Steam drying and fluidized-bed calcination of lime mud

Charlotta Hanson and Hans Theliander

ABSTRACT: A novel lime-mud-reburning system is described wherein lime mud is dried in a steam dryer and then calcined in a fluidized bed. Energy in the flue gases leaving the calcination unit is recovered in a flue-gas boiler. Calculations show that a system with a steam dryer utilizes the energy input better than conventional systems, where flue gases dry the lime mud. Experiments in a pilot-scale steam dryer show that lime mud (initial dry solids of 69–71%) was dried to >99% dry solids. The dried lime mud was then heated and calcined in a pilot fluidized-bed reactor at about 900° C. The resulting "soft burned" lime was highly reactive during slaking and causticizing. However, it produced a lime mud with a higher specific filtration resistance than reburned lime from industrial rotary kilns.

KEYWORDS: Calcination, drying, fluidized beds, steam, white liquor mud.

There are two commercial systems used to reburn lime mud. One is based on a rotary kiln, as illustrated in **Fig. 1**. The first rotary kiln for reburning of lime mud was supplied by F.L. Smidth in 1905 to Stora Kopparberg AB, Sweden *(1)*. The other system is based on a fluidized bed combined with a flue-gas dryer, as seen in **Fig. 2**. The Fluosolids fluidized-bed system has been in use since 1963 *(2)*.

In both systems, the heat for heating, drying, and calcination of lime mud is supplied by flue gases from the combustion of organic fuels. The carbon dioxide content in the flue gases determines the lowest flue-gas temperature needed for the calcination, according to the equilibrium of the reaction. If the heat transfer from the gas to the solid phase inside the reactor is insufficient, the temperature sometimes has to be much higher than the equilibrium temperature to give a reasonable reaction rate.

In the rotary kiln, the solid phase remains at the bottom of the reactor while the flue gases sweep over the bed. The rate of heat transfer between the flue gases and the solids in the bed is low. Furthermore, investigations have shown that a radial temperature profile is developed in the bed *(3)*, indicating that the solid material does not have a uniform temperature. This implies that the flue-gas temperature must be held at a high level in some regions of the kiln to ensure total calcination. This—plus the fact that a temperature profile develops along the kiln *(4)*—makes it difficult to control the quality of the produced lime.

The fluidized-bed system gives a lime product of consistent quality, according to the manufacturer *(5)*. A fluidized bed provides fast heat and mass transfer *(6)*, the result of a large area for transfer of heat and mass as well as high heat-and mass-transfer coefficients. The good mixing conditions in a fluidized bed also help maintain a uniform temperature. These conditions in the fluidized bed are favorable for producing a reburned lime mud of uniform quality. The fast heat and mass transfer also has positive effects on the calcining temperature, since it can be kept close to the equilibrium temperature.

In both of these conventional systems, high-temperature (900°C) flue gases from the calcination process are used to heat and dry the lime mud. From a technical standpoint, a temperature of 200°C is sufficient to dry lime mud. The use of 900°C flue gases to dry lime mud is clearly a waste of high-quality energy.

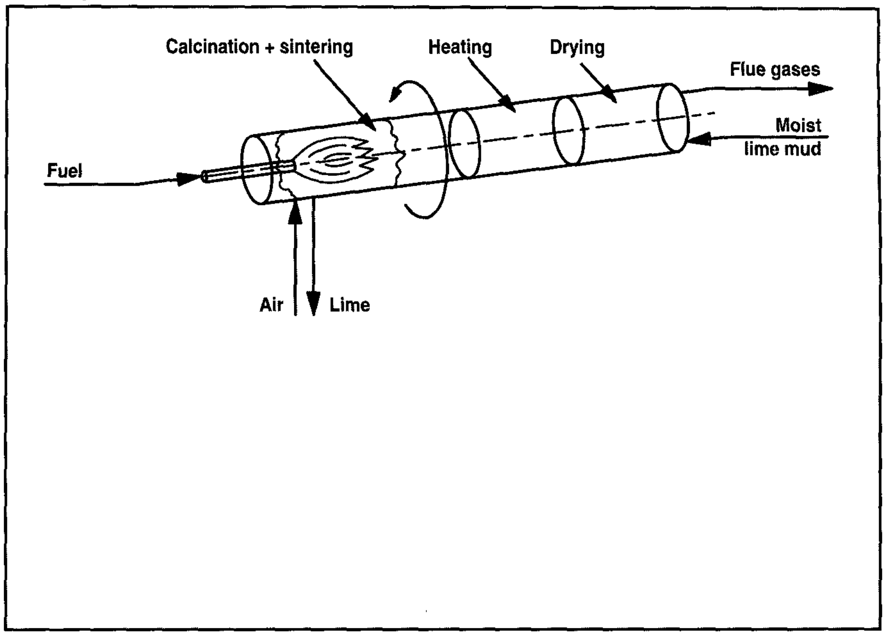
Two novel lime-mud-reburning systems have been developed recently. One system, developed by Fuller Co., is based on a flash calciner and a flue-gas dryer *(7)*. The second system, de-

Hanson, graduate student, and Theliander, associate professor, are affiliated with the Dept. of Chemical Engineering Design, Chalmers University of Technology, S-41296 Göteborg, Sweden.

veloped at Chalmers University of Technology *(8)*, is based on a fluidized bed, a steam dryer, and a flue-gas boiler. This report describes the second system and compares its performance with that of conventional systems.

