D (2011)

Getting mercury concentrations down to the levels shown in Table 1 will require very high mercury removal rates (greater than 98 percent removal based on the mercury concentrations shown in Table 2).

As Table 3 highlights, the presence of a wet FGD system is not a guarantee of high flue gas mercury removal rates (see bins 10, 11, 19, 20, 36, and 38). Elemental mercury must be converted into an oxidized form in order for a wet FGD system to effectively capture mercury from the flue gas. Therefore, attention must be paid to the chloride levels in the fuel source(s) and the corresponding mercury oxidation rates leaving the boiler.

Sorbent injection systems can be employed to inject halogenated compounds (typically bromine

based compounds) onto the fuel to increase the percent of mercury oxidation in the boiler when the fuel chloride content is too low to achieve sufficient mercury oxidation. Fuel sources with higher chloride content will result in higher mercury concentrations in the FGD wastewater.

Alternatively, dry sorbent injection systems like activated carbon injection systems can adsorb mercury directly from the flue gas before the wet FGD system. It may be more economical and practical to capture mercury from the flue gas through use of a dry sorbent injection technology instead of trying to later remove mercury from the wet FGD system wastewater down to the ultra-low concentrations required by the proposed ELGs.

CHLORIDE CONCENTRATION IN FGD SYSTEMS. The blow down rate from wet FGD systems is usually set to maintain a predetermined chloride concentration within the recirculating slurry. The maximum allowable chloride concentration is determined by the materials of construction within the wet FGD system.

Operating the wet FGD system at lower chloride concentrations will result in blow down whereas higher chloride concentrations will result in less blow down generated. The size of the wastewater treatment system and its corresponding capital and operating costs are directly related to the blow down treatment rate. Minimizing the wet FGD system blow down results in higher concentrations of dissolved species that need to be treated. Higher concentrations of arsenic, mercury, nitrate, and selenium in the wet FGD blow down make it harder for the downstream wastewater treatment system(s) to achieve compliance with the proposed ELG limits.

In addition, chloride concentrations in wet FGD systems that produce a saleable grade gypsum byproduct are usually limited. It becomes increasingly difficult to wash the final gypsum product to the typical chloride concentration of less than 150 mg/L when the chloride concentration in the FGD blow down is high (i.e., greater than 20,000 mg/L).

Under-deposit corrosion mechanisms are impacted by elevated concentrations of aggressive anions, like chlorides. Acidic conditions are established beneath deposits as chlorides segregate to the shielded region beneath the deposit formation.

CHLORIDE LEVELS IMPACT TREATMENT OPTIONS. Some of the biological wastewater treatment options are influenced by elevated chloride concentrations in the wastewater to be treated. Elevated chloride concentrations will impact the selection of suitable bacteria for fixed film, biological reactors. Elevated chloride concentrations will also impact the selection of suitable plant species for constructed wetland systems. Dilution of the wastewater to lower the chloride concentration may be necessary before

treatment in a constructed wetland. However, the proposed ELGs appear to disallow dilution of wastewater streams as part of the treatment process.

The chloride concentrations in the wet FGD blow down do not significantly impact most physical / chemical treatment options. Elevated chloride concentrations can interfere with the absorption of metal species onto ferric hydroxide. Elevated chloride concentrations can also interfere with the softening reactions making it more difficult to achieve good metal removal rates in the metal hydroxide precipitation, iron co-precipitation, or metal sulfide precipitation processes.

In addition elevated chloride concentrations can impact the overall cost of the wastewater treatment systems as more expensive materials of construction are required to withstand the corrosive environment.

The operating chloride levels in wet FGD systems impact a number of issues beyond just the materials of construction used within the wet FGD system. A holistic approach needs to be considered when establishing the operating chloride concentration. For example, it may be more economical to line the existing wet FGD system with a higher grade alloy and operate at higher chloride concentrations to reduce the volume of wastewater blow down and subsequently the overall cost of the wastewater treatment system.

pH LEVELS

TYPICAL pH RANGES. The chemical processes involved in a wet FGD system are complex and not fully developed here for simplicity. In summary, an alkaline material such as limestone or lime is fed to react with dissolved SO₂ to form insoluble salts of calcium sulfite or calcium sulfate which are precipitated from the recirculating FGD slurry.

