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CONF-77/219-3

THE PREDICTION OF LIMESTONE REQUIREMENTS FOR
SO₂ EMISSION CONTROL IN ATMOSPHERIC
PRESSURE FLUIDIZED-BED COMBUSTION

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Prepared for

5TH International Conference on FBC
Washington, D.C.
December 12-14, 1977

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PRESSURE FLUIDIZED-BED COMBUSTION

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ABSTRACT

The limestone-SO₂ reactivity as determined on a thermogravimetric analyzer was used to estimate the quantity of limestone required to meet the SO₂ emission standard in atmospheric fluidized-bed coal combustors. Comparison of laboratory TGA calcium utilization with pilot plant experimental results were made. Estimates of SO₂ retention in a carbon burnup cell were made for various limestone feed options. From this information, estimates of limestone requirements for an AFBC-CBC system were made for Tymochee dolomite and for Greer and Germany Valley limestones. The results indicate that dolomites may require smaller quantities to meet the SO₂ standard than limestones.

INTRODUCTION

Atmospheric-pressure fluidized-bed combustion (AFBC) is one of the new methods being considered for producing power while meeting EPA SO₂ emission standards with high-sulfur coal. In this process, coal is burned at 850-950°C in a fluidized bed consisting of partially sulfated solid particles (SO₂-sorbent). Limestones (or dolomites) are usually the bed material of choice due to their high calcium content, high reactivity with SO₂ at 850-950°C, low cost, and wide availability.

To reduce the size and cost of an AFBC, a high superficial gas velocity (3-4.6 m/s) is required. However, high gas velocities cause high dust loadings (limestone and unburned coal) in the effluent gas stream. High gas velocities, therefore, produce low combustion efficiencies. Pope, Evans and Robbins (PER)¹ reported combustion efficiencies of ~85% at a fluidizing gas velocity of 3.8 m/s in a PDU combustor. To increase overall combustion efficiency, PER incorporated into their process a carbon burnup cell (CBC) which operates at a higher temperature than the combustor and at a lower fluidizing-gas velocity (~1100°C, 1.8 m/s). Unburned coal dust removed from the combustor effluent stream by cyclones is injected into the CBC. Pope, Evans and Robbins estimates that the overall combustion efficiency can be increased to approximately 99%.¹

It has been estimated by Babcock and Wilcox² that the weight ratio of unburned sulfur to carbon in the coal dust elutriated from the combustor is one-half that in the original coal. Thus, at a combustion efficiency of 85% in the combustor, approximately 93% of the sulfur in the coal is oxidized to SO₂ in the combustor; the other 7% is unburned and leaves the combustor in the coal dust (along with partially sulfated limestone) with the effluent gas stream. Conflicting results³⁻⁵, have been found by investigators as to the fate of the elutriated sulfur which is released as SO₂ in the CBC. The National Coal Board⁴ and Argonne National Laboratory⁵ found that the SO₂ reactivity of several limestones dramatically decreased above 870°C. In contrast, PER³ found that the addition of limestone 1359 to their CBC caused a two-thirds reduction in SO₂ emissions. Due to the high operating temperature (1100°C) in the CBC, the SO₂ might not be captured by either partially sulfated limestone or fresh limestone injected into the CBC. As a result, 7% of the sulfur would bypass the sulfur-removal system of the combustor and be released as SO₂ in the CBC. Consequently, a greater percentage of the sulfur (which is released as SO₂) must be captured in the combustor to meet the EPA SO₂-emission standard (1.2 lb SO₂/million Btu produced by the entire system). Because of this increased requirement for sulfur retention in the combustor, a greater overall Ca/S feed ratio would be required (at higher sulfur retention, calcium utilization is lower). Thus, if it is assumed that no sulfur is retained in the CBC, much larger amounts of limestone would be required to meet the EPA SO₂-emission standards.

Presently available experimental results³⁻⁵ and analyses⁶ of sulfur capture in the CBC are ambiguous. It is the purpose of this paper to estimate the quantity of limestone required for an AFBC-CBC coal combustion plant. Also, four limestone feed configurations to the AFBC-CBC plant are considered.

