Effect of Flue Gas Oxygen Content to Gas-Gas Heater Requirement in Limestone Forced Oxidation Desulfurizer System

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Abstract. To comply the regulation, the sulfur dioxide (SO$_2$) concentration should be reduced to designated standard of emission which was stated in the regulation. It causes many coal fired power plant (CFPP) need to procure a new or upgrade their desulfurization equipment. In this study the desulfurization method focused on the usage of Limestone forced oxidation (LSFO). The study is based on subcritical 300 MW CFPP and the specification of coal for the calculation was taken from coal properties of Palu 3 CFPP. There are two cases and two configurations in this study. The first case is flue gas oxygen content of 3.5% and the second is 7%. Then in the configuration of with or without gas-gas heater (GGH). The oxygen content condition in the flue gas have an influence to the presence of GGH within LSFO desulfurizer system. The lower of oxygen content in the flue gas then increase the tendency to use GGH.

1. Introduction

Electricity power generation within coal-fired power plant (CFPP) generates a byproduct of Flue Gas Emission which will be exposed to the environment. This Flue Gas Emission is composed by carbon dioxide, nitrous oxides, particulates, and sulfur dioxide. Excessive SO$_2$ concentration in flue gas can cause acid rain phenomenon in the local surrounding area of CFPP. Thus, the concentration limit of SO$_2$ in flue gas is regulated by the Indonesian Government in the newly adopted Ministry of Environment and Forestry Regulation No 15 year 2019 regarding Power Plant Flue Gas Emission that supersedes Regulation No. 21 year 2008. For power plant that operate before 2019 the maximum SO$_2$ concentration is 550 mg/Nm$^3$ and for after 2019 is 200 mg/Nm$^3$ (Dry basis, 7% O$_2$)

In order to comply the regulation, the SO$_2$ concentration should be reduced to designated standard of emission which was stated in the regulation. Since coal fired power plant still dominating in Indonesian electricity [1], many CFPP need to procure a new or upgrade their desulfurization equipment. However, upgrading desulfurization is not easy at all. For example, before this regulation take a lead several power plants are not installed that system due to comply the previous regulation. Various desulfurization methods were already used around the world CFPP. Numerous study conducted to compare how is the efficiency, sorbent typical, until the cost estimation of each method [2][3]. In this study the desulfurization method will focus on the usage of Limestone forced oxidation (LSFO) that implemented with the intention of reducing SO$_2$ concentration in the flue gas. The main equipment of LSFO is absorber, it conducts a desulfurization reaction between the flue gas and limestone slurry. Besides absorber, LSFO sometimes using gas-gas heater (GGH) to exchange the heat between flue gas inlet and outlet. However, not all power plant engineer knows what are the parameters which influence the GGH requirement. This condition made a misunderstanding in
designing LSFO system, So this study have a purpose to give a reference to the reader what is the condition that recommend LSFO to use GGH.

2. Methods

This study has several steps to explain how the variable influence the design. There are two things that used to estimate the result, those are heat & mass balance and psychrometric. Based on the variables that stated in this section then processed to the calculation in section three. Begin with flue gas entered the LSFO, calculate the desulfurization process, define the properties of each stream, and compared the the result of every variable. The variables were explained by several following points.

2.1. Coal basis

The specification of coal for the calculation was taken from coal properties of Palu 3 CFPP (2x50 MW). Two analysis were given to determine the quality and rank of the coal. With the proximate and ultimate analysis of its, the calorific value of the coal could be determined by Dulong formula [4]. The coal high heating value (HHV) was determined in the value of 4,159.46 kcal/kg. This coal could be classified as lignite or brown coal.

2.2. Cases

The calculation was based on subcritical 300 MW pulverized coal (PC) fired power plant. The total air to boiler was basically the sum of primary air and secondary air, where Force draft (FD) fan(s) supplied the secondary air for combustion. The FD fans must supply secondary air in sufficient quantity to achieve the intended excess air at the boiler outlet. PC boilers typically operated with 15 to 20% excess air at full load, and with higher excess air as load is reduced. The combustion product calculation was assuming on full conversion reaction mass balance [5]. The basis of combustion calculation was plant heat rate of 2,647 kcal/kWh and the coal consumption of 190.91 ton/h.