The pH in the wet FGD system reaction tank is usually maintained between 5.2 and 6.2. The equilibrium pH for the dissolution of calcium sulfite is approximately 6.3. Operating the FGD system at a pH higher than 6.3 would result in

undesirable scaling within the system due to sulfite precipitation.

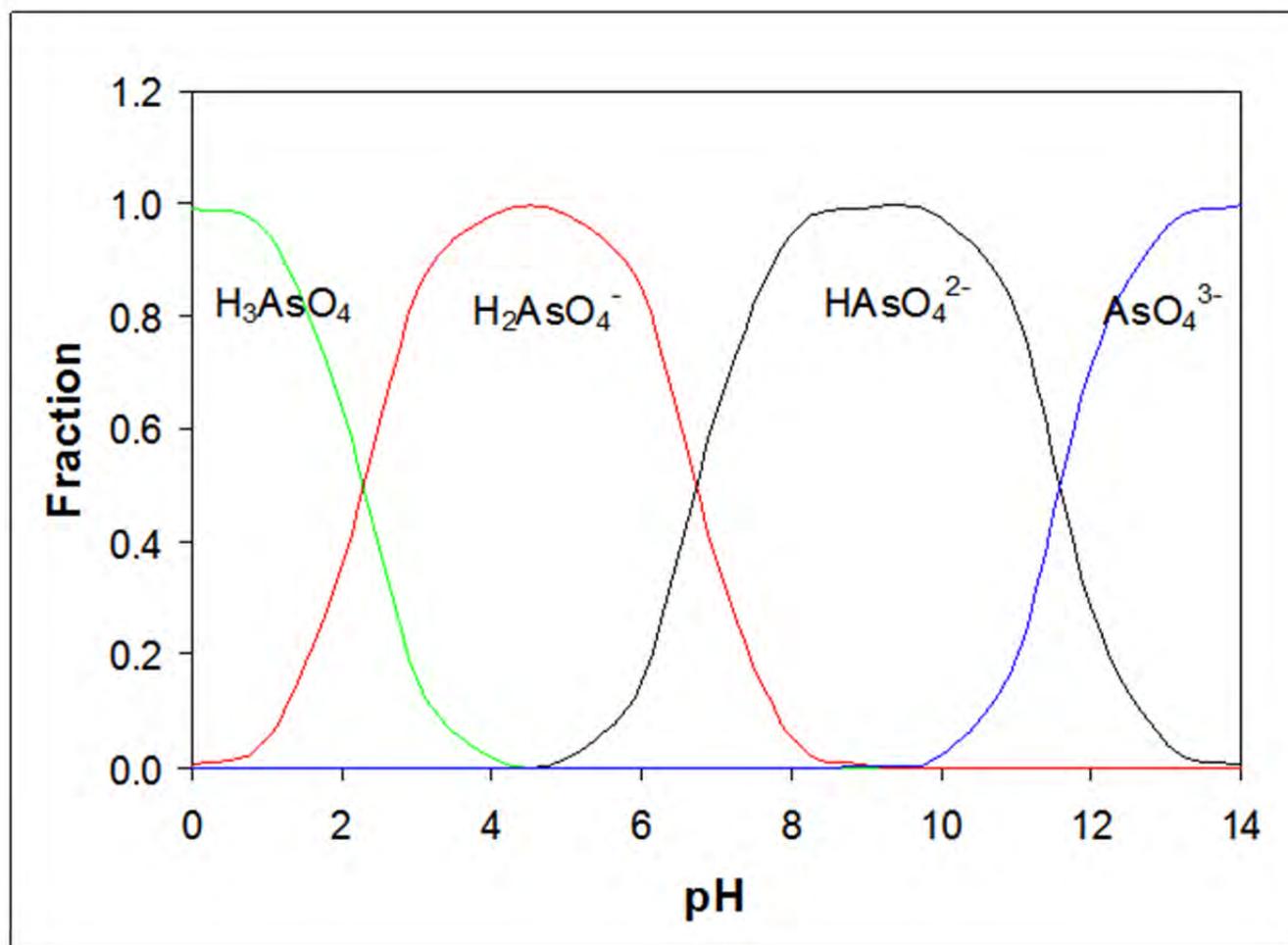
The SO₂ removal rate in the wet FGD system is generally higher with excess lime or limestone feed at a high operating pH and lower with little excess lime or limestone at a low pH. Lower pH at any point in the system results in lower SO₂ removal rates. The pH level at the wet FGD system inlet is also an indicator of excess lime or limestone in the system; with a higher pH indicating more excess lime or limestone available for reaction with SO₂.