EXPERIMENTAL

A thermogravimetric analyzer (TGA) was used to study the reaction of various limestones with SO_2 and O_2 . A 0.3% SO_2 -5% O_2 in N_2 synthetic combustion gas was used for all reactions. The limestone- SO_2 reactions were performed at either 900 or 1100°C to represent the operating temperatures for the combustor and CBC, respectively. Before sulfation, fresh limestones were precalcined at 900°C in 20% CO_2 (balance N_2). The TGA system has been described in detail elsewhere.⁷

Three limestones were studied: (1) Tymochtee dolomite, a highly reactive stone, which contains 52% CaCO_3 and 43% MgCO_3 , (2) Greer limestone, which contains 80% CaCO_3 , 3.5% MgCO_3 , 10% SiO_2 , and a high sodium content (Na_2O , ~0.23%), and is highly reactive compared with high-calcium limestones, and (3) Germany Valley limestone containing 98% CaCO_3 and 0.6% MgCO_3 , which is a high-calcium limestone with low reactivity.

RESULTS AND DISCUSSION

Four design options for feeding limestone into the AFBC-CBC power plant were considered (Fig. 5). In option 1, virgin limestone is fed only to the combustor. In option 2, virgin limestone is fed to both the combustor and the CBC. Virgin limestone is fed only to the CBC in option 3; the partially sulfated bed material from the CBC is then fed to the combustor. In option 4, fresh limestone is fed to the combustor and a portion of the partially sulfated limestone from the combustor is fed to the CBC.

Limestone SO_2 Kinetics

The limestone- SO_2 kinetic results and their conversion to a " SO_2 retention vs. Ca/S ratio" plot are presented in detail for Greer limestone. Similar information was obtained for Germany Valley and Tymochtee.

In order to determine the limestone requirements for the four feed options, the reactivities of fresh and partially sulfated limestones with SO_2 were determined, using a TGA (as described in the previous section). Results for Greer limestone are shown in Fig. 1. Virgin limestones were reacted with SO_2 and O_2 at both 900 and 1100°C to determine the reactivity of fresh limestone at the conditions prevailing in a combustor and CBC (curves 2 and 3, respectively). These experiments provided the information necessary to determine limestone requirements for options 1 and 2 and parts of options 3 and 4. Limestones which had been partially sulfated (5% conversion of CaO to CaSO_4 and heat treatment for 30 min at 1100°C) were reacted with SO_2 at 900°C to determine the combustor limestone requirements for option 3 (curve 4). A total heat-treating time of 30 min was chosen to simulate the estimated limestone CBC residence time. For option 4, fresh limestones were partially sulfated (reacted for 30 min with SO_2) at 900°C. This material was then tested for SO_2 reactivity at 1100°C (curve 5).

The initial reactivity of Greer limestone at 900°C is higher than at 1100°C, as expected; however, the extent of conversion of CaO to CaSO_4 after three hours is the same. Partial sulfation (5% conversion) of Greer limestone at 1100°C, had a detrimental effect on its reactivity at 900°C (discussed later). Partially sulfated (at 900°C) Greer limestone at 1100°C had the highest reactivity (conversion of calcium oxide to CaSO_4 at 1100°C was 48%). These results show that Greer limestone does react with SO_2 at 900 and 1100°C. However, this information is not in a useful form to predict limestone requirements for AFBC-CBC plants. This information can be converted to the proper form, which is a plot of internal SO_2 reduction (ratio of absorbed SO_2 to that released in the reactor) vs. internal Ca/S ratio (S is the number of moles of sulfur released as SO_2 in the reactor), by using a fluid-bed desulfurization equation developed by Westinghouse.

