

Verification of Microfine Lime[®] Theoretical Model for SO₂ Removal in Precalciner Cement Plants

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ABSTRACT

Envirocare International has developed a performance model for use in predicting SO₂ removal from cement kiln exhaust gases using Microfine[®] lime slurry injection. This model was verified through field testing in a full-scale Portland cement plant. The physical system was determined to be robust and commercially viable for use. SO₂ removal was within expected requirements and the Envirocare theoretical model was verified by field results. This model allows end users to submit permit applications with assurance that they will meet required SO₂ emission limits. This paper describes the variables and physical/chemical reactions used in predicting expected performance and the accuracy of the model.

INTRODUCTION

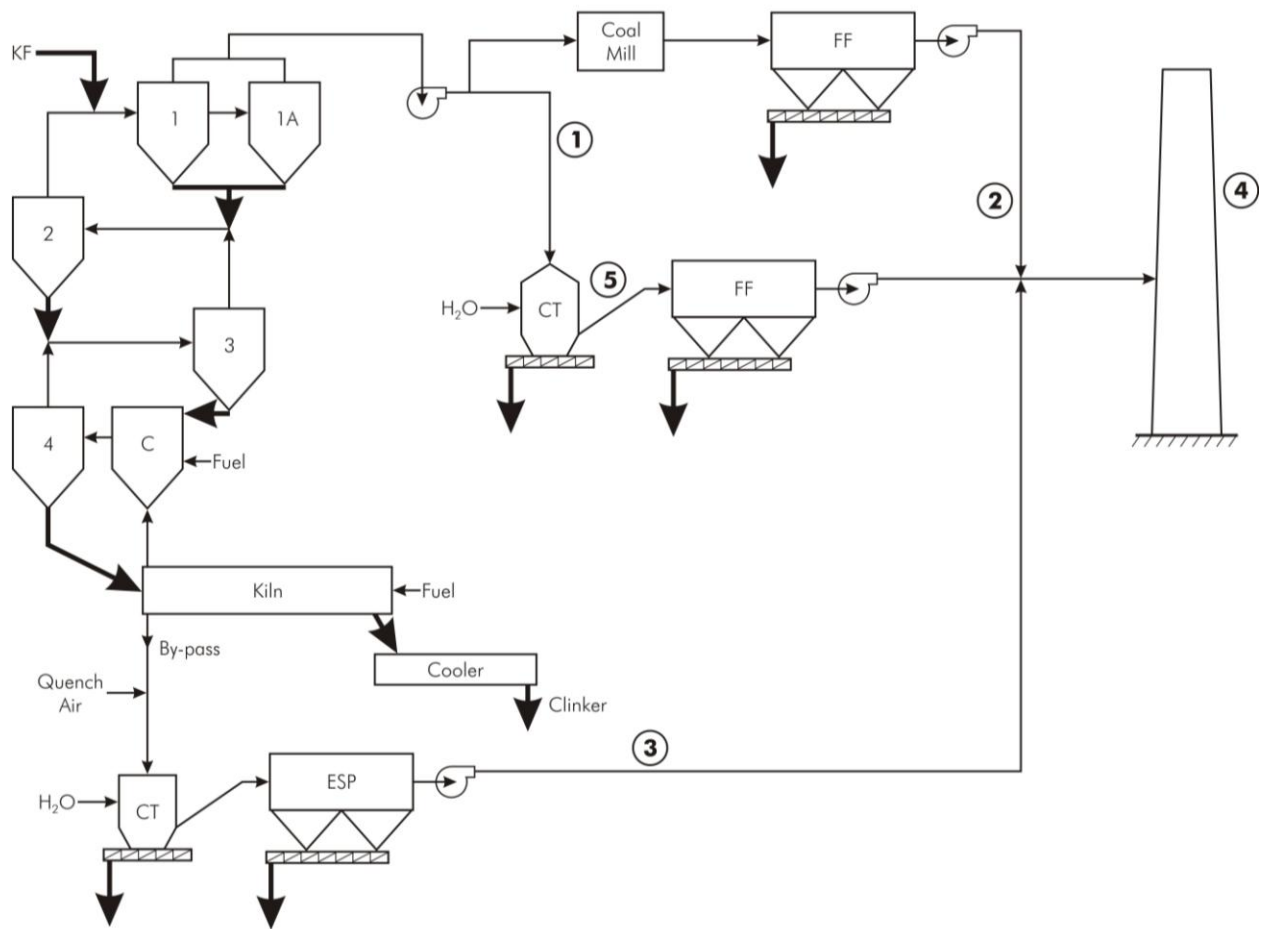
Envirocare International has developed a patented method for removal of SO₂ gas from flue gases employing a high surface area hydrated lime (i.e. Microfine Lime[®]).¹ This material used in combination with dual fluid spray nozzles manufactured by Envirocare has been demonstrated to exhibit high SO₂ removal when used in conditioning towers or down-comer ducts in preheater/precalciner cement kilns. As a demonstration project, a Portland cement plant completed a field evaluation of the process in its facilities. The evaluation was used to demonstrate the reliability of the physical system (pumps, meters, lances, and nozzles) and the effectiveness of the chemistry (i.e., percent removal, hydrate usage, and impact on plant operation). As part of the demonstration, the theoretical predictive model used by Envirocare was compared to the field results.

