

Dry Flue Gas Desulfurization for Power Generation

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Abstract

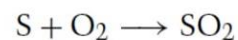
Sulfur Dioxide (SO₂) emission in Power Plant flue gas is one of the controlled emission by Ministry of Life and Environment with maximum concentration of 600-800 mg/Nm³ for various power plants. Such limit will be exceeded by Coal Fired Power Plant that utilizes coal with sulfur content of more than 0.4-0.6% weight, or Diesel Power Plant that Utilizes Marine Fuel Oil (MFO) fuel with sulfur content up to 2-5% weight. One of the most effective method of Flue Gas Desulfurization (FGD) for coal fired power plant is by using Wet Limestone Forced Oxidation System (LSFO). However, the investment cost of LSFO FGD is prohibitively expensive, and retrofit to existing pulverized coal fired power plant is virtually impossible due to its space requirement. This paper explains an alternative FGD systems that is less effective, but much more suitable for retrofit purposes, which is Dry Flue Gas Desulfurization, using Natrium Bicarbonate (NaHCO₃). The effectiveness of reagent, and application to Coal Fired Power Plant and Diesel Power Plant will also be discussed.

Keyword : Flue Gas Desulfurization, Combustion, Pulverized Coal Power Plant, Sulfur Dioxide emission, Diesel Power plant, Marine Fuel Oil utilization

I. Introduction

Pulverized Coal Fired Power Plant (CFPP) is the major source of electricity in Indonesia. Coal is used as fuel because of its low cost per unit of energy compared to other sources of fuel. Marine Fuel Oil (MFO) is another fuel that is widely used alongside High Speed Diesel Fuel (HSD) that is used for diesel engine power generation.

Power generation using Coal and fuel oil will generate Sulfur Dioxide (SO₂) that is diluted in the flue gas. SO₂ gas is a product of burning (oxidizing) Sulfur contained in the fuel, with following reaction:



High concentration of SO₂ in the environment can form acid rain that will affect the local area. Ministry of Life and Environment (MOLE) regulates the flue gas Emission of power plant by regulation No. 21 Year 2008 as Follow:

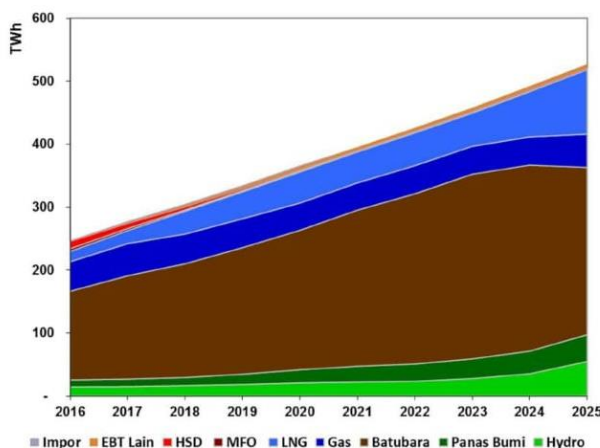


Figure 1: Indonesia Energy Mix Forecast [7]

Table 1. Indonesia Powerplant Emission regulation

Parameter (24 Hour Average)	Coal Powerplant		Diesel Powerplant	
	Before 2008	After 2008	Before 2008	After 2008
SO ₂ (mg/Nm ³)	750	750	800	600
NO ₂ (mg/Nm ³)	850	750	1000	1000
Particulate (mg/Nm ³)	150	100	150	120
CO (mg/Nm ³)	-	-	600	540
Opasitas	20 %	20%	20%	20%
Koreksi terhadap kadar O ₂ (Selain Opasitas)	7%		13%	

Such limit will be exceeded by Coal Fired Powerplant that utilizes coal with sulfur content of more than 0.4-0,6% weight, or diesel power plant that utilizes MFO fuel with sulfur content up to 2-5% weight. Such as that the sulfur content in fuel that dictates the SO₂ concentration in flue gas is a crucial parameter that need to be carefully controlled.

