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EKONO, Inc, Bellevue, Wash

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This publication, directed towards the process and design engineer, describes types, quantities, and sources of emissions, presents the latest control device alternatives, and estimates costs for implementing the air pollution control systems. Emphasis is placed on explanation of chemical and physical processes which generate emission in specific unit operations so that the advantages and disadvantages of both internal and external process control methods can be understood. Actual field installations have provided the basis for the majority of design data.

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CHAPTER 11

LIME BURNING AND LIME DUST HANDLING

The causticizing of liquor, commonly called green liquor, from the smelt dissolving tank by addition of lime or calcium oxide (CaO) results in the generation of a lime mud or calcium carbonate ($CaCO_3$) sludge. The lime and sludge are then washed and calcined at elevated temperatures in either a rotary kiln or a fluidized bed calciner to recover calcium oxide. This oxide can then be reused for reclaiming additional white liquor, the chemical solution for digesting pulp. The normal auxiliary fuels used as heat sources for lime mud burning are natural gas and residual fuel oil. The two major potential air pollutants from lime mud burning are the gaseous emissions and the particulate emissions of entrained lime dust from the burning zone. The gaseous emissions are H_2S from the lime mud and, possibly, organic sulfur compounds from the scrubbing water.

11.1 Rotary Lime Kilns

11.1.1 Design Features

The rotary kiln is the most commonly employed device for lime mud reburning in kraft pulp mills. The device is an open-ended inclined cylinder that is rotated so that lime mud added at the upper end gradually passes to the lower end and drops out into a bin as dry lime. Fuel and air flow countercurrently to the lime from the lower end of the kiln. The kiln exhaust gases normally pass through a mechanical cyclone collector for lime dust recovery and finally through a liquid scrubber for particulate control (1).

Rotary lime kilns employed in kraft pulp mills can range from about 2.4 to 4.0 m (8 to 13 ft) in diameter and from about 30 to 120 m (100 to 400 ft) in length. They are designed to burn 36 to 360 t (40 to 400 tons) of lime (as dry CaO) per day (2). The lime kilns are normally inclined at a slope of about ten degrees from the horizontal plane and can be supported by two- to four-supports, depending on their length. The lime kilns must be designed with a number of auxiliary components, including a lime mud feed system, hot lime conveying system, air inlet and preheating system, gas exhaust system, kiln rotation system, and instrumentation systems (3). Major kiln design variables include kiln length, kiln diameter, rotation speed and angle of incline, which influence solids retention time, gas-solids contact area, and temperature.

11.1.2 Operating Parameters

Lime mud at 55 to 65 percent solids and with sodium content of 0.1 to 2.5 percent by weight (as Na_2O) enters at the upper end of the lime kiln and passes through successive stages of water evaporation, mud preheating, and lime calcination. Temperatures in the kiln vary from 150 to 260° C (300 to 500° F), at the upper or wet end, to 1200 to 1300° C (2200 to 2400° F) at the hottest part of the calcination zone near the lower or dry end. Energy requirements for the lime kiln operation are for water evaporation, preheating and calcining the lime mud, and power to rotate the lime kiln, drive the air fans and flue gas fans, pump the scrubber liquid, and convey the lime mud and reburned lime. The major types of fuels burned in lime kilns are natural gas and residual fuel oil; turpentine and coal may also be used. Fuel requirements for lime kilns and fluidized bed calciners are listed in Table 11-1.

Two of the major design variables affecting particulate emissions from lime kilns are kiln length and diameter. These variables can affect the amount of particles swept from the kiln exhaust gases by governing the gas velocity and the gas-solids contact area.

TABLE 11-1
ENERGY REQUIREMENTS FOR LIME MUD CALCINING SYSTEMS (3) (4)

Rotary Kiln		Fluid Bed Calciner	
kJ/t	(BTU/ton)	kJ/t	(BTU/ton)
$2.3\text{-}4.7 \times 10^6$ *	$(2\text{-}4 \times 10^6)$*	$2.1\text{-}2.5 \times 10^6$	$(1.8\text{-}2.0 \times 10^6)$*
$9.3\text{-}17.4 \times 10^6$ **	$(8\text{-}15 \times 10^6)$**	$8.0\text{-}9.2 \times 10^6$	$(7\text{-}8 \times 10^6)$**

*per metric (t) or short ton (ton) of pulp.
**per metric (t) or short ton (ton) of lime, as CaO.