When comparing different systems, there are many aspects that should be taken into account. It is not possible to treat all of them in the available space. Consequently, key aspects are selected and examined in greater detail. The report presents a theoretical discussion that focuses on energy demand and utilization in the various components of the lime-mud-reburning system. Results of an experimental investigation of the properties and quality of the product lime also are presented.

1. *Rotary kiln*



Novel lime-mud-reburning system

The novel lime-mud-reburning system consists of a steam dryer, a fluidized-bed reactor, and a flue-gas boiler, as illustrated in **Fig. 3.**

The steam dryer resembles a vertical-tube heat exchanger. Moist lime mud is fed into the steam dryer and suspended in superheated carrier steam. The pressure of the carrier steam is preferably chosen to match the low-pressure steam network in the mill. The degree of superheating of the carrier steam is controlled by letting steam of higher pressure and temperature condense on the outside of the tubes. The pressure on the heating steam can be chosen to fit the back-pressure steam network.

The dry lime mud is separated from the carrier steam in a cyclone. The lime mud is thereafter removed from the dryer. A certain amount of carrier steam, corresponding to the amount of liquid vaporized from the lime mud, is also removed from the dryer. This steam can be re-formed and used for various heating purposes in the mill. The remaining part of the carrier steam is recirculated into the feed of moist lime mud. Thus, in this type of dryer, steam of approximately 10 bars is used to heat and dry the moist material. However, the heat of vaporization is recovered in steam at a useful pressure level, close to 5 bar.

The heart of the novel system is the reactor, a fluidized bed (Fig. 3). The reactor actually consists of two or three fluidized beds. The upper bed is the calcining bed, and the lower beds are combined lime coolers and air preheaters. The dry lime mud and the fuel are fed directly into the calcining bed, similar to the "Fluosolids" reactor.

The third part of the system is the flue-gas boiler, where the heat of the flue gases leaving the reactor is recovered. If (a) the calcining temperature is 900°C, (b) the flue gases leaving the boiler have a temperature of 190°C, and (c) the dry content of the incoming lime mud is 65%, then the heat of vaporization in the produced steam (10 bar) corresponds to the heat demand in the dryer. If the dry content is lower than 65%, external heating steam must be supplied to the system. This would imply production of more low-pressure steam from the extra moisture in the lime mud. On the other hand, if the dry content is higher than 65%, a surplus of steam will be produced in the flue-gas boiler.

If the price of electric power is high, an alternative configuration may be of interest. **Figure 4** illustrates a system with a flue-gas boiler designed to produce high-pressure steam for generation of electric power. In this modified process, the heating steam to the dryer would be taken from the mill's steam network.

Energy demand and utilization

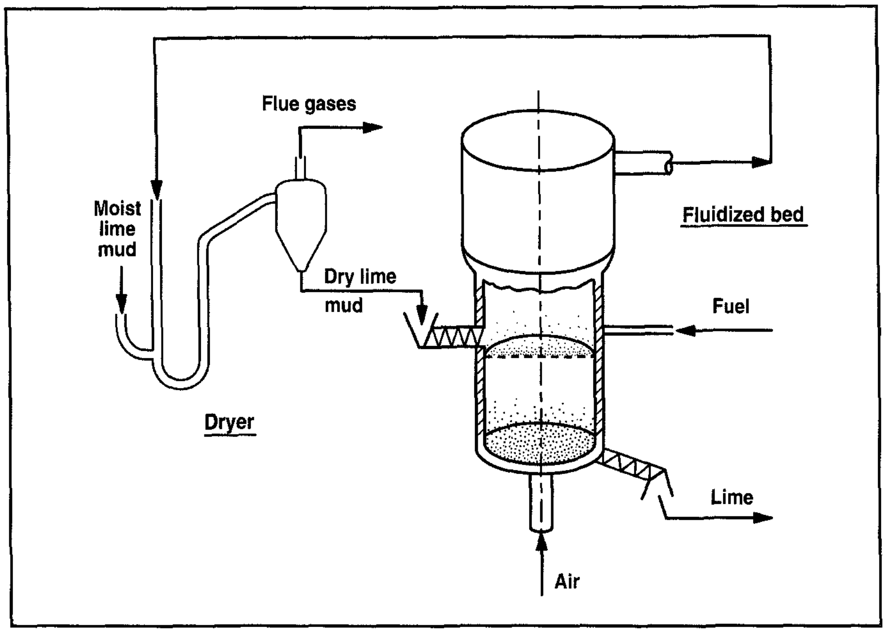
The input of primary energy to the lime-mud-reburning system is mainly determined by the calcining temperature. In this discussion, a calcining temperature of 900°C has been assumed. If either the calcining temperature or the temperature of the flue gases leaving the heating section could be decreased, the demand for primary energy would of course decrease in any of the investigated systems. A condition for such a decrease would be that the input of energy to the drying stage remain sufficient.

Table I lists the values for parameters used to calculate energy demand and utilization for the novel system and conventional systems. The energy demand is represented by a change of enthalpy in the system. The utilization of energy is represented by a change of entropy.