II. Energy Conversion by combustion

Conversion of energy of Coal and MFO is done by combustion, which is essentially an oxidation reacton to release heat. A reference of the combustion process is sub bituminous coal with medium sulfur content (0.9-1.1% weight), that is consumed by a powerplant that also utilize Wet Limestone Forced oxidation (LSFO) Flue Gas Desulfurization system to comply with emission regulation. The power plant is a subcritical type with nett power production of 660 MW per Unit, and a thermal efficiency of 36-37%. Table II explains the mass and stoichiometry ratio of combusting 100 kg of coal typical to with the following conditions:

- Actual O₂ content in flue gas of 3,3% causing excess air of 21% above stoichiometry (wet basis)
- Calculated Excess Air is based on 7% O₂ content in flue gas, 55% Excess Air.
- Flue gas mass of 1.225 kg/m³

Total actual flue gas mass generated by combusting 100 kg of Coal is 1041 kg of Flue Gas, which amounts to an actual SO₂ concentration of 874mg/Nm³ in Flue gas. But when calculated for emission regulation, the O₂ is increased to 7%, which amounts to 1258 kg of flue gas / 100 kg of fuel, yielding an SO₂ gas concentration of 752 mg/Nm³. These

figures are within the borderline of emission regulation. The theoretical composition of flue gas is explained in Table 2

Table 2. Combustion of typical sub-bituminous coal

REACTANT = 100 kg Fuel Basis				
Species		Ultimate Analysis	M	N
Fuel = As Received		Mass (kg)	molar mass (kg/kmol)	mol (kmol)
C	Carbon	58.50	12	4.88
H	Hydrogen	4.70	1	4.70
O	Oxygen	12.51	16	0.78
S	Sulfur	0.57	32	0.018
N	Nitrogen	1.00	14	0.071
	Ash Content	4.48		
H ₂ O	Moisture	18.24	18	1.01
Total Fuel		100.0		11.46
Stoichiometric Air		779.6	28.84	5.68
Excess Air		378.9	28.84	2.75
Total Reactant		1258.5		17.14
PRODUCT				
Species		Ultimate Analysis	M	N
(Exhaust Gas)		Mass (kg)	Molar mass (kg/kmol)	mol (kmol)
CO ₂	Carbon Dioxide	214.5	44	4.88
H ₂ O	Moisture	60.5	18	3.36
SO ₂	Sulfur dioxide	1.1	64	0.018
N ₂	Nitrogen	889.6	28	31.771
O ₂	Oksigen	88.3	32	2.76
	Ash Content	4.48		
Total flue gas + Excess Air		1258.5		42.79

Furthermore, for 660 MWh the powerplant produces, 2995,57 kg of SO₂ gas is produced.

III. Wet Limestone FGD as Baseline

In the powerplant in discussion, the SO₂ in flue gas is captured using Wet Limestone Forced Oxidation (LSFO) Flue Gas Desulfurizer (FGD) system with the principle of binding SO₂ in flue gas with CaCO₃ slurry, originating from limestone. The reaction absorber is located between the Electrostatic Precipitator (ESP) and Chimney as follow:

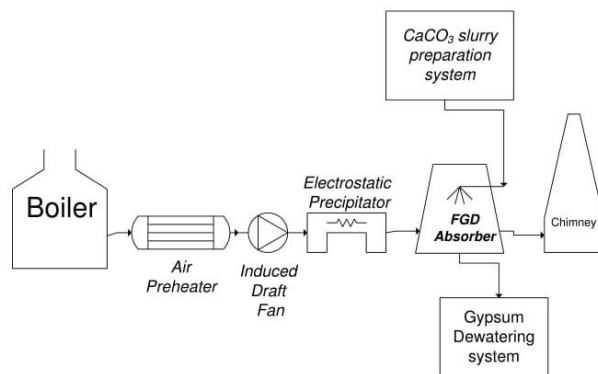


Figure 2. Wet Limestone FGD Absorber Location [3]
To enable desulfurization reaction, the limestone is pulverized and mixed with water to make a slurry with composition of 20-30% limestone, and is introduced with the flue gas with a Ca:S molar ratio of 1,1:1. For 660 MW CFPP, height of the absorber is approximately 30 m, and 10 m in diameter, as depicted in figure 3.