H_2S emissions from the lime kiln are affected by the Na_2S content of the lime mud (particularly the aqueous phase) and by the presence of Na_2S in the scrubber wash water. The use of digester and evaporator condensate as lime kiln scrubber water can result in the stripping of organic sulfur compounds into the exit flue gas. The presence of sufficient excess air in the kiln can reduce the concentration of H_2S in the exhaust by providing an oxidizing atmosphere sufficient for H_2S conversion to SO_2 (5).

11.2 Fluidized Bed Calciners

Fluidized bed calciners are alternatives to rotary lime kilns for the calcination of lime mud to lime. The lime mud is first washed to reduce soluble sodium compounds to a sodium content of 0.1 to 0.5 percent by weight (as Na_2O) and then dried on a vacuum filter. The dried lime mud at 55 to 65 percent solids is then suspended in the flue gas from the fluidized beds at a temperature of about 150° C (300° F) to evaporate the water. The solids are then passed through a two-stage cyclone system to recover the dried lime mud solids and then fed into a bed of fluidized lime pellets formed by calcination. The bed is kept in suspension by the action of an air fan located below the cooling chamber from which reburned lime is removed. Natural gas or fuel oil, is injected into the suspended bed and burned to provide the heat necessary for the calcination reactions to take place at a temperature of about 825 to 875° C (1500 to 1600° F). The entrained particles and the combustion gas products then pass out from the calciner to entrain the wet mud and pass through the two-stage cyclone system and a venturi scrubber for particulate removal (5).

Fluidized bed calciners are employed at several kraft pulp mills and have lime burning rates ranging from 23 to 136 t per day as CaO (25-150 ton/day). Fuel requirements for fluidized bed calciners are generally lower than for lime kilns because of the small combustion chambers used which have smaller radiation heat losses. Electricity requirements for fluidized bed calciners, however, are generally greater than for rotary kilns because of the energy required for suspending the bed and operating the venturi scrubber. Major operating variables affecting fluidized bed reactor operation are the mud drying temperature, the calcination zone temperature, the excess air level, the bed fluidization level, and the sodium content of the lime mud.

Major operating variables affecting particulate emissions from the calciner unit are the air sweep velocity and the lime feed rate. Variables affecting gaseous emissions from fluidized bed calciners are the same as those affecting reduced sulfur emissions from rotary lime kilns.

11.3 Particulate Emission Control

The major means of controlling particulate emissions from lime kiln and fluidized bed calciner exhaust gases are liquid scrubbing, using either an impingement or venturi-type scrubber, and, recently, electrostatic precipitation. The scrubbing devices are usually placed following a mechanical cyclone collector used either for removal of the larger lime dust particles, as with lime kilns, or for predrying the lime mud for fluidized bed calciners. Particulate inlet loadings to scrubbing devices from lime kilns can range from 7 to 35 g/m^3 (3 to 15 gr/cu ft) at standard conditions of 21.1° C, 1.0 atmosphere, dry gas (70° F, 29.92 in. Hg, dry gas). The dust losses constitute about 1 to 5 percent of the total dry solids load to the kiln. Particle size measurements for the above mass concentrations are not reported with these data, but the lime particles generally comprise the larger sizes and sodium particles the smaller ones (4). Comparable data are not available for fluidized bed calciners, but it is necessary to use a two-stage cyclone mud drying system to avoid overloading the venturi scrubber.

11.3.1 Scrubbing Systems

The major types of scrubbers employed on lime kilns, to date, are the impingement and venturi types, with cyclonic scrubbers also employed, but to less extent. Impingement type scrubbers were extensively employed in the past for particulate scrubbing on lime kilns and have the advantages of relatively low pressure drop and scrubber shower rate, with resultant reduced operating costs. The devices are limited, however, in their maximum scrubber slurry water solids concentrations due to possible scrubber plugging. In addition, they normally have lower particulate removal efficiencies because of less efficient gas-liquid contact. Impingement type scrubbers have higher capital costs than venturi scrubbers on similar installations basically due to their larger size and greater complexity.