Energy demand

The energy demand in the calcining and heating steps is mainly influenced

2. Fluidized-bed system



by the calcining temperature, while the amount of heat needed for drying depends on the dry content of the entering lime mud. **Figure 5** shows the changes of enthalpy in the conventional system and in the novel system. The negative values represent the cooling of lime and carbon dioxide from the calcination and—in the case of the novel system—the condensation of steam from the steam dryer.

As long as the flue-gas temperature is kept at the same level in both systems, the enthalpy changes are almost the same in the heating and calcining stages. The difference appears in the drying stage, where the novel system has an advantage over the conventional system. Because the moisture from the lime mud is recovered as steam in the novel system, the heat of condensation is easily utilized. This utilization is more difficult in the conventional system, where the vapor is mixed with flue gases from the combustion and with the carbon dioxide released from the calcination.

The mixture of vapor in the flue gases of a conventional system also makes it less profitable to increase the dry substance in the lime mud. A decrease in the demand for drying heat means higher temperatures in the flue gases leaving the kiln. This also means higher temperatures for the vaporized moisture from the lime mud. The surplus of energy added in the drying stage is difficult to recover. In the novel system, a higher dry content of the lime mud implies a surplus of steam produced in the flue-gas boiler, steam that can be used elsewhere in the mill.

An advantage of the rotary kiln is that the heat in the flue gases is delivered countercurrently with the solid phase in the rotary kiln. This makes it possible to decrease the energy demand to some extent when the dry content is decreased, provided the kiln is long enough. This could be achieved in the fluidized-bed system as well, if the lime mud is heated countercurrently by the flue gases either before entering, or inside, the reactor.

Energy utilization

The increase of entropy in a system should be minimized in order to utilize the energy input efficiently. The increase of entropy in the calcining, heating, and cooling stages will be about the same in the investigated systems. **Figure 6** shows that the entropy increase is much lower in the novel drying system. This is because, in a conventional rotary-kiln system, the drying is performed directly in high-temperature flue gases. The novel system uses steam in the dryer. The flue gases can then be used to produce electricity.

Characteristics of product lime

Experimental methods

Steam dryers. Two pilot-scale steam dryers—one batch and one continuous—were used to investigate the properties and quality of steam-dried lime mud. Both are of single-tube construction, with a tube diameter of 100 mm. All tubes in the drying sections have steam-heated jackets.

The pilot steam dryer in **Fig. 7** is a batch dryer and has a height of 11 m. A batch of lime mud was loaded into this dryer and circulated until drying was completed. The drying process was monitored by reading the temperature inside the dryer. Drying was assumed to be complete when the temperature was stabilized. The pressure of the carrier steam was about 2.5 bar, and it was superheated to 170°C before a test was started. The heating steam was saturated steam at 180°C.

The pilot steam dryer in **Fig. 8** is a continuous dryer with a total length of 200 m. The moist lime mud is fed via a rotary vane feeder and a transport screw into the dryer. The dry lime mud is removed from the dryer via a cyclone and a second rotary vane feeder. The surplus carrier steam is leached out, and the remaining carrier steam is recirculated via two blowers and an electrical superheater. The pressure of the carrier steam was about 2 bar, and it was superheated to 200–250°C before the moist lime mud was suspended. The heating steam was saturated steam at 170°C.

Pilot calciner. The semi-batch calciner consisted of a lime-mud feed system, a reactor (i.e., the fluidized bed), four cyclones, and a scrubber, as seen in **Fig. 9**.

Dry lime mud is fed from a small silo via two transport screws to the reactor. The inner diameter of the reactor is 150 mm, and the height is 2 m. Heat is supplied by two LP-gas burners, one on each side of the distribution plate. The upper burner was used only during heatup to the operating temperature. The reactor is followed by four cyclones. The first one is large and separates the large particles. The three small parallel cyclones separate the small particles.

Tests were carried out using two different lime muds. Average calcination temperatures ranged from 800°C to 920°C. Temperature was controlled to within ± 30°C by the feed of lime mud, with peaks up to ± 60°C because of variations in the degree to which the screw feeder was filled.

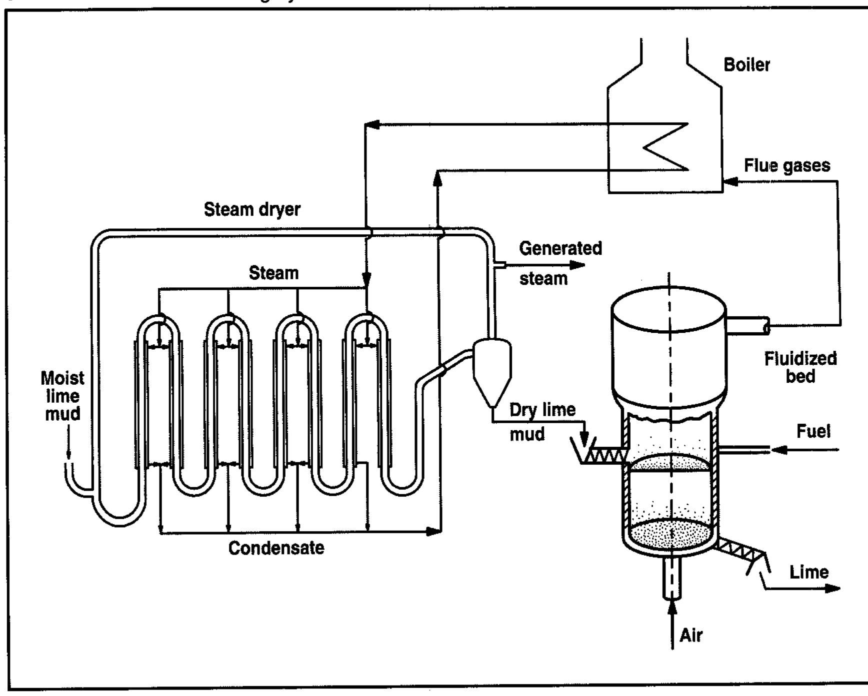
Each calcination run began by heating the system to the desired temperature. After system heatup, dry lime mud was fed into the reactor for 60 min. Thereafter, the reactor was shut down and the system was allowed to cool down. The lime was removed about 12 h later at a temperature of about 200°C.