At the completion of the demonstration, it was concluded that the physical system was robust and commercially viable for use. SO₂ removal was within expected requirements and the Envirocare theoretical model was verified by field results.

PROCESS OVERVIEW

The cement process involves the heating, calcining, and burning of cement rock to produce clinker. In this process sulfur in fuels and kiln feed (pyrites) is oxidized to SO_2 and is removed with combustion gases. These gases are cooled in a conditioning tower (CT) using water sprays and then passed through a fabric filter. In the process test, addition gases by-pass the CT and are used in coal grinding and are then combined at the kiln stack (Figure 1). Gases are also removed at the kiln exit (i.e., bypass system), cleaned in an ESP, and combined with the main CT gases at the stack. This project involved removal of SO_2 in the main CT using injection of Microfine Lime[®].

Figure 1. K5 Process Flow Diagram.



Test Procedure

Environmental Quality Management, Inc. (EQ) used measurements of flue gas sulfur dioxide at the main CT inlet, coal mill exit, by-pass exit, and main stack to determine the removal of SO₂ in the main CT.

In order to complete an SO₂ mass balance, SO₂ concentration, percent O₂, temperature, and gas flow volume were measured at Points 2, 3, and 4. In addition, percent oxygen and SO₂ concentration were measured at Point 1. Due to high dust loading, gas flow could not be accurately measured at Point 1. The flow at Point 1 was calculated by subtraction of mass flows at Points 2 and 3 from that at Point 4 with correction for false air (leakage) and the water injected in each CT. As a result, the SO₂ mass rate at the main CT inlet could be calculated.

In addition, a full mass balance was completed using kiln fuel rates, kiln feed rates, added moisture, and combustion air (forced and false) to verify mass flow at each point. The full mass balance (Table 1) agreed reasonably with the short method. Table 1 presents the SO₂ balance completed for the kiln baseline period prior to the injection of lime. This verified that no removal of SO₂ was occurring across the main fabric filter at the measured gas stream temperature and moistures.

Table 1. Example of Kiln Mass Balance for Baseline Condition Prior to Lime Injection.

Preheater Exit		
Gas Volume	Calculated	150,470 Nm ³ /hr
Temperature	Calculated	360°C
O ₂	Measured	2.92%
H ₂ O	Calculated	5.58%
SO ₂	Calculated	211 ppm (wet)
SO ₂	Calculated	90.85 kg/hr
Coal Mill System		
Preheater Gases	Calculated	25,271/Nm ³ /hr
Temperature	Measured	360°C
O ₂	Measured	2.92%
H ₂ O	Calculated	5.58%
Quench Air	Calculated	37,090 Nm ³ /hr
Temperature	Measured	21.1°C
Water Evaporated From Coal	Calculated	1,289 kg/hr
False Air	Calculated	4,166 Nm ³ /hr
Mill Outlet Volume	Measured	68,131 Nm ³ /hr
Temperature	Measured	81.3°C
O ₂	Measured	6.66%
H ₂ O	Calculated	13.2%
SO ₂ (Point 2)	Measured	13.2 ppm (wet)
SO ₂ (Point 2)	Calculated	15.26 kg/hr

Table 1 (continued).