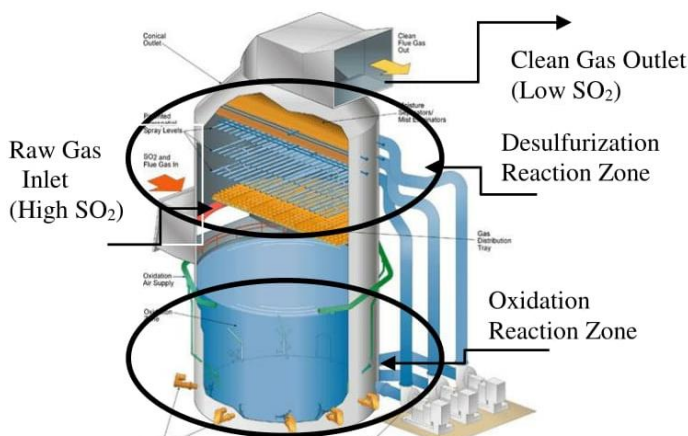
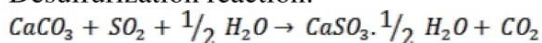


Figure 3. Wet Limestone FGD Absorber Cutaway [3]

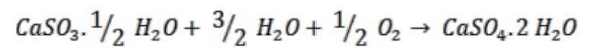
In wet LSFO FGD, 2 types of chemical reaction occurs. The first chemical reaction is desulfurization reaction occurring in the reaction tray, which is between SO_2 and CaCO_3 that creates $\text{CaSO}_3 \cdot 1/2\text{H}_2\text{O}$ (Calcium Sulfite). In this reaction, 95% of SO_2 in the flue gas is captured.

Desulfurization reaction:



1 gram of CaCO_3 can absorb 582 mg of SO_2 . The result of Desulfurization is a physically undesirable Calcium Sulfite compound. However, Calcium Sulfite can be oxidized into Calcium Sulfate (gypsum) that have some commercial value and is physically easier to handle.

Oxidation reaction is as follow:



Based on calculation described in table 2, the combustion of 100 kg coal with sulfur of 0.57% weight will create 1.14kg of SO_2 that is diluted within 968 kg of flue gas with 1487 mg/Nm^3 SO_2 concentration. The mass balance for desulfurization with wet LSFO FGD is described in Table IV:

Table 3. Wet Limestone Desulfurization calculation

Desulfurization Reaction (100 kg Fuel Basis)				
Exhaust Gas		Weight	molar Mass	mol
Desulfurization reactant		kg	kg/kmol	kmol
SO_2	Sulfur Dioxide	1,14	64	0.018
CaCO_3	Calcium Carbonate	1,78	100	0.018
H_2O	Water	0,2	18	0.009
Desulfurization Reactant		3,1		0.045
Desulfurization Product				
Clean Flue Gas		Weight	molar Mass	mol
Desulfurization Result		kg	kg/kmol	kmol
$\text{CaSO}_3 \cdot 1/2 \text{H}_2\text{O}$	Calcium Sulfite	2,8	156	0.018
CO_2	Carbon Dioxide	0,8	44	0.018
Total Desulfurization product		3,6		0.036
Oxidation Reaction				
O_2	Oxygen	0,3	32	0.0089
H_2O	Water	0,5	18	0.0267
Hasil akhir desulfurisasi				
$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$	Gypsum	3,56	199	0.018

Thus, for each kWh of electricity produced ;