In this calculation the assumption was given at two cases with different oxygen content in the flue gas entering the LSFO. The first case combustion calculated for normal excess air with oxygen content of 3.5% and the flue gas mass flow (dry condition) was 1,339.76 t/h. The second case, the flue gas containing 7% oxygen and the mass flow is 1,654.34 t/h. From flue gas output the absolute humidity could be obtained by dividing with the mass of moisture. For case 1 the absolute humidity is 92.22 g/kg dry and case 2 was 74.68 g/kg dry. The absolute humidity of flue gas from both cases, before entering the absorber would give the different of heat transfer characteristic.

2.3. Configuration types

Flue gas from the Electrostatic Precipitator (ESP) was drawn by Induced Draft (ID) fan (and sometime use booster fan too) into LSFO system to control the SO\textsubscript{2} content of the flue gas to meet the emission regulation. The LSFO main equipment were Absorber and Gas - Gas Heater (GGH). Flue gas would be contacted with reagents inside the absorber. The reagent which used in LSFO system was limestone that dissolved in water or can be called as limestone slurry. A cleaned exit flue gas was discharged from the absorber and gypsum would be produced as the by-product of the desulfurization reaction [6]. Assuming there was no flue gas bypassing the LSFO system.

The flue gas handling design from the exit of the absorber to the stack discharge was an important consideration in LSFO system. Flue gas left the absorber at near the adiabatic saturation temperature and includes some entrained liquid or slurry. Because the flue gas was saturated with water vapor, surface condensation is inevitable [7]. The acidic droplet might be occurred at the stack exit that gave bad effect to environment. To minimize the acidic droplet occurred in the chimney outlet, the regenerative heat exchanger (gas – heater) might be added at the inlet and outlet absorber. The gas – gas heater will transfer heat from the hot flue gas inlet to the cooler flue gas outlet.

The configuration of the absorber system with and without gas - gas heater is used depending on the overall power plant system design and the ambient condition of the power plant. This paper would study when the gas – gas heater is required. The typical LSFO without heat exchanger is shown in figure 1, while figure 2 will show the typical LSFO system with GGH. Stream 1 and 2 are explained as flue gas enter and out the LSFO system. Then stream 1A and 2A are explained as hot and cold flue
3. Result and discussion

3.1. Absorber system inlet

With sulfur content at coal input of 0.50 % as received basis, the boundaries of the absorber system is depicted in the figure 1 and figure 2. The parameter for the absorber inlet is shown in table 1 below.

| Absorber Inlet Parameter | Case 1 | Case 2 |
|--------------------------|--------|--------|
| O₂ content (% dry)       | 3.50   | 7.00   |
| Absolute humidity (g/kg dry) | 92.22 | 74.69 |
| Relative Humidity (%)    | 4.18   | 3.47   |
| Flue Gas Inlet Temperature (°C) | 135   | 135   |

The parameter difference between case 1 and case 2 were the oxygen content in the flue gas. The variation of the O₂ dry percentage on the case 2 was twice as much as the case 1. The extreme value difference of the O₂ dry percentage would show the effect of the addition of gas-gas heater to the LSFO system because the initial moisture content percentage in the flue gas also have effect to the LSFO design consideration.

3.2. Desulfurization

The main part to reduce SO₂ content in the flue gas was the desulfurization reaction itself. The basic reaction of desulfurization in LSFO can be seen in following equation:

$$\text{CaCO}_3 + \text{SO}_2 \rightarrow \text{CaSO}_3^- + \text{CO}_2$$

Limestone or calcium carbonate reacted with SO₂ and producing CaSO₃ and CO₂. CaSO₃ or calcium sulphite then oxidated to form gypsum crystal (CaSO₄.2H₂O). O₂ or oxygen was obtained from air that injected to absorber bottom. This air was called oxidation air.