To test whether this cooling period influenced the final results, the 12-h cooling period was repeated in a laboratory furnace. It was found that the lime mud did not calcine even at the highest temperature measured during the cooling process. Still, samples removed from the pilot-scale refiner before and after the cooling period had different contents of calcium carbonate. The probable explanation of this is that the before-and-after samples were obtained in different ways. Samples before cooling were taken just below the screw feeder, while cooled samples were taken out of the mixed bed. The samples obtained before cooling may have contained remnants of unreacted lime mud.

Laboratory oven. Lime muds were also reburned in a laboratory furnace to provide a reference for comparison with the lime muds that had been reburned in a fluidized bed. The investigated temperature interval in this case was 850–1300°C.

The laboratory oven is a high-temperature furnace (PHOENIX Sigma 3) equipped with a programmable temperature controller. The inner dimensions are 355 mm x 180 mm x 130 mm.

3. Novel lime-mud-reburning system



The atmosphere inside the furnace was controlled by sweeping 10 L/min of nitrogen over the sample.

Evaluation of calcined lime mud. The quality of the calcined lime mud was evaluated by measuring the contents of $CaCO_3$ and CaO, specific surface area (B.E.T. theory), slaking and causticizing rates, and specific filtration resistance. Test methods are described elsewhere *(12, 13)*.

Results

Steam dryer. Tests in the batch dryer showed that the drying of lime mud was easy to accomplish. The dry content of the lime mud increased from 63–69% to 85–95% within 10–20 s.

Several minor tests in the continuous dryer during the past few years have produced a lime mud with a dry content between 96% and 99%. After a major rebuilding of the dryer, a larger test was performed. The test period was 3 days, during which 9 tons of moist lime mud were dried. The dry content of the incoming lime mud varied between 69–71%, and it was dried to a dry content higher than 99%.

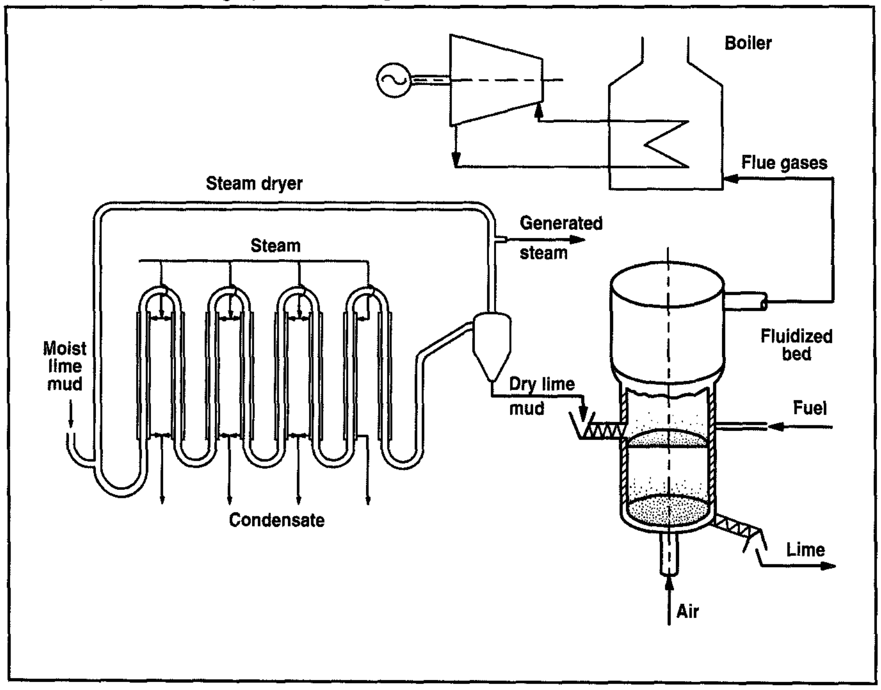
Figure 10 shows the temperature of the carrier steam measured at different positions in the steam dryer during a test run. Temperatures were measured in the superheated steam before the moist lime mud was suspended and after the first, second, and third riser, as illustrated in Fig. 8.

As can be seen, the temperature drops very swiftly in the first riser (Curve 2). Thereafter, it increases to approximately 170°C, which is the saturation temperature of the heating steam. Thus, the lime mud has become dry at some position between the first and second riser. This translates to a drying time of 1–2 s, which indicates that the drying of lime mud is very fast and easy to accomplish.