- 0.00454 kg SO_2 s formed with a concentration of 752 mg/Nm^3 at 7% O_2 content in flue gas.
- 0.00743 kg CaCO_3 is needed for desulfurization reaction
- 0.00289 kg of water is needed for slurry dilution and oxidation reaction.
- 0.00512 kg of air is needed for oxidation reaction
- 0.0144 kg Gypsum is formed
- With an limestone cost of Rp 500/kg, thus desulfurization cost is Rp3,71/kWh

IV.Duct Sorbent Injection FGD

Due to the complexity of LSFO FGD System, and the near impossibility for retrofit to

existing CFPP, other types of flue gas desulfurization system for retrofit purposes is explored with the following requirements:

- Not needing to build absorber (minimum footprint)
 - Not needing complicated reagent preparation system
 - Low Investment Cost
 - Has sufficient desulfurization efficiency, enough to lower SO₂ level to below 750 mg/Nm³ as per Indonesian Regulation.
- Thus, the following system is proposed:

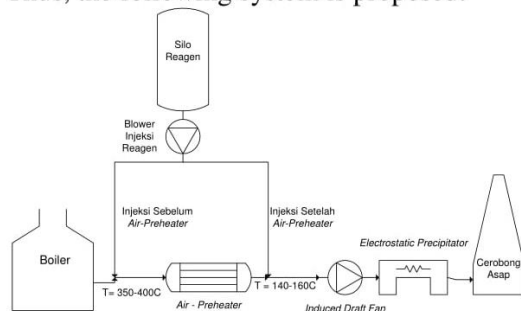


Figure 4. Duct Sorbent Injection FGD

The system that fulfill the above mentioned criteria is Duct Sorbent Injection (DSI) system, which is a desulfurization process involving Injecting Sorbent in the flue gas duct. In Coal Fired Power Plant, the injection location is described in Figure 3. The injected adsorbent reacts with SO₂ in the flue gas and capture it, thus reducing the SO₂ concentration in flue gas. Calculation shows that for a typical 660 MW powerplant, the time needed from injection point before air preheater to after air preheater is approximately 3-4 seconds, and from after air preheater to electrostatic precipitator is another 3-4 seconds. The reacted adsorbent is then captured by the Electrostatic Precipitator, and is mixed with fly ash for disposal. The advantage of using DSI as method of Flue Gas Desulfurization are :

- Minimum footprint
- Minimum Investment

The Disadvantage of DSI are :

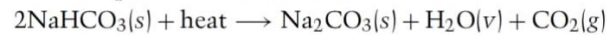
- Less Effective than LSFO
- Higher Absorbent Cost
- Increased Fly Ash Production

The desulfurization absorbent deemed suitable for this type of system is Sodium Bicarbonate (NaHCO₃). One of the reasoning behind the absorbent selection are the stability of

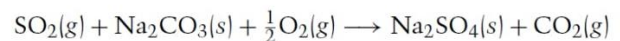
compound, for long period of time in room temperature.

The Desulfurization reaction of the adsorbent are as Follow:

In the Injection point, flue gas temperature must be above 140 Celcius to enable NaHCO₃ to decompose into Na₂CO₃ as follow:



The thermal decomposition process release H₂O vapor and CO₂ gas, leaving Na₂CO₃ in solid form, thus creating porosity, and increasing surface area , promoting SO₂ to react with Na₂CO₃ as described in Figure 5. However, Thermal decomposition is limited to temperature of 400 Celcius, because sintering of sodium carbonate occurs, and the benefit of high surface area is reduced, decreasing effectiveness of the reaction[8]. The Na₂CO₃ then reacts with SO₂ as follow:



The final solid product is Sodium Sulfite (Na₂SO₄) in solid powder form. In stoichiometric condition, 1 Gram of Na₂CO₃ can absorb 380 mg SO₂, that will be referred to as maximum adsorbent capacity of Na₂CO₃.