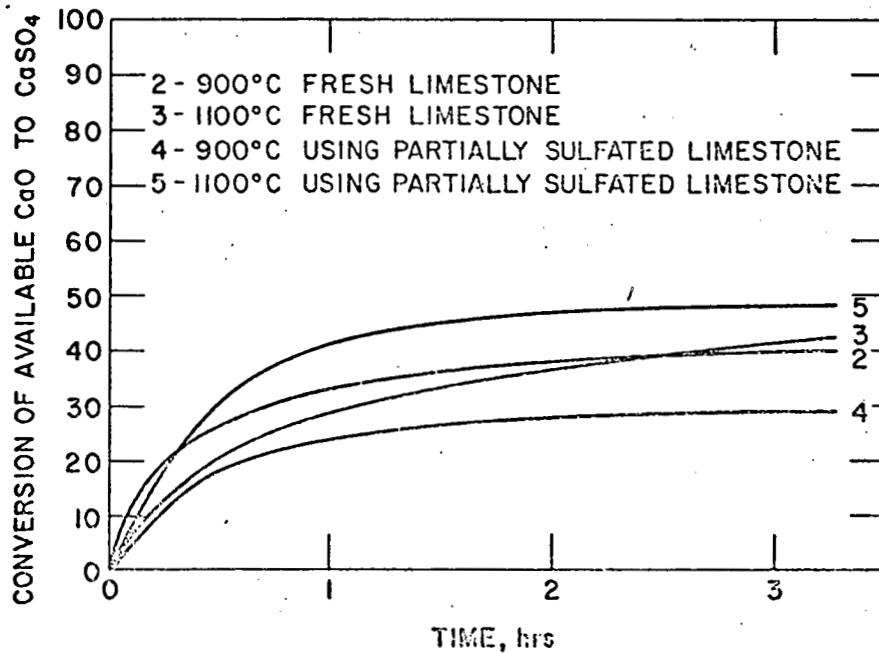


Figure 1. Conversion of CaO to CaSO₄ in fresh or partially sulfated Greer limestone at 900 and 1100°C.

$$U = \frac{1}{Ca/S} \left[1 - \frac{V}{kH\epsilon} \left(1 - e^{-\frac{kH\epsilon}{V}} \right) \right] \quad (1)$$

where U = calcium utilization, fraction
 Ca/S = calcium to sulfur mole ratio
 V = superficial gas velocity, m/s
 H = fluidized-bed height, m
 ε = bed voidage, assumed to be 0.5
 k = average particle reaction rate constant, s⁻¹.

This fluid-bed desulfurization equation gives the calcium utilization as a function of the "average" reaction rate constant of the particles in the bed (provided the superficial gas velocity and bed height are known). Thus, in order to determine U, the "average" rate constant, k, must be known and is obtained using the kinetic information shown in Fig. 1. (See references 7 and 8 for calculation details.)

The results of converting the kinetic data to SO₂ reduction vs. Ca/S ratio for Greer limestone are shown in Fig. 2. (PER experimental results are also included.) The internal SO₂ reduction should be distinguished from SO₂ reduction normally reported by FBC workers, which is the percentage of the total sulfur that does not leave the combustor as SO₂. That is, it includes both sulfur captured by the limestone and unburned sulfur and is not useful for the process analysis presented below.

Curve 1 in Fig. 2 represents the corrected (internal) experimental results obtained by PER⁹ using Greer limestone in the combustor, assuming that 7% of the total sulfur remains in the unburned coal dust.