As the wet FGD slurry recirculates through the system, the drop in slurry pH depends on the extent to which the lime or limestone dissolves and replenishes the alkalinity of the solution.

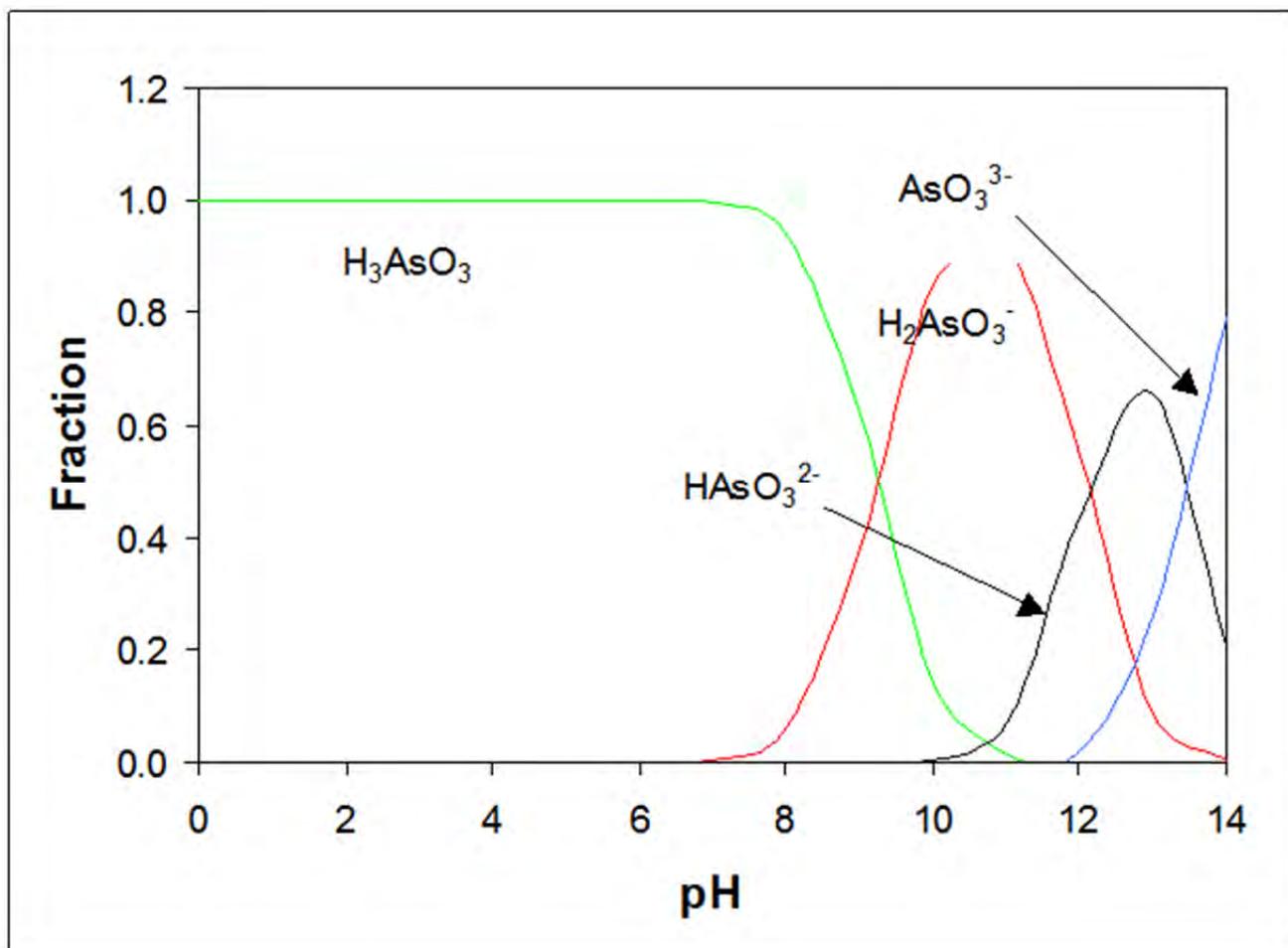
Therefore, it is both undesirable to let the pH of the recirculating slurry get too high (> 6.3) or too low (< 5.0). At elevated pH values sulfite precipitation and scaling will occur. At low pH values, not enough alkalinity is present in the system to react with the dissolved SO₂ and the system will be unable to achieve the desired SO₂ removal rate.