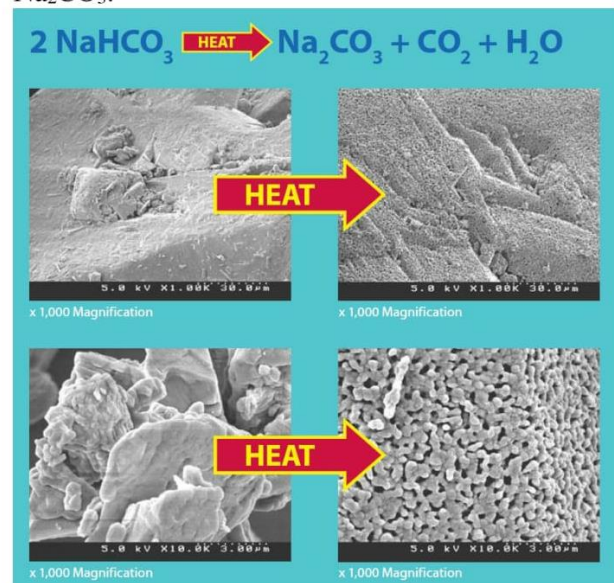


Figure 5. Thermal Decomposition of NaHCO₃ [8]

V. Laboratory Scale Dry FGD

Experiment

To understand of the characteristic of Adsorbent Chemical used for Dry Desulfurization, a laboratory scale experiment is conducted. The idea is to simulate the limited reaction time of SO₂ and adsorbent in elevated temperature. The installation of experiment apparatus is described in Figure 6:

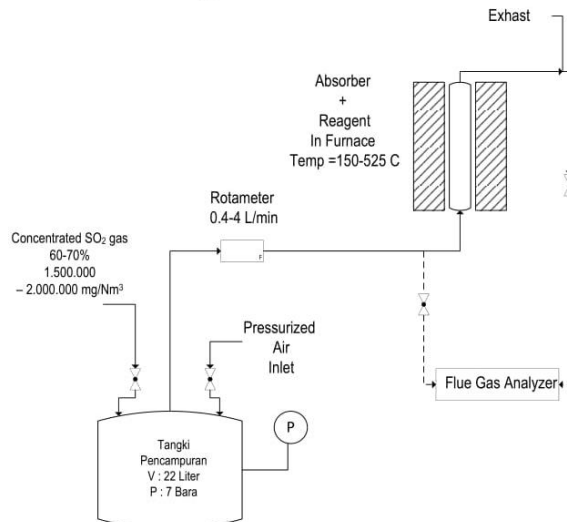
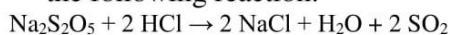


Figure 6. Laboratory Scale FGD Installation

The operating principle of the apparatus is as follow:

SO₂ gas is produced by reacting Sodium Metabisulfite (Na₂S₂O₅) with Hydrochloric Acid (HCL) in a closed 100 mL container with the following reaction:



Every 100 mL injection will contain 150-200 mg of SO₂ Gas with Very high Concentration of 1.5-2 Million mg/Nm³. The SO₂ is the injected into an unpressurized 22 Liter tank. The tank is then pressurized to 6 bar gauge to dilute the SO₂ concentration and enable flow to the reactor. Each 100 ml injection will yield 1200-1400 mg/Nm³ in the gas exiting the tank, and SO₂ concentration can be controlled by varying the amount of SO₂ injection in tank.

The diluted SO₂ gas is then throttled and flowed into a ¼ inch diameter stainless steel adsorber by feedback of rotameter with range of 0.4-4 L/min. The absorber is filled with 0.25 to 1 gram of reagent of 28- to 400- mesh size, and is located inside a furnace with temperature up to 525 °C. The reagent is not distributed, and only occupy less than 1

centimeter of the absorber height. Thus, reaction time between flue gas and reagent is very brief. For a gas flow of 1,209 L/min, it will yield contact time of 0,062 second /cm adsorber height. Which is much less than what would be expected when the system is applied in the actual power Plant. Typical application in 660 MW power plant will yield about 8 seconds of reaction time, if injected in the area between Economizer and Air Preheater, until the flow reach Electrostatic Precipitator.