Main Stack		
Gas Volume	Measured	357,220 Nm ³ /hr
Temperature	Measured	1.79°C
O ₂	Measured	11.61 %
H ₂ O	Calculated	11.6%
SO ₂ (Point 4)	Measured	90.91 ppm (wet)
SO ₂ (Point 4)	Calculated	92.8 kg/hr
CT Inlet System		
Flue Gas Volume (CT inlet)	Calculated	125,198 Nm ³ /hr
Temperature	Measured	360°C
H ₂ O	Calculated	5.58%
O ₂	Measured	2.92%
SO ₂ (Point 1)	Measured	215 ppm (wet)
SO ₂ (Point 1)	Calculated	83.8 kg/hr
Water Rate	Measured	318 l/min.
Flue Gas Volume (CT Outlet)	Calculated	161,776 Nm ³ /hr
Temperature	Measured	160°C
H ₂ O	Calculated	17.7 %
O ₂ (wet)	Measured	5.05%
SO ₂ (Point 5)	Calculated	151.2 ppm (wet)
SO ₂ (Point 5)	Calculated	75.59 kg/hr
By-Pass System		
Flue Gases (Kiln)	Calculated	3,557 Nm ³ /hr
By-Pass	Calculated	5.2% Volume
Temperature	Measured	1,110°C
Quench Air	Calculated	11,302 Nm ³ /hr
Temperature	Measured	21.1°C
Flue Gases Quenched	Calculated	14,860 NM ³ /hr
Temperature	Measured	439.8°C
Water Rate	Measured	53.99 l/min.
Flue Gas (ESP Exit)	Measured	18,887 Nm ³ /hr
Temperature	Measured	164.5°C
H ₂ O	Calculated	23.2%
O ₂ (wet)	Measured	14.05%
SO ₂ (Point 3)	Measured	36.66 ppm (wet)
SO ₂ (Point 3)	Calculated	1.98 kg/hr

Microfine Lime[®] was introduced through dual flood nozzles under three conditions:

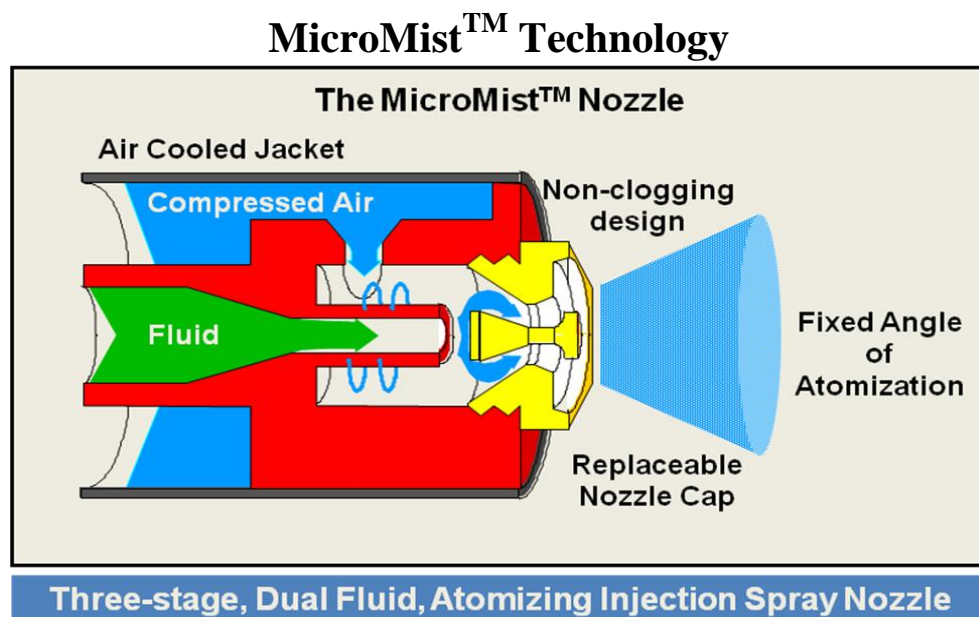
- 1 Three nozzles at 1.53 Ca/S molar ratio.
- 2 Six nozzles at 1.58 Ca/S molar ratio.
- 3 Nine nozzles at 1.57 Ca/S molar ratio.