The sharp peaks in temperature after the first riser are due to variations in the feed rate and in the dry content of the moist lime mud. After approximately 3500 s, the load was increased, and this caused the average values of the temperatures after the first and second riser (Curves 2 and 3, respectively) to decrease.

Pilot calciner. The lime mud formed pellets almost immediately. After 10 min, particles/pellets could be seen jumping around inside the reactor. This activity was observed through a sight hole located 10 cm

4. Novel system with high-pressure flue-gas boiler



I. Input data for calculations

Lime mud	
Temperature	60°C
Dry solids	70%
Lime	
Temperature	200°C
CaO content	100%
CO_2 temperature	200°C
Flue gases	
Temperature	200–900°C
CO_2 content	30%
N_2 content	70%
Conventional system	
Vaporized moisture	1 bar, saturated
Novel system	
Steam	5 bar, saturated
Condensate	5 bar, saturated liquid

Steam data from Kell (*9*)
Solid-phase data from Barin& Knacke (*10*)
CO_2 and N_2 data from Van Wylen (*11*)

above the perforated plate. The sight hole then slowly filled up with particles. These particles could be blown away by air, but they immediately refilled the sight hole. This made it impossible to observe the conditions inside the reactor as the experiment proceeded.

As can be seen in **Fig. 11**, most of the pellets were 1–5 mm in diameter. The mechanical stability of the pellets was poor, but more important is the fact that the reburned lime mud was not a dusty product. The lime pellets captured in the first cyclone were smaller than 0.5 mm. A fine powder was taken from the last three cyclones.

Layers of lime were found on the walls and the perforated plate within the reactor. These layers were soft, a few millimeters thick, and easily removed. This lime was not further analyzed.

Calcination index. The calcination index (CI) is defined as the ratio between calcium oxide (lime) and calcium oxide plus calcium carbonate.

$$CI = W_{CaO}/\{W_{CaO} + [W_{CaCO3}(M_{CaO}/M_{CaCO3})]\} \times 100\%$$

where

W = weight fraction

M = molar weight.

Thus, if the calcination index is 100%, the lime mud has been completely reburned. Limes produced in the pilot fluidized bed had calcination indices between 70% and 98%.

Slaking rate. **Figure 12** shows the slaking rate of lime produced in different ways vs. the specific surface area of the lime. Lime from the pilot-scale fluidized bed had a high reaction rate and a high specific surface area compared with the limes produced in the laboratory oven or in an industrial rotary kiln. The data in Fig. 12 also reveal a trend that is independent of the choice of equipment. Generally, a higher specific surface area yields a higher slaking rate.

Specific filtration resistance. **Figure 13** shows the specific filtration resistances of the lime muds produced from the different limes. The lime mud from the pilot-scale calciner gave the highest specific filtration resistance. As seen in Fig. 13, specific filtration resistance of the lime mud tended to increase with increasing specific surface area of the calcined lime.

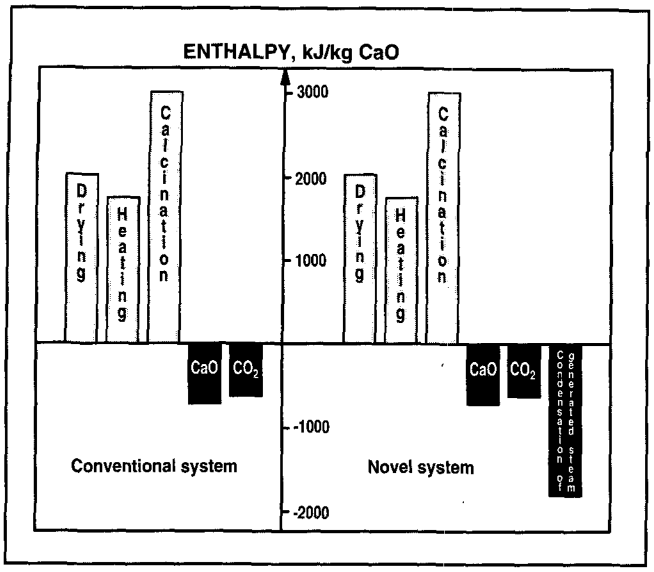
Discussion

The novel system is flexible, with the calcining and drying equipment operating independently of each other. Thus, controlling the novel system is much simpler than controlling either of the two conventional systems. It is also possible to find different ways of utilizing the energy in the high-temperature flue gases from the calcination process.