pH RELATIONSHIPS. Understanding the impact of pH on the form of each individual species is essential to determine the best method of removal. Various species take on different forms as the pH changes. Removal of one species may be improved at lower pH while removal for another species may get worse. As an example, Figures 1 and 2 show the effect of pH on the form of Arsenic (V) and Arsenic (III).

Figure 1: Acid-Base Chemistry of Arsenic (V)



Source: Chwirka, J (2005)

Figure 2: Acid-Base Chemistry of Arsenic (III)

Source: Chwirka, J (2005)

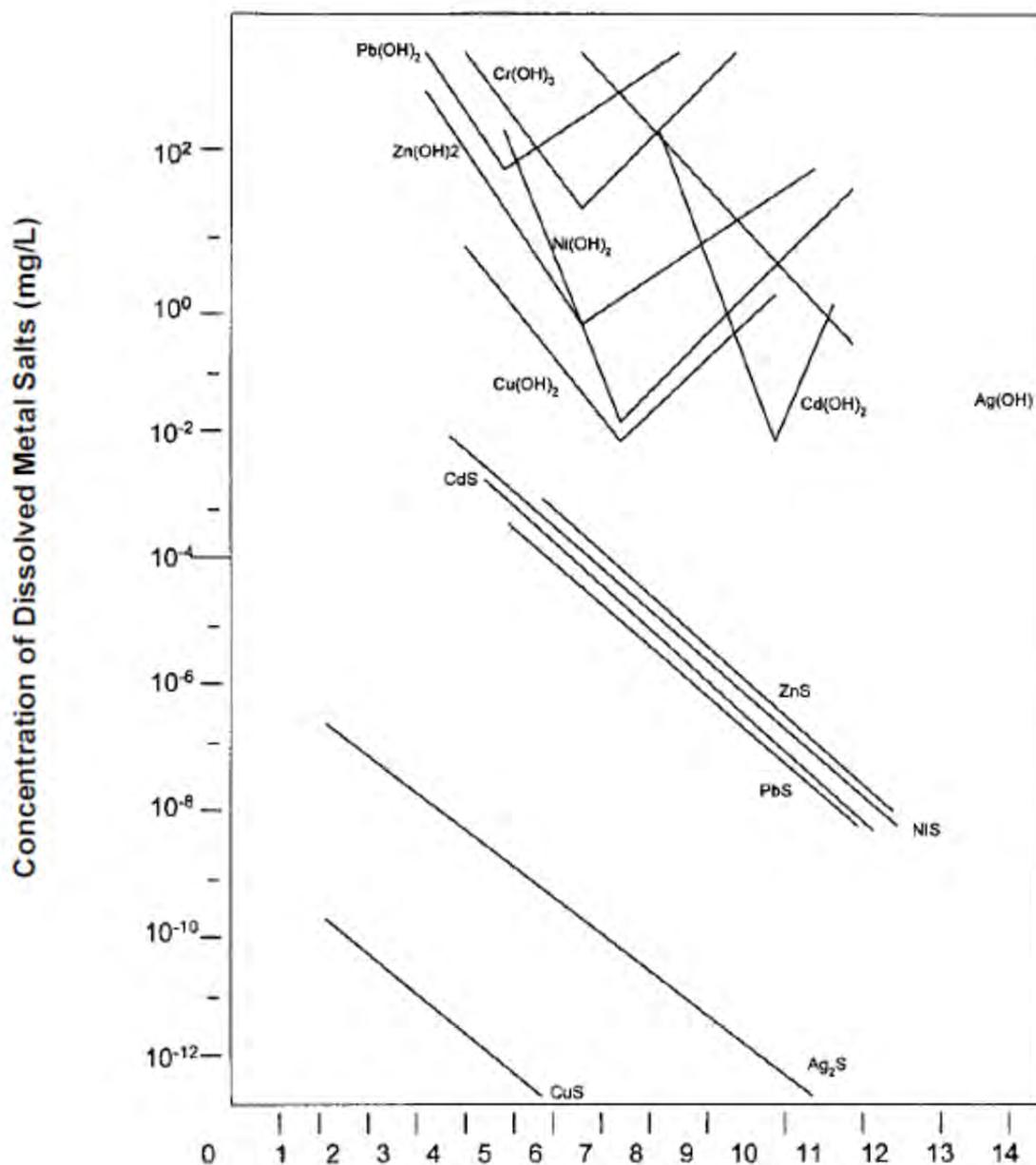
Based on the typical operating pH for wet FGD systems, the most anticipated form of arsenic is either $H_2AsO_4^-$ or H_3AsO_3 .