The injection of hydrate was completed in three steps for a period of 90 minutes at each condition, after which the injection of hydrate was stopped allowing the post-test confirmation of the pre-test baseline SO₂ emission rate.

ECI Predictive Model

The Envirocare SO₂ removal model is based on the physical/chemical reaction occurring as a water droplet, containing a hydrated lime solids suspension, evaporates in a flue gas stream which contains sulfur dioxide gas. As applied in the main CT, the solution is introduced through dual fluid nozzles (i.e., air/water)² to produce a mono-dispersed water droplet between 120 and 150 μm in diameter (Figure 2). The droplet size is specifically defined by the nozzle design and air/water pressures. A mono-dispersed droplet is necessary to accurately control evaporation time. A skewed droplet distribution produces larger diameter droplets which can cause a wet bottom in the evaporation tower.

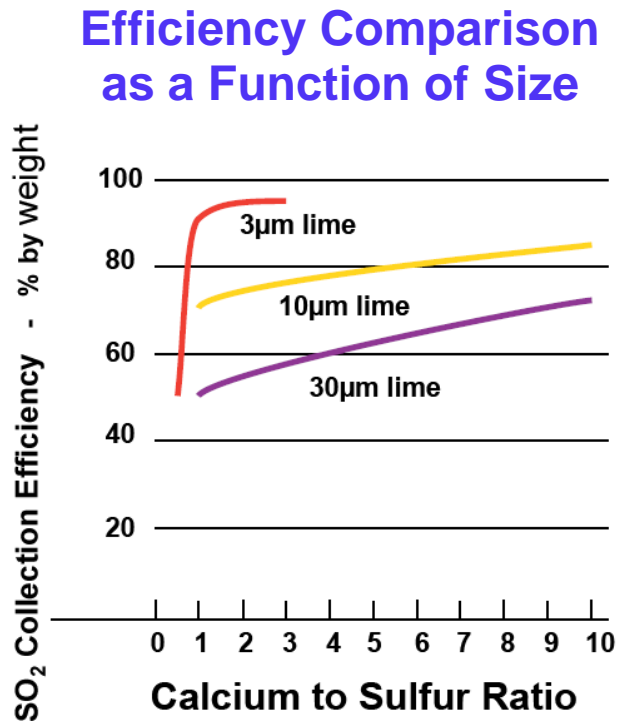
Figure 2. ECI Nozzle Design.



Hydrated lime is suspended in the water droplet, producing a dissolved solution which reacts with SO₂ adsorbed at the droplet surface. As the soluble hydrate is consumed, the solution is refreshed by the hydrate suspension. The surface area of the hydrate solids determines the refresh rate and controls the SO₂ removal rate in the CT. Microfine Lime[®] is a lime product specifically manufactured to achieve maximum SO₂ adsorption (Figure 3).

The difference between normal spray drier systems during SO₂ removal processes and that used by Envirocare is that this process is designed to remove SO₂ in the droplet evaporation phase and does not depend on SO₂ removal in the particulate dust layer in downstream fabric filters.³ For this reason, the final gas temperature does not require a close approach to the moisture dewpoint for high SO₂ removal. It therefore can be applied in CT and/or duct locations without fouling or leaving a build-up on walls.

Figure 3. Expected SO₂ Removal as a Function of Lime Particle Size.



System Design

The main CT was retrofitted with nine spray lances, a valve rack, and a lime slurry pump to allow injection of lime slurry at predetermined injection rates with the ability to control the air/water rate and pressure at the lance (Figure 4). This allowed the water rate (l/m), lime rate (l/min), and air pressure to be adjusted. As a result, the droplet size, CT bottom temperature, and SO₂ removal could be controlled during the test. Lime from the slurry pump is combined at the spray lance nozzle at the point of injection (Figure 5).

The nozzle pressure and sizing was specified to produce a non-overlapping spray pattern with all nine sprays in use and complete evaporation in 2 seconds residence time (Figures 6 and 7).

Figure 4. General Arrangement of Air/Water Control System for the CT Lances.