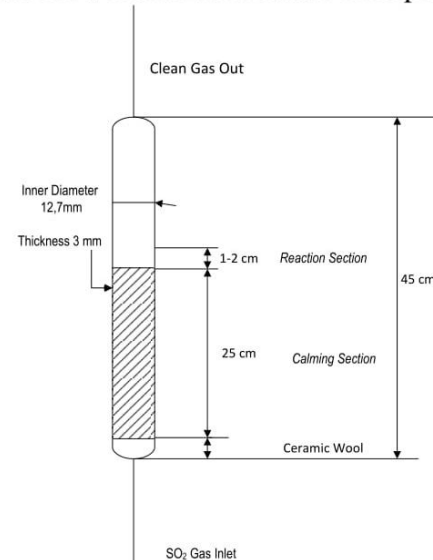


Figure 7. Adsorber Configuration

The Raw and clean gas SO₂ concentration is measured using BACHARACH Gas Analyzer model "Portable Combustion Analyzer 3" (PCA 3) capable of measuring SO₂ up to 15.000 mg/Nm³ within 5% accuracy. It is known that there is a 50 - 100 mg/Nm³ SO₂ concentration reduction for gas flowing through an empty reactor in elevated temperature. Furthermore, the SO₂ concentration in the gas flowing from the tank gradually decreases over time, typically 100-200mg/Nm³ per hour as described in Figure 8. Such difference is accounted for in the Adsorbent effectiveness calculation.

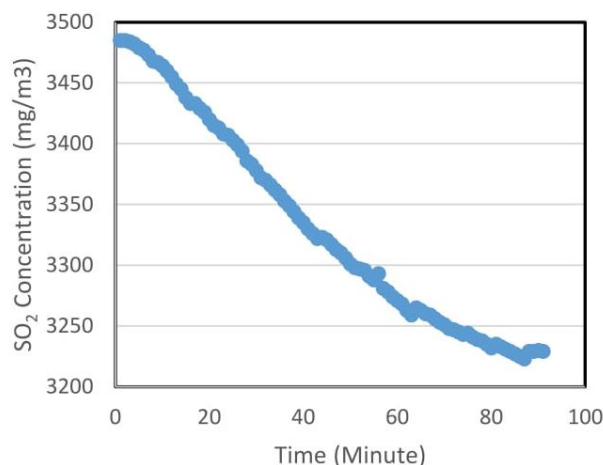


Figure 8. Example of reduction of raw gas SO_2 concentration ejected from tank over time

VI. Method of experiment calculation

The NaHCO_3 adsorbent is located inside the reactor, and concentration of SO_2 in the raw gas is measured once, using the Flue Gas Analyzer. The gas is then continuously flowed into the reactor within the furnace, in which the adsorbent has been heated for at least 15 minutes. The clean gas exiting the reactor is then measured and logged continuously with a resolution of 1 minute. With most of the gas flowrate being set of 1,209 L/min, the total data acquisition period is 90-110 minutes. After the mixing tank is nearly depleted, the raw gas is measured once again to find out the reduction of SO_2 concentration. Figure 9 shows detail of the raw Data acquired from the flue gas analyzer. It shows data of the following parameter:

Adsorbent mass: 0,5 g
Particle size : Below 149 micron
Initial raw gas concentration : 2482 mg/Nm³
Reactor temperature : 170 C

VII. Effectiveness of desulfurization Experiment

Effect of mass of reagent: In this set of 3 experiment an SO_2 concentration of 2500 mg and 4000 mg/Nm³ is flowed into a reactor of temperature 170 °C, with reagent mass of 0,25 g (#7), 0,5g (#6), and 1g(#5).