Free available alkalinity in the form of hydroxide ions can form insoluble metal complexes which precipitate from solution. Figure 3 shows the relationship between various metal hydroxides and metal sulfide species. In general, the metal sulfides are much less soluble than the metal hydroxides and the solubilities decrease with increasing pH concentration. For comparison, the solubility of mercury hydroxide is about 3.9×10^{-4}

and the solubility of mercury sulfide is about 9.0×10^{-20} .

Also, it is interesting to note that all of the metal hydroxides decrease in solubility up to a maximum pH value, after which their solubility increases with increasing pH. The point of minimum solubility differs for each metal hydroxide species.

Figure 3: Solubility of Metal Hydroxides and Sulfides as a Function of pH



Source: Industrial Environmental Research Laboratory (1980)

OXIDATION

TYPES. There are three types of oxidation systems utilized with wet FGD systems. The most common of these is forced oxidation where air is injected into the reaction tanks and effectively converts the calcium sulfite into calcium sulfate (gypsum) which can then be dewatered and sold as a marketable byproduct. Forced oxidation systems can achieve nearly 100 percent oxidation of the sulfite to sulfate.

Natural oxidation is similar to forced oxidation except that air is not manually introduced into the reaction tanks. Some oxidation of sulfite to sulfate occurs in the gas-liquid contact zone of the wet FGD system from the oxygen present in the flue gas. Natural oxidation can achieve oxidation rates as high as 30 to 50 percent.

Inhibited oxidation is an alternative to natural or forced oxidation systems. In the inhibited oxidation process, emulsified sulfur, or sodium

thiosulfate, is added to the reagent feed tank. The addition of the emulsified sulfur controls the sulfite oxidation rate to less than 15 percent. This helps control scaling and pluggage from forming in the wet FGD system. The dewatered solids from the inhibited oxidation system must be blended with fly ash and lime to form a stable solid that can be disposed of in a landfill.

Operating in an inhibited oxidation mode may cause re-emission of mercury from the FGD slurry due to reduction of oxidized mercury to its elemental state in the slurry. Re-emission of mercury into the flue gas is undesirable because of the impact this has on the facilities ability to comply with the MATS requirements that have been recently finalized by the EPA requiring mercury emissions of less than 1.2 lb/TBtu.

Conversion of an existing natural or forced oxidation system to an inhibited oxidation system would require some modifications to the dewatering equipment. Conversion to an inhibited oxidation system could potentially eliminate the need for a wastewater treatment system as the wastewater blow down could be blended with fly ash and lime and landfilled. The cost of the conversion to inhibited operation would have to be weighed against the potential lost revenue from fly ash sales and/or gypsum sales to determine the most economical solution.