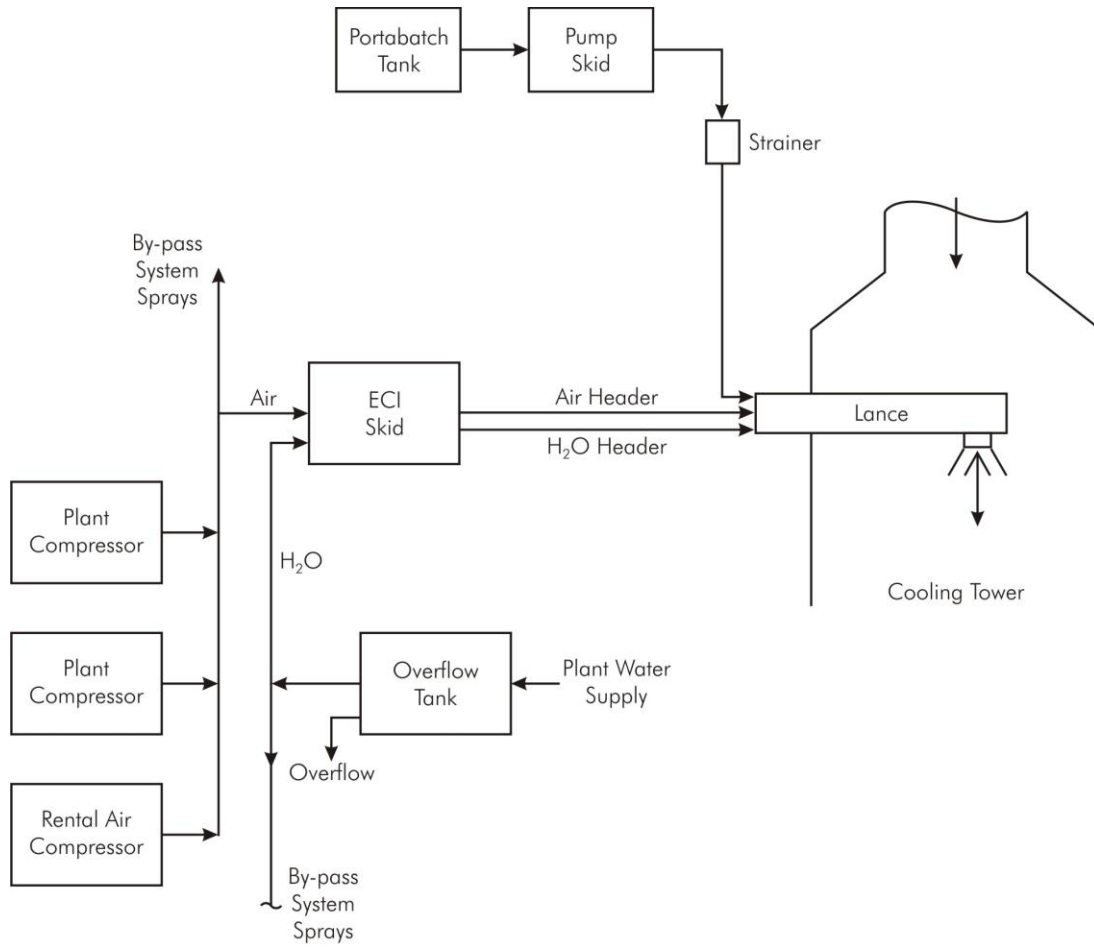


Figure 5. Lance Assembly With Lime Slurry Injection Port.



Figure 6. Elevation View of CT.