Venturi scrubbers are commonly used on lime kilns at the newer kraft pulp mill installations primarily because of higher particulate removal efficiencies than achievable by the older impingement type scrubbers. Venturi scrubbing systems can operate with slurry water solids concentrations of up to 30 percent by weight without excessive plugging. A summary of operating characteristics for kraft lime-kiln scrubbers is presented in Table 11-2 (4).

TABLE 11-2
OPERATING CHARACTERISTICS FOR PARTICULATE LIQUID SCRUBBERS EMPLOYED ON KRAFT LIME KILNS (4)

	Scrubber Type	
Parameter	Impingement	Venturi
Shower rate ratio, l/m^3	0.54-2.0	1.73-3.21
($gal/10^3\ ft^3$)	(4-15)	(13-24)
Slurry solids, % by wt.	1-2	10-30
Pressure drop, mm Hg	9-13	19-28
(in H_2O)	(5-7)	(10-15)
Power required,* kW per t/day	0.041-0.049	0.082-0.099
(hp per ton/day)	(0.05-0.06)	(0.10-0.12)
Power required,** kW per t/day	0.13-0.16	0.27-0.34
(hp per ton/day)	(0.16-0.20)	(0.33-0.42)

*per mass of pulp.
**per mass of lime.

11.3.2 Performance Characteristics

A number of studies were conducted to determine particulate collection efficiencies of lime kiln scrubbers. Stuart and Bailey (6) report that venturi scrubbers were able to achieve 96-97 percent particulate removal from lime kiln exhaust gases at pressure drops of 1.7 to 2.8 kPa (7 to 10 in water); while Landry and Longwell (7) report that venturi scrubbers can achieve particulate removal efficiencies of 98-99 percent at pressure drops of 2.4 to 3.7 kPa (10 to 15 in water). A series of studies were conducted on a joint basis by the National Council for Air and Stream Improvement and the U.S. Environmental Protection Agency to establish the particulate collection efficiencies of 66 existing lime kiln scrubbers. Venturi scrubbers were able to produce consistently higher particulate collection efficiencies than the impingement scrubbers, as shown in Table 11-3 (2).

TABLE 11-3
PARTICULATE COLLECTION EFFICIENCIES FOR LIQUID SCRUBBERS ON KRAFT PULP MILL LIME KILNS (2)

Parameter	Impingement Scrubbers		Venturi Scrubbers	
	Average	Range	Average	Range
Inlet concentration,* g/m^3	27.38	8.00-33.96	18.60	5.85-31.83
(gr/cu ft)	(11.94)	(3.50-14.81)	8.11	(2.55-13.88)
Outlet concentration,* g/m^3	1.78	0.99-3.56	0.73	0.27-2.29
(gr/cu ft)	(0.78)	(0.43-1.56)	(0.32)	(0.12-1.00)
Removal efficiency, % by wt.	92.2	86.8-96.9	94.8	85.5-99.1
Emission rate,** kg/t	1.78	1.14-2.09	1.01	0.33-2.60
(or lb/ton)	(3.55)	(2.28-4.18)	(2.02)	(0.66-5.19)

*Concentrations are reported at standard conditions of 21.1° C and 760 mm Hg (70° F and 29.92 in Hg), dry gas.
**Emission rates are based on an air-dried ton of pulp basis (i.e., 10% moisture, by weight).

Information developed during the study indicates that high pressure drop venturi scrubbers can achieve significantly lower particulate levels than reported in Table 11-3 (2). Particulate concentrations at standard conditions of between 0.02 and 0.11 g/m^3 (0.01 to 0.05 gr/cu ft), corresponding to emission rates of 0.01 to 0.05 kg per air dried metric ton of pulp (0.02 to 0.1 lb/ton), were measured. Very little information exists regarding particulate emission control following fluidized bed calciners. Erdman (8) reports on a high pressure drop venturi scrubber following a two-stage cyclonic mud drying system. The pressure drop through the venturi is 5.4 kPa (22 in water). Although the dust carryover from the calciner section is 12 percent, the scrubber emits a particulate concentration of 0.16 g/m^3 (0.07 gr/cu ft), which corresponds to an emission rate of 0.24 kg per metric ton of pulp (0.49 lb/ton).