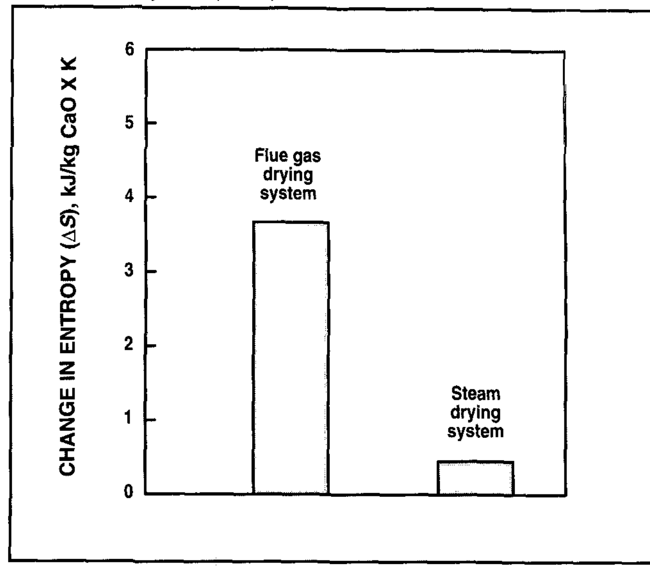
Steam drying. Valuable operational experience was obtained during the drying tests, especially during the period where 9 tons of lime mud were dried in the continuous steam dryer. Problems during this test period were mainly related to the lime-mud feed system. The greatest problem was ensuring that the temperature of the feed equipment was kept above the condensing temperature. If the steam condensed, the lime mud began to clog and arches were formed. This problem can be avoided if the equipment is insulated. As it was, the dryer operated satisfactorily most of the time.

The difference between the pilot steam dryer and a full-scale steam dryer is the size of the feed and discharge equipment, the type of superheater, and the numbers of tubes. The design of the pilot system simplifies the scale-up of industrial equipment. Data from this pilot dryer have been used to develop steam-drying systems for a variety of materials, including pulp, hogged fuel, lumber, and bark (*14*).

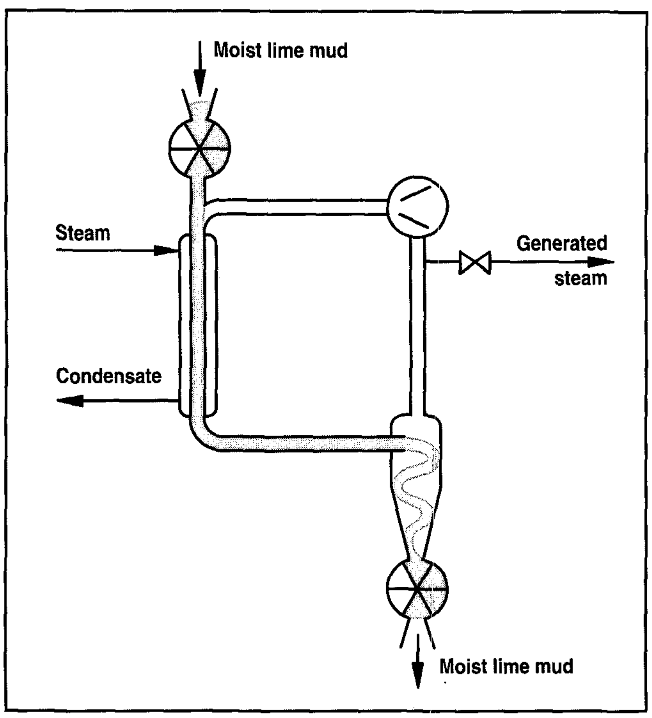
5. Enthalpy changes in conventional and novel systems



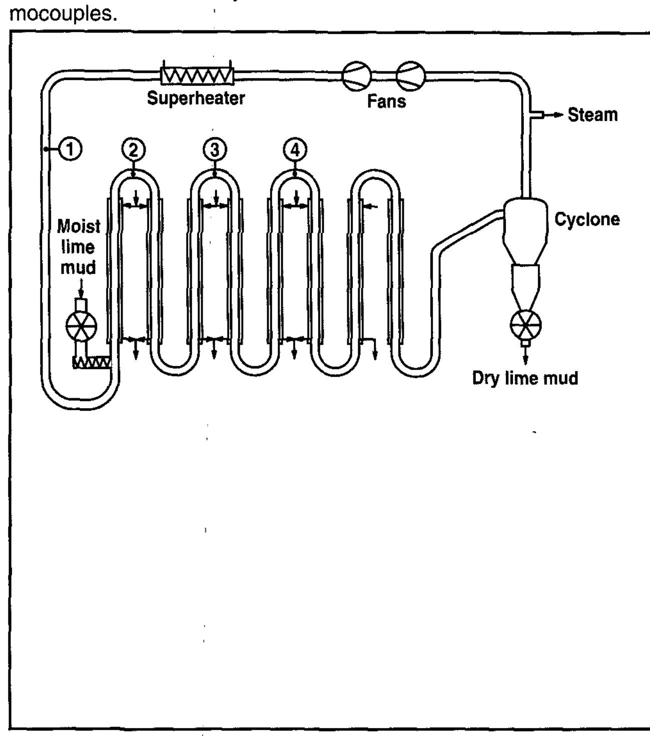
6. *Increased entropy in flue-gas drying system (conventional) and steam drying system (novel)*