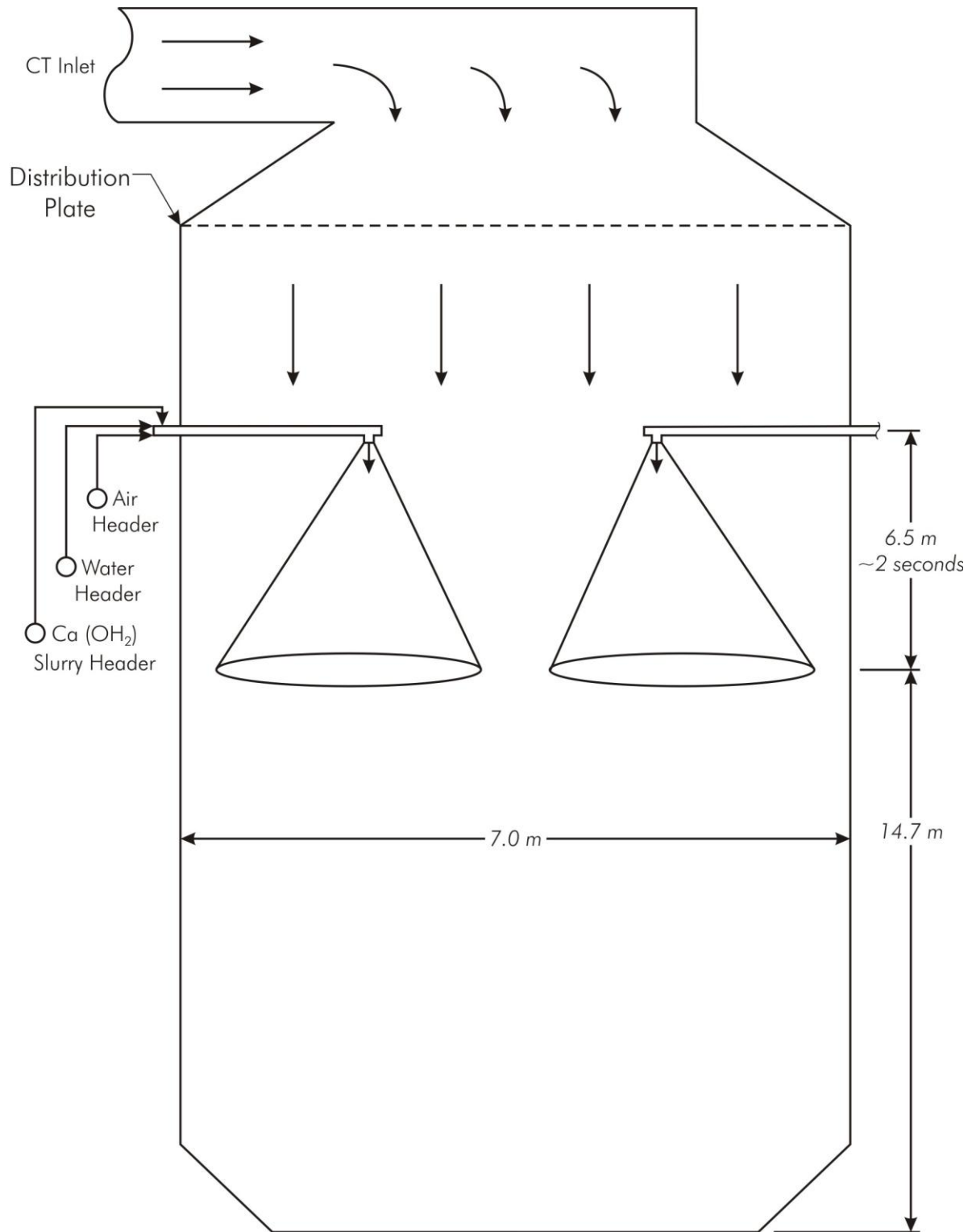
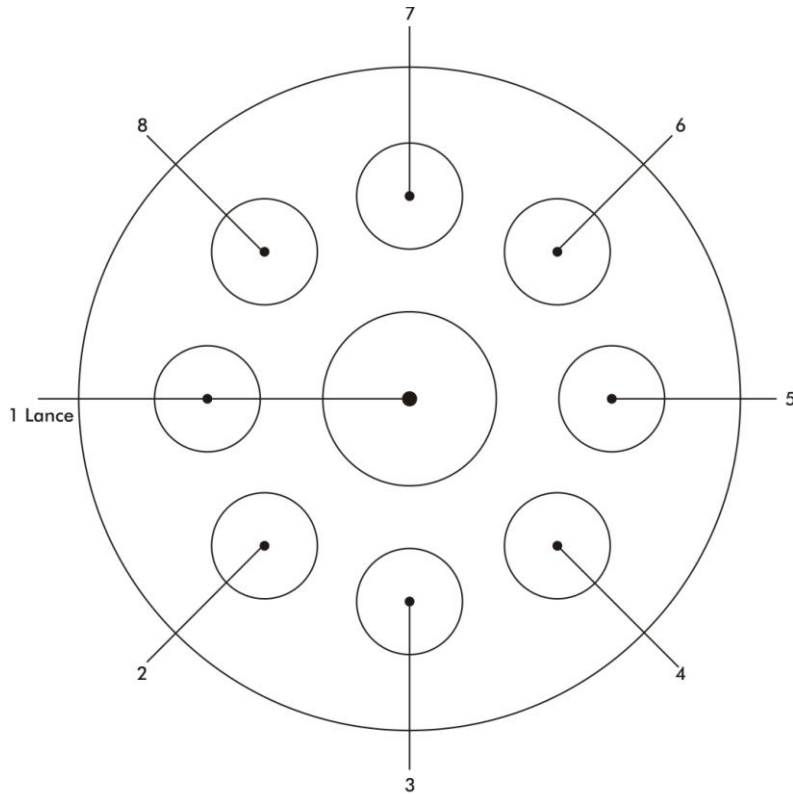