11.4 Gaseous Emission Control

Lime mud calcining in rotary kilns or fluidized bed reactors can emit H_2S, organic sulfur, SO_2, and nitrogen oxides to the atmosphere. The gaseous emissions result either from materials entering the calcining unit system or from materials entering the kiln. The major process operating variables affecting gaseous emissions include excess air level, operating temperature, and solid and gas-phase retention times.

Major input material properties affecting gaseous emissions include the respective Na_2S contents of the input lime mud and scrubber water, organic sulfur levels in the inlet scrubber water, and the moisture content of the lime mud. The major design variable affecting gaseous emissions from the calcining system are the length and, to a lesser extent, the diameter for rotary kilns, and the diameter and height for fluidized bed calciners.

A summary of gaseous emissions from rotary lime kilns and fluidized bed calciners is presented in Table 11-4.

TABLE 11-4
GASEOUS EMISSIONS FROM KRAFT PULP MILL LIME KILNS (2)

Gaseous Constituent	Concentration Average	Concentration Range	Emission Rate Average	Emission Rate Range
	ppm, by volume		kg sulfur per t pulp (lb sulfur per ton pulp)	
H_2S	108	0-500	0.24 (0.48)	0-1.88 (0-3.76)
CH_3SH	14	0-90	0.03 (0.07)	0-0.17 (0-0.34)
CH_3SCH_3	27	0-245	0.02 (0.05)	0-0.22 (0-0.43)
CH_3SSCH_3	5	0-11	0.01 (0.03)	0-0.10 (0-0.20)
TRS	–	–	0.31 (0.63)	0-2.37 (0-4.73)
SO_2	34	0-140	0.14 (0.28)	0-1.11 (0-2.20)

11.4.1 Lime Mud

The most important gaseous emissions from lime reburning systems are malodorous reduced sulfur compounds. Hydrogen sulfide can be volatilized from the Na_2S present in the lime mud by contact with CO_2 from the flue gas. Above a threshold Na_2S concentration of 0.2 percent by weight, the generation of H_2S is directly proportional to the residual Na_2S content of the lime mud. This linear relationship is similar to that for direct contact evaporation (9). The amount of H_2S released can be controlled by reducing the residual Na_2S level by more efficient lime mud washing. It is not normally feasible, however, to reduce the residual sodium content in the lime mud to less than 0.1 percent by weight because of possible mud ring formation.

Prakash and Murray (5) report that H_2S emissions from the lime mud occur primarily from Na_2S dissolved in the aqueous portion and not from the solid portion of the lime mud. The H_2S emissions can be reduced by drying the mud to a solids concentration of 70 percent by weight or more before burning.

11.4.2 Scrubbing Water

The scrubber water can be a source of both H_2S and organic sulfur compounds emissions from kraft mill calcining units equipped with scrubbers. The presence of Na_2S in the scrubber water can result in the release of H_2S by contact with CO_2 if the liquid pH is sufficiently low. The emission rate of H_2S and organic sulfur compounds increases with the inlet Na_2S and organic sulfur concentrations, with rising liquid- and gas-phase temperatures, and with an increasing degree of gas-liquid contact, as represented by the scrubber pressure drop. The potential for organic sulfur release is particularly great if untreated digester or evaporator condensates are used as lime kiln scrubber makeup water.

Caron (11) reports that using lime mud wash water instead of fresh water for lime kiln scrubbing results in stripping of 0.10 to 0.22 kg sulfur per metric ton of pulp (0.2 to 0.4 lb sulfur/ton) as compared to an absorption of only 0.035 kg sulfur per metric ton of pulp (0.07 lb sulfur/ton) with fresh water. Normally, fresh water should be employed as the scrubbing medium to avoid the stripping of odorous gases. If condensate waters are employed, steam stripping should be employed prior to the scrubbing operation.