7. Batch steam dryer



8. Continuous steam dryer. Circled numbers denote location of thermocouples.

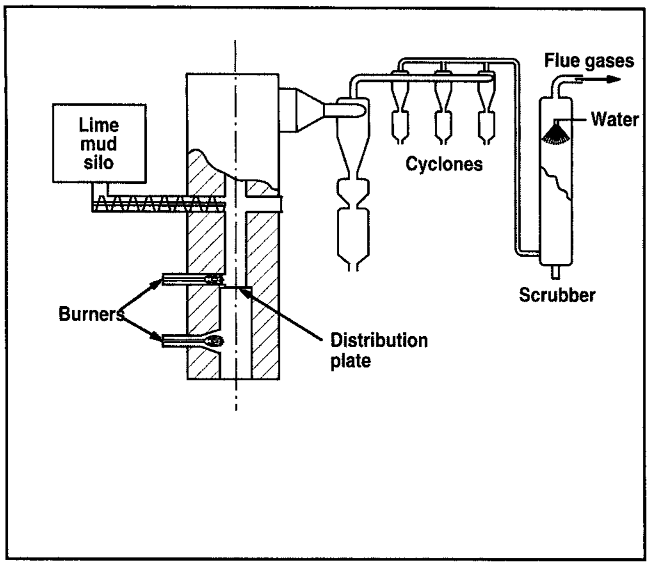


Pilot calciner. The spontaneous formation of pellets in the calciner was an important result. The mechanism of the pelletizing is not known, but the same results were obtained with the two investigated lime muds. Some of the smaller pellets may have been formed when the lime-mud powder became attached to the grains in the bed. The other larger pellets probably were not fluidized and may have increased in size by granulation at the bottom of the bed.

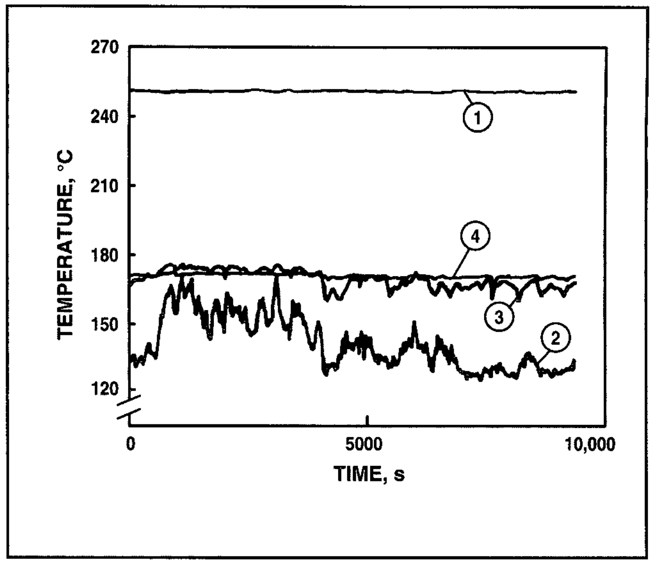
Because the experiments in the pilot-scale reactor were run batchwise, it was not possible to evaluate the uniformity of the product lime. Furthermore, the design of the burner and the cooling compartment differs from that of an industrial plant. Since these circumstances may have influenced the results, complementary experiments are now being performed in a larger-scale reactor.

The lime calcined in the pilot fluidized-bed calciner was more reactive than the other limes investigated. It

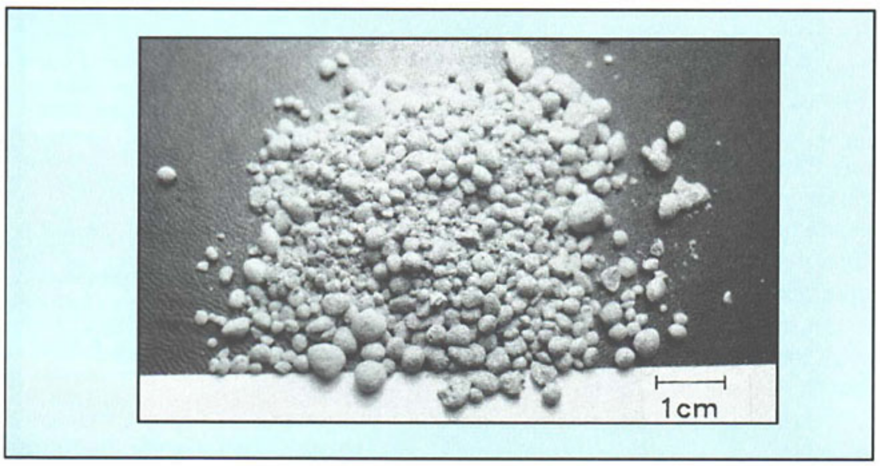
9. Pilot calciner



10. Temperature profiles at four locations in continuous pilot steam dryer. Numbers correspond to thermocouple locations depicted in Fig. 8.



11. Lime pellets from pilot calciner



also gave a lime mud with a higher filtration resistance. High reaction rates are reported from the Fluosolids system as well *(15)*. The limes calcined in a fluidized-bed thus can be characterized as "soft burned."

The quality of reburned lime is dependent on the specific surface area of the lime *(13)*. The large specific surface area of the lime obtained in this study may explain why it is reactive and yields a lime mud with high specific filtration resistance. Since the temperature is relatively low in the fluidized bed, the sintering processes are slow, which may explain the large specific surface area of the lime. The calcination index also may influence the specific surface area of the product lime *(12)*.