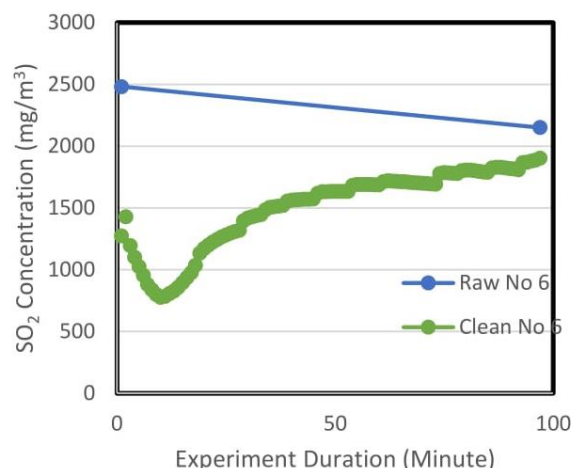


Figure 9. Example of NaHCO_3 desulfurization raw data Based on the SO_2 Concentration and flowrate, the Y Axis data is converted into mg SO_2 /min, and total SO_2 Mass flow is known. The rate of adsorption is calculated from the total SO_2 in the raw flue gas, deducted by the total SO_2 in the Clean Flue gas.

For the Graph Above, the calculated data is shown in Table 4.

Table 4. Calculation of NaHCO_3 Desulfurization

Parameter	Figure
Adsorbent Mass	0.5 g
Particle Mesh Size	100
Total SO_2 Raw Gas	268.86 mg
Total SO_2 Clean Gas	175.82 mg
Total SO_2 Absorbed	93.02 mg
Fraction SO_2 Adsorbed	34.61%
Adsorbent Capacity (mg SO_2 /g adsorbent)	380
Actual Adsorbtion capacity (mg SO_2 /g adsorbent)	186.08
Fraction Adsorbent Capacity	48.85%

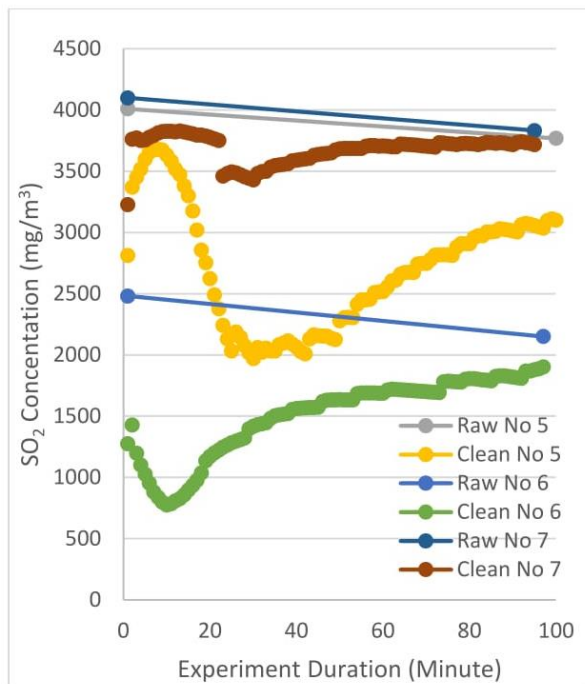


Figure 10. Comparison of raw SO₂ data

The experiment shows different SO₂ adsorption figures, but yield relatively similar NaHCO₃ conversion, as explained in table 5.

Table 5. Desulfurization effectiveness as function of mass

Exp no	Raw SO ₂ (mg/Nm ³)	Experiment Duration (minute)	NaHCO ₃ Conversion	SO ₂ Conversion	Reagent Mass (gram)
5	4010	99	24.67%	30.28%	1
6	2482	96	48.85%	34.61%	0.5
7	4100	94.5	31.67%	6.66 %	0.25

NaHCO₃ Characteristic curve: Figure 10 shows the typical “dip” in the clean gas curve, also apparent in figure 11, with 170°C, and 0,5g of NaHCO₃. This phenomenon is also apparent in figure 11, and is most likely due to the completion of decomposition process from NaHCO₃ to Na₂CO₃. The NaHCO₃ is heated with the set temperature, approximately 15-20 minutes before SO₂ gas is flowed. The experiment result is explained in table 6.