ORGANIC ADDITIVES

Organic additives are sometimes used to help improve the overall wet FGD system SO₂ removal rate. Some of the more common organic additives include: formic acid, acetic acid, adipic acid, succinic acid, and dibasic acid.

Organic additives function as a buffer to control the hydrogen ion concentration in the gas-liquid contact zone which in turn helps maximize the SO₂ absorption rate. Because the organic additives function as pH buffers in the system and help improve SO₂ removal rates, the immediate impact is an overall increase to the amount of calcium sulfite or calcium sulfate produced by the system.

Impacts on the downstream wastewater treatment systems are not fully known at this time. However, it is anticipated that additional chemical feed(s) will be required in the downstream wastewater treatment systems to overcome the pH buffering capacity of the organic additives leading to higher operating costs. The impacts of organic additives on the solubility of various metal hydroxides or sulfides and the impact on the proposed ELGs need further research.

MERCURY RE-EMISSION ADDITIVES

The revised MATS standard requires most units to comply with a 1.2 lb/TBtu mercury emission rate.

Recent testing efforts have determined that small quantities of oxidized mercury are able to be reduced back to elemental mercury within the wet FGD systems. The elemental mercury is then re-released into the flue gas stream reducing the overall mercury emission removal rate. These small quantities of mercury that are reintroduced into the flue gas are enough to potentially cause a facility to violate the stringent 1.2 lb/TBtu mercury emission rate required by the revised MATS standard.

To counter this phenomenon, chemical additives have been used to maintain the mercury in a soluble / oxidized form in the wet FGD slurry. The primary impact of these chemical additives is the net increase in the total mercury concentration that must be treated in the wet FGD wastewater stream. The use of these mercury re-emission additives are relatively new and further research is needed to determine the impact of these chemicals on the proposed ELGs.

SOLIDS DEWATERING

Wet FGD systems typically have primary and secondary dewatering systems. Primary dewatering usually consists of hydrocyclones. The hydrocyclone overflow is returned to the reaction tank and the underflow is processed by the secondary dewatering equipment.

Secondary dewatering equipment typically consists of rotary drum vacuum filters or horizontal belt vacuum filters. Rotary drum vacuum filters are generally less expensive and require less physical space. Rotary drum filters are able to dewater solids to about 80 to 90 percent solids. Horizontal belt vacuum filters can dewater solids to as high as about 95 percent solids.

In order to minimize the impact on the wastewater treatment system, operations should consider use of rotary drum vacuum filters and operating at around 85 percent solids. This results in more wastewater entrained in the solids disposed of in the landfill and reduces the overall amount of wastewater that must be treated in the wastewater treatment system.

Plants that currently sell their gypsum product will need to evaluate the cost of the new wastewater treatment system versus operating at a lower percent solids concentration and landfilling the material instead of selling the gypsum product.

Purge water from the wet FGD system is typically taken from the filtrate tank after the secondary dewatering equipment. The percent solids in the filtrate tank are typically 1 to 3 percent solids but can be as high as 5 percent solids. This corresponds with 10,000 to 50,000 mg/L of suspended solids. The wastewater treatment system will need to be designed to treat and remove these suspended solids to achieve compliance with the proposed ELG limitation of 30 mg/L or less on average.

CONCLUSIONS

Multiple operating aspects of a wet FGD system must be considered and evaluated in conjunction with the addition of any new wastewater treatment system for compliance with the proposed ELGs.

Some of the key areas for consideration include:

- Installation of ACI system for mercury capture prior to the wet FGD system.
- Installation of halogen feed system for the fuel source to convert more elemental mercury into an oxidized form.
- Conversion of wet FGD system to inhibited oxidation and elimination of wastewater blow down.
- Testing of organic additives and mercury re-emission additives to determine impact these chemicals may have on any downstream wastewater treatment system.
- Operating the secondary dewatering equipment at a lower percent solids concentration so more wastewater is leaving the facility with the dewatered solids.

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