With refer to flue gas combustion mentioned in subsection 2.2, the SO₂ concentration for both cases were in the number of 1,419.71 mg/Nm³. To meet the regulation for power plants which have been operated before 2019, plant should reduced the SO₂ to 550 mg/Nm³. With the stoichiometric
calculation [5] and 95% purity of limestone. After the limestone required was calculated, the oxidation air mass flow based on CaSO$_3$ oxidation reaction was calculated. The oxidation air needed was 1,246.81 kg/h. Note this oxidation reaction is using 0% excess air, then the additional nitrogen from the air was carried by the flue gas since did not react with any compound in the absorber.

### 3.3. Heat balance

To define the condition of each stream of LSFO system heat balance calculation should be conducted. Assuming the heat transfer of absorber and GGH were the same in each case and configuration which used these. Then from heat balance, estimated the stream condition by psychrometric. Since the properties of dry flue gas was relatively same with air (Nitrogen content almost same), so dry flue gas psychrometric was refer to air.

#### a. Without gas-gas heater

Exhaust flue gas exchanged heat with limestone slurry in the absorber. Transferred heat was calculated using $C_p$ of fluid. The formula is given below [8]:

$$ C_p = a + b(T) + c(T)^2 + d(T)^3, \quad (2) $$

While for the heat exchanged was calculated using following equation:

$$ \dot{Q} = \dot{m} \int_{T_0}^{T_1} C_p \, dT \quad (3) $$

The coefficient of a, b, c, and d of each component that mentioned in equation (1) could be seen in Himmelblau et al [8]. Using equation (2) to define the enthalpy that carried by the stream, the enthalpy of flue gas entered the LSFO system for case 1. Assuming adiabatic process for the heat transfer between flue gas and limestone slurry, hence there was no heat loss or leakage during desulfurization. From psychrometric theory, this phenomenon lead flue gas to move adiabatically approaching the saturated line or its wet bulb temperature. It was need to know that there was no change in wet bulb temperature of flue gas enter and leaving the absorber since it was assumed for perfect adiabatic process [9].

Then assuming the flue gas temperature leaving the absorber (stream 2) was $62^\circ$C for case 1. The number of heat transferred could be estimated by looking at the difference between the enthalpy of stream 1 and 2. In case 1, the dry flue gas heat transfer was 98,768.82 MJ/h. this heat transfer was being the maintained parameter for the next case. By using this, the next case flue gas outlet temperature could be defined. The case 2 was calculated with the same step likes case 1, but the heat transfer was defined before the exit temperature.

After all cases was calculated, define the absolute humidity and relative humidity by psychrometric [10]. The calculation result of energy balance for case 1 and case 2 provided significant result (see table 8). Flue gas containing heat entered absorber and contacts with limestone slurry which form of droplets. Heat transfer and reaction occured followed by drop in temperature of flue gas leaving absorber with formation of CaSO$_3$ in the absorber. Mass transfer also took place in this process and lead to increase of flue gas humidity. Table 2 shows that less excess of O$_2$ has higher relative humidity (RH). Its mean case 1 has higher probability to form plume after flue gas leaving the stack.

| Stream 1                  | Case 1 | Case 2 |
|---------------------------|--------|--------|
| Temperature (°C)          | 135.00 | 135.00 |
| Absolute humidity (g/kg dry) | 92.23  | 74.69  |
| Relative Humidity (%)     | 4.18   | 3.47   |

| Stream 2                  |        |        |
|---------------------------|--------|--------|
| Temperature (°C)          | 62.00  | 76.12  |
b. With gas-gas heater

Notice that the calculation result on table 2 for case 1 has high value of relative humidity, thus in the practice the flue gas leaving absorber must be heated until certain value. Heated flue gas must be dryer or has less relative humidity to avoid plume formation while contacted with ambient air. However, this application required additional cost for such system. This problem could be solved using heat integration by connecting flue gas before entering absorber with flue gas leaving absorber in one heat exchanger, well known as Gas-Gas Heater (GGH). The phenomenon of fluids in GGH called as sensible cooling or heating. This phenomenon could be illustrated by horizontal movement in psychrometric chart where there was no change in dew point and absolute humidity value between GGH inlet and outlet.