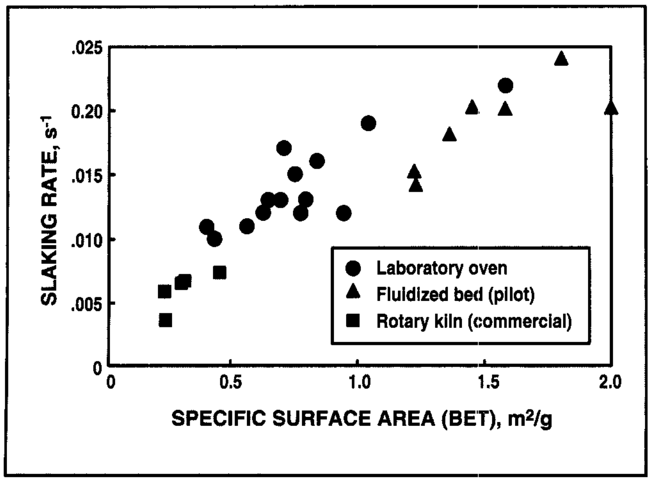
The lime showed the same tendencies regardless of whether it was produced in the pilot fluidized bed, the laboratory oven, or in an industrial rotary kiln. An increase of the specific surface area of the lime was associated with an increase in the reaction rates in the slaking and causticizing stages. A larger specific surface area of the lime also increased the filtration resistance of the filter cake when the lime mud was separated from the liquor. Thus, the requirements of a fast-reacting reburned lime mud and a low specific filtration resistance are contradictory.

The lime reburned in the pilot fluidized bed was more reactive in the slaking and causticizing processes than the lime from an industrial rotary kiln. High reaction rates translate to a shorter residence time for slaking and causticizing. This, in turn, decreases the dead time in the measuring and control loops of the system. The result is more accurate control of the slaking and causticizing units and, in turn, better control of white-liquor quality. With a fast-reacting lime, it is also easier to obtain a white liquor with concentrations of hydroxide and carbonate ions close to the equilibrium conditions. This eliminates the risk of overliming, which is of utmost importance for the filtration operation.

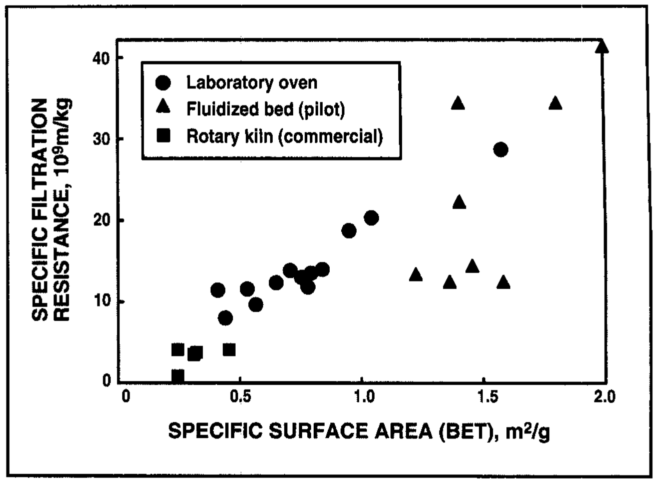
Filtration resistance was higher for the lime mud produced using lime obtained from the pilot-scale calciner. The higher resistance implies a need to increase the filter area. However, since the risk of overliming decreases and the quality of the lime can be more uniform, the filtration is more easily controlled. To some extent, the higher filtration resistance will be compensated by the fact that the filter may be easier to control and optimize.

In cases when it is important to obtain a more "hard burned" lime, it is

12. Slaking rate of lime as a function of specific surface area of the lime



13. Specific filtration resistance of lime mud as a function of specific surface area of the lime



possible to increase the calcining temperature, although this increases the energy demand. To keep down the input of primary energy, it is of utmost importance to preheat the dry lime mud before it is fed to the calcining bed.

Whether to choose a "hard burned" or a "soft burned" lime is a manner of optimization. A low energy demand in the kiln is reached when the calcining temperature is low. A low calcining temperature yields a lime with a high specific surface area, which can be characterized as "soft burned." The "soft burned" lime is reactive but has a higher filtration resistance. A higher calcining temperature yields a more "hard burned" lime, a material that reacts more slowly but produces a more easily filtered lime mud.

Concluding remarks

The novel lime-mud-reburning system dries the lime mud in a steam dryer and calcines the dried lime mud in a fluidized bed. The energy in the flue gases leaving the calcination unit is recovered in a flue-gas boiler. Such a system is more energy efficient and flexible than lime-mud-reburning systems that dry lime mud with flue gases.

Pilot-plant experiments showed that steam drying is a useful technique for lime-mud drying. Lime mud with an initial dry content of 69–71% was dried to a dry content higher than 99%. Furthermore, it was shown that the lime mud dried very quickly, within 1–2 s.

Tests in a pilot-scale reactor confirmed that lime mud dried in a steam dryer can be calcined in a fluidized bed. The product lime was "soft burned." Lime mud produced from "soft burned" lime has a higher specific filtration resistance than lime mud produced from a more "hard burned" lime. On the other hand, "soft burned" lime reacts swiftly in the slaking and causticizing reactions.

The results from this study provide a glimpse of the expected quality of the lime produced in the novel system. Further studies in a larger reactor are ongoing.

The novel lime-mud-reburning system has built-in flexibility, since the different process stages are performed in separate devices. With this flexibility, the novel system should find application in a wide variety of chemical-recovery systems.

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