One U.S. mill has significantly reduced TRS emissions from a lime kiln venturi scrubber by adding sodium hydroxide to the scrubber water to raise the pH. The scrubber water is recycled to the causticizing system (10).

11.4.3 Combustion Variables

The major combustion variables that can affect reduced sulfur emissions from lime mud calcining operations are the excess air level, the temperature profile, and the mud retention time in the kiln. Caron (11) reports that the TRS emissions from the combustion zone are minimized at excess oxygen levels of four percent by volume or greater. Though no definite patterns have been established, the kilns that have cooler wet-end temperatures tend to have relatively higher reduced sulfur emissions because the sulfur compounds can be volatilized without burning. Sufficient retention time must be provided at temperatures above 760° C (1400° F) to oxidize the reduced sulfur compounds.

Walther and Amberg (12) report that shorter lime kilns tend to have lower reduced sulfur emissions than longer lime kilns, though no definite correlation could be established. The probable reason is that short lime kilns must operate at higher average temperatures throughout than the long kilns to achieve an equivalent degree of calcination. The result is a more complete oxidation of reduced sulfur compounds. An additional factor is that the evolution of Na_2S at low temperatures in oxidizing atmospheres promotes H_2S formation; its evolution at higher temperatures promotes SO_2 formation (13).

Limited data indicate that reduced sulfur emissions from fluidized bed calciners are minimal. This may be due to the relatively long retention time at uniformly high temperature which provides for efficient oxidation of the sulfur compounds (14). One test shows an emission rate of less than 0.01 kg sulfur per metric ton of pulp (0.02 lb/ton). Flash drying of the mud tends to minimize H_2S formation in the fluidized bed units.

The burning of digester and evaporator noncondensable gases in the lime kilns brings an additional source of sulfur compounds to the units. The conversion of these materials to SO_2 is essentially complete because they are added with the primary air at the hot end of the lime kiln and so have sufficient retention time for complete combustion to take place (15). The addition of green liquor dregs with the lime mud to the cold end of the lime kiln can substantially increase the reduced sulfur emissions, because these materials are normally contaminated with Na_2S from the green liquor. There is also insufficient retention time at high enough temperatures for complete oxidation to take place.

11.4.4 Sulfur and Nitrogen Oxides

The concentrations of sulfur oxides in lime-kiln exhaust gases are normally minimized because the CaO can act as an efficient adsorption and reaction medium to form $CaSO_3$ and $CaSO_4$. Long kiln length, with sufficient oxygen and high calcination efficiencies, promote efficient SO_2 removal. To date, no adverse effects on lime kiln operating efficiency were traced to the sulfur released by the burning of either residual fuel oil or noncondensable gases. In a limited series of tests, it was not possible to measure the presence of SO_2 in the exhaust gases of a fluidized bed calciner, probably because the calcining and flash drying provided a two-stage removal system.

Galeano and Leopold (16) report that the lime kiln is the only major process source where significant quantities of nitrogen oxides can be measured. The primary reasons for the presence of oxides of nitrogen are that there is sufficient excess air at a temperature of 1200 to 1300° C (2200 to 2400° F) to promote the reactions. The amounts of nitrogen oxides formed in the fluidized bed calciners are probably significantly less than from rotary kilns because of the lower operating temperatures of 825 to 875° C (1500 to 1600° F). To date, no specific tests have been conducted to determine the amount of oxides of nitrogen in fluidized bed calciner exhaust gases.

11.5 Oxygen Addition

Molecular oxygen can be added to the combustion air of a lime kiln to control H_2S generation from the lime mud in the combustion zone. The oxygen must be added together with the primary air to the firing zone at the dry end of the lime kiln. This practice will promote effective mixing with the combustion gases and will provide for complete oxidation of any H_2S released from the mud. Precautions should be taken to assure that overheating of the kiln does not occur in localized areas. Such overheating could damage refractory materials, interfere with kiln operation, or result in increased emissions of oxides of nitrogen.