Begin with case 1, the same condition of flue gas inlet the LSFO system, GGH would cool it until the temperature of 120°C. Enthalpy of the stream 1 and 1A were calculated same as equation (1) and (2) then the heat transfer could be defined. The heat transfer of GGH was based on wet basis due to all of component release and received heat without phase change. So, the GGH wet basis heat transfer for case 1 was 24,013.98 MJ/h. After passing GGH, the flue gas entered the absorber (stream 1A) that contacted with limestone slurry and would be cooled adiabatically like explained in previous configuration. With the same absorber dry heat transfer, the flue gas leaving the absorber (stream 2A) was in temperature of 46.69°C before went to stack, stream 2A received heat from stream 1 in GGH. By assuming there was no heat loss in GGH, the temperature of stream 2 could be estimated in the number of 62.51°C.

The distribution of absolute and relative humidity for case 1 with GGH was totally different against the without one. Then recalculated for case 2 with similar wet heat transfer within GGH and similar dry heat transfer within absorber. The result can be seen in table 3.

Application of Gas-Gas Heater also gave impact on absorber performance toward the exchanged fluids as shown in table 2 and 3. It shows that, for case 1, flue gas reached its saturation condition because the inlet temperature was lower thus the capability of fluid to absorb heat also reduced. On the other hand, while the oxygen content was higher, fluid did not reach its saturation temperature. For case 1, the water content in flue gas leaving absorber would be lower than without GGH because some waters were condensed.

### Table 3. Flue gas properties with GGH

| Parameters                  | Case 1          | Case 2          |
|-----------------------------|-----------------|-----------------|
| Absolute humidity (g/kg dry) | 128.70          | 102.92          |
| Relative Humidity (%)       | 78.99           | 35.37           |

3.4. Plot on Psychometric chart
To facilitate reader how the flue gas condition moved due to adiabatic cooling, sensible cooling and heating, author illustrated with psychrometric schema for all cases. The illustration can be seen in figure 3. Note that ambient condition used in this study are 30°C and 80% relative humidity. In the case 1, the present of GGH would reduce the water content in the flue gas significantly (see figure 3 (ii)). It happened because water condensation was occurring in absorber. It was proven there was absolute humidity drop between stream 1A and 2A (see table 3). It occurred because the flue gas heat release to slurry was more than the heat release to reach saturation condition or 100% relative humidity. So, the movement continued inline with saturation condition and condensed some water content. For case 1 without GGH configuration (figure 3 (ii)), it was seen that the flue gas in stream 2 would cross the saturation line when it mixed with ambient air (represented by black line). Then white plume would be happened. Different from with GGH configuration that tended to be safe from white plume phenomenon. So, for the case 1, GGH was required to prevent white plume. For case 2, there was no significant effect in the use of GGH. Either with or without GGH the flue gas condition tended to be safe from white plume phenomenon.

4. Conclusion

The oxygen content condition in the flue gas have an influence on the presence of gas-gas heater (GGH) within limestone forced oxidation (LSFO) desulfurizer system. The lower of oxygen content in the flue gas then increase the tendency to use GGH. The oxygen content in the flue gas inversely proportional with the value of absolute humidity. When the oxygen content is low, the absolute humidity will relatively high that indicate the flue gas containing more water and finally increase the tendency to form white plume in the chimney outlet. So GGH will be required. This study answered one of the aspects why there are some CFPP’s LSFO using GGH and some are not.

Environmental engineer especially in designing LSFO shall be consider the probability of the lowest flue gas oxygen content. It can be happened when boiler operate with minimum air-fuel ratio (AFR) or there are some failures in combustion air supply. Otherwise, when boiler operate with excessive combustion air it will result high oxygen content in flue gas and lessen the tendency to form white plume. But in other hand the boiler efficiency will decrease, and the power needed to operate draft fan is higher.

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