Table 6. Desulfurization experiment result of similar parameter.

Exp no	Raw SO ₂ (mg/Nm ³)	Experiment Duration (minute)	NaHCO ₃ Conversion	SO ₂ Conversion
4	3663	59	30.47%	68.07%
5	4010	99	24.67%	30.28%

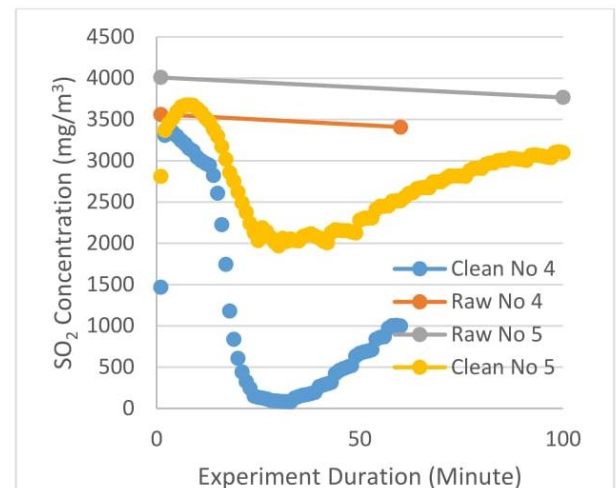


Figure 11. Comparison of SO₂ raw data.

Effect of temperature : Literature states that above 400°C, the NaHCO₃ coagulates, thus reducing effectiveness. The data acquired confirms such claim, but not in a very drastic measure. Furthermore, maximum desulfurization effectiveness is achieved in temperature of 425 °C.

Table 7. Effect of temperature on desulfurization effectiveness

Exp no	Raw SO ₂ (mg/Nm ³)	Experiment Duration (minute)	NaHCO ₃ Conversion	SO ₂ Conversion	Temperature
19	1750	92	37.53%	38.69%	325
20	1780	94	43.45%	42.35%	375
21	1877	86	52.18%	52.43%	425
22	1780	97	35.04%	33.30%	475
23	1800	83	23.47%	25.40%	525

VIII. Conclusion of Experiment Result

1. Data shows a suitability for NaHCO₃ for dry desulfurization system in CFPP due to its temperature compatibility with flue gas temperature.
2. Data Shows an increase in desulfurization effectiveness of NaHCO₃ SO₂ loading in higher reactor temperature, with a limiting temperature of about 425 °C.
3. Experiment shows quite good desulfurization result in very short reaction time of less than 0,6 second. A higher reaction time in actual application should yield better desulfurization result.

IX. Economic Calculation of DSI FGD

Based on the laboratory desulfurization result, it can be assumed that the NaHCO₃ Conversion is 80%, meaning 1 gram of NaHCO₃ can capture 304 mg of SO₂. With

the Target clean gas of 376 mg/Nm³, which is approximately half of what is regulated. The cost prediction for desulfurization based on the above assumption is explained in table. Also to be considered is the additional desulfurization byproduct that will be added to the fly ash.

Sulfur Content (% weight)	SO ₂ Production (kg/660 MWh)	Raw SO ₂ Concentration (mg/Nm ³)	Clean SO ₂ Concentration (mg/Nm ³)	Reagent Consumption (kg NaHCO ₃ /660 MWh)	Desulfurization cost (Rp/kWh)	Fly Ash Byproduct (kg/660 MWh)
0,57	2995	752	376	4926,9	26,87	4315
0,85	4493	1128	376	9389,4	53,67	8616
1,14	5991	1504	376	14.786	80,54	12.719

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