Figure 7. Plan View of CT Spray Pattern.



Model Input Parameters

The following variables are used to estimate SO₂ removal in the CT and ducts employing Microfine Lime[®] lime as a reagent:

Item	Unit
hydrate diameter	μm
hydrate surface area	m ² /g
Ca/S ratio (molar)	
slurry concentration (droplet)	% wt
SO ₂ inlet concentration	ppm (wet)
dust diameter	μm
dust mass rate	g/sec
gas mass rate	kg/sec
gas temperature	°K
gas humidity	% (vol)
gas CO ₂	% (vol)
CT area	m ²
CT length	m
water mass rate	g/sec
water velocity	m/sec
droplet diameter	μm

Model Output Parameters

Item	Unit
distance for evaporation	m
time for evaporation	sec
calcium utilization	%
SO ₂ capture	%
gas temperature	°K
gas humidity	% (vol)
solid diameter	μm
gas velocity	m/sec

Model Results

The removal of SO₂ in the CT is a function of the coverage of the tower cross-section and contact time with the gases before the droplet fully evaporates. In the experimental data, the three conditions represent increasing coverage areas in the tower. The gases were cooled to the same temperature by controlling the total water injected. A total of nine nozzles were in operation at all times (Table 2).

Due to the CT design, complete cross-section coverage with nine nozzles was not optimum and a total of 18 would be needed to give 75% coverage. Without tower re-design, a coverage of >80% would result in water/slurry impingement on walls and is not recommended. This limits the removal efficiency to 60% (Table 3).

Table 2. CT Operating Conditions.

Flue gas inlet condition	125,200 Nm ³ /hr 360 °C 5.5 % H ₂ O 83.8 kg/hr
Flue gas outlet conditions	161,776 Nm ³ /hr 160 °C 22.0 % H ₂ O
Diameter	7.0 meters
Length	21 meters
Water spray rate	270 l/min
Lime injection	248.8 kg/hr (3) 255.6 kg/hr (6) 254.6 kg/hr (9)

Table 3. Summary of Results of Predictive Model vs. Field Data.

Total Nozzles in Use	Nozzles Supplied With Ca(OH) ₂	Percentage Ct Area Covered	Measured SO ₂ Removal, %		Predicted Removal, %
			Stack	CT Outlet	
9	3	19	11.76	14.17	15
9	6	38	29.1	31.7	31
9	9	57	40.75	43.8	45
18 ^a	18	75			60
18 ^b	18	80			70

^aRedesign of lance system for maximum lance penetrations.

^bRedesign of CT to improve flow pattern for maximum area coverage.

CONCLUSIONS

The data obtained through this study verified the accuracy of the predictive model in the design of Microfine Lime[®] SO₂ abatement systems. This model allows end users to submit permit applications with assurance in meeting required SO₂ emission limits.

REFERENCES

1. Schwab, J., U.S. Patent No. 6,464,952, October 15, 2002, Sulfur Dioxide Abatement Method.
2. MicroMist Cooperative Gas Cooling and Conditioning Systems, Envirocare Systems International, Navato, California.
3. Sheth, S.H., SO₂ Emission and Scrubbing Systems, 33rd IEEE Cement Industry Conference, Mexico City, May 1991.