There is very limited field experience, to date, with the addition of oxygen to lime kilns for reducing H_2S emissions. Singman (17) reports that oxygen addition to lime kilns can substantially increase the lime mud throughput rates for previously overloaded lime kilns without excessive lime losses. An addition of 0.454 kg (1 lb) of oxygen results in a net decrease in lime makeup rate of 1.8 kg (4 lb) as CaO and, consequently, a considerable savings in operating costs for causticizing. Decreases in H_2S emissions may result for relatively short lime kilns, particularly where higher temperatures are maintained at the wet end. Additional process variables that would affect H_2S emissions in addition to kiln length include kiln diameter, mud washing efficiency and inlet sulfide level, mud firing rate and solids concentration, and gas velocities at different locations in the kiln.

11.6 Process Economics

The primary economic factor to consider for effective air pollution control of lime-calcining systems is the installation of devices for particulate control. The respective capital and operating costs for impingement and venturi scrubbing devices are presented in Table 11-5 (4).

Gaseous emission control does not normally require substantial capital investment unless flash drying of lime mud must be instituted. Maintenance of sufficient excess air, proper washing of lime mud, and the use of fresh water normally are sufficient to minimize gaseous

TABLE 11-5
CAPITAL AND OPERATING COSTS FOR LIME KILN PARTICULATE SCRUBBERS (4)

Cost Item	Scrubber Type: Impingement	Scrubber Type: Venturi
Capital cost,* \$/daily t pulp	30-33	22-27
(\$/daily ton pulp)	(27-30)	(20-25)
\$/daily t lime	99-110	71-93
(\$/daily ton lime)	(90-100)	(65-85)
Annual operating cost,** \$/t pulp	3-7	7-11
(\$/ton pulp)	(3-6)	(6-10)
\$/t lime	11-22	22-38
(\$/ton lime)	(10-20)	(20-35)

*Based on 1966 data.
**Based on 0.9 cents/kWh.

emissions. Addition of NaOH to the particulate scrubber makeup water to minimize H_2S emissions by increasing liquid pH levels will increase operating costs.

11.7 Lime Dust Handling

A minor source of fugitive particulate emissions from the causticizing system of a kraft pulp mill consists of lime dust releases from storage tanks and bins, and conveying and transfer facilities. Activities where lime dust is loaded or unloaded, dumped, or transferred are particular problems because of the dryness of the material that is handled. The lime dust is a localized emission source within the immediate area of the causticizing plant.

The three major approaches to control fugitive lime dust emission are to:

1. Confine the potential emission sources to prevent air leakage,

2. Wet the dust to prevent its becoming airborne by wind or by transfer operations, and

3. Use special air pollution control equipment.

The first method is effective in limiting the potential sources or fugitive emissions by effective housekeeping. It also facilitates the subsequent installation of particulate air pollution control equipment. Wetting the dust is effective in controlling fugitive dust emissions, but it can make the lime difficult to handle if overdone. It generally is not recommended as an effective technique for controlling lime dust emissions.

The third method for controlling dust emissions from lime storage and transfer facilities involves the use of particulate control techniques, such as centrifugal separation, liquid scrubbing, and fabric filtration. Keeping the dust as dry as possible facilities its recovery; therefore, liquid scrubbing is undesirable. Centrifugal collectors are not advantageous in that they require high pressure drops and tend to have low collection efficiencies.

The only known installation for particulate lime dust recovery from storage facilities employs a fabric filter baghouse for a 907 metric ton per day (1000 ton/day) kraft pulp mill. The air vents from the lime storage tanks are vented into a central duct and passed to a baghouse with a design flow rate of 4,400 m^3/h (2,600 cfm) at a maximum temperature of 290° C (550° F). The filter bags have a total surface area of 121 m^2 (1,300 ft^2) with a cleaning cycle of once each 20 minutes. The filter bags are made of a siliconized glass cloth with a design ratio of air to cloth filter area of 22.8 to 27.4 $m^3/h/m^2$ (1.25 to 1.50 cfm/ft^2).

Total capital cost for the system was $5,900 in 1966. The fans have a total capacity of 13.4 kW (18 hp) with a resultant direct annual operating cost of $1,080 per year. The system can recover 225 to 450 kg/day of lime (500 to 1,000 lb/day), which is equivalent to an annual savings of $1,800 to $3,600, based on a lime price of $22/t ($20/ton) (18, 19).

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