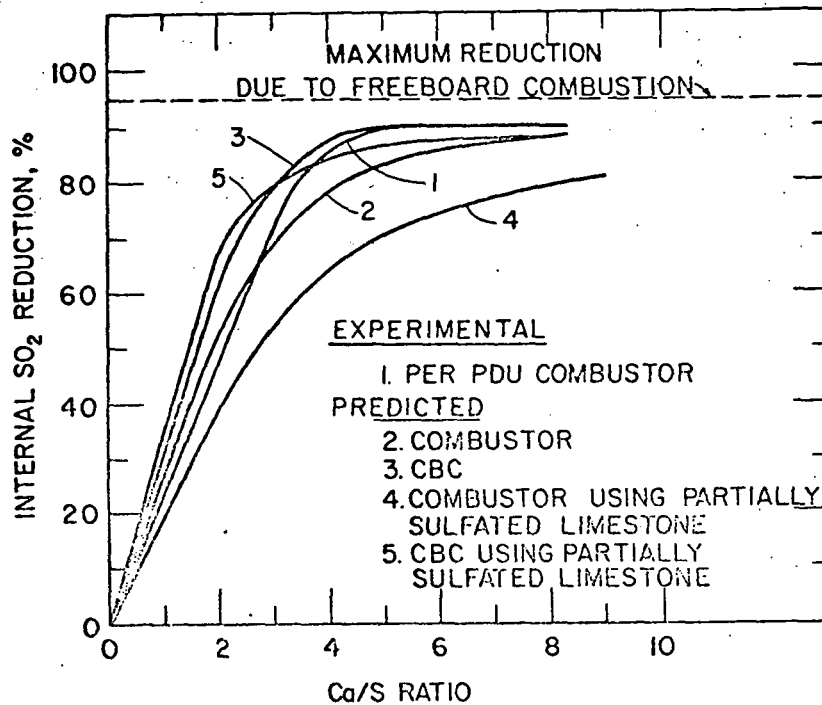


Figure 2. Predicted SO₂ Retention vs. Ca/S Ratio for Greer Limestone

Curve 2 is the predicted results for Greer, in the combustor, based on the laboratory (TGA) experimental data. The laboratory experimental results predict a somewhat higher Ca/S requirement (Curve 2) for a given SO₂ reduction than has been found in PER's pilot plant (Curve 1). Since the overall Ca/S ratio for the AFBC-CBC plant is sensitive only to the calcium requirements of the combustor, PER's experimental results (Curve 1) were used instead of the ANL TGA data (Curve 2) to provide more realistic estimates of overall limestone requirements. TGA data was used to estimate CBC limestone requirements since pilot plant data is unavailable. The resulting comparisons are qualitatively correct; quantitative comparison can be made as sufficient FBC data becomes available.

Curve 3 shows a higher SO₂ reduction for a given Ca/S ratio for fresh limestone in the CBC than in the combustor even though the SO₂ reactivity is lower in the CBC (Fig. 1). This is because the fluidizing velocity is lower in the CBC, producing a longer SO₂ residence time in the fluid bed and allowing more SO₂ to be captured by the limestone.

Curve 4 (TGA data was used since no experimental results exist for this option) projects a lower SO₂ retention in the combustor by Greer limestone that has been partially sulfated in the CBC. This is due to its low SO₂ reactivity (Fig. 1, Curve 4). It has been shown¹⁰ that limestones which contain small quantities of CaSO₄ sinter rapidly at high temperatures (1100°). Sintering causes small pores to coalesce into large pores, and lowering of total porosity results.¹⁰ This sintered limestone now has a low SO₂ reactivity compared with fresh limestone at combustor conditions (900°C). The reactivity loss by the sintered limestone is, however, experienced only at the lower temperature (900°). Although sintering does occur at higher temperatures, it is speculated that mobility of CaSO₄ within the limestone particle at high temperature allows reaction of fresh calcium with SO₂ to continue and thus the limestone retains its reactivity at the higher temperature (see Curve 3, Figs. 1 and 2). However, removing the limestone from the CBC (~1100°C) and injecting it into the

combustor at the lower temperature ($\sim 900^{\circ}\text{C}$) causes a loss of CaSO_4 mobility in the partially sulfated and sintered limestone. With the CaO less available (reduced porosity and CaSO_4 mobility), the limestone is less reactive.

Curve 5 (Figs. 1 and 2) shows the highest limestone- SO_2 retention for a given CaO/S ratio. Curve 5 represents the reactivity of partially sulfated (900°C) Greer in the CBC for option 4. Calcination at the lower temperature (900°C) produces a pore structure which is somewhat more favorable to the SO_2 diffusion-reaction process. It is believed that removing the partially sulfated limestone from the combustor and injecting it into the CBC causes the CaSO_4 to become more mobile. This exposes fresh calcium for reaction with SO_2 .

The " SO_2 retention vs. Ca/S " results for Germany Valley limestone and Tymochee dolomite are shown in Figs. 3 and 4. Notice that for Germany Valley, higher Ca/S ratios are required to obtain the same SO_2 retention as for Greer limestone, while for Tymochee dolomite smaller Ca/S ratios are required. This is simply due to high Tymochee- SO_2 reactivity and very low Germany Valley- SO_2 reactivity.

Evaluation of AFBC Limestone-Feed Options

Greer Limestone. The SO_2 reduction results in Fig. 2 were used to determine the overall Greer limestone requirements for the four process options. The requirements for Greer limestone are given in Fig. 5. The required SO_2 removal in these calculations was based on using Sewickley coal, which is a Pittsburgh seam coal (4.3% S) having a heating value of 12,200 Btu/lb. This coal requires an overall retention of 83% of the SO_2 to meet EPA SO_2 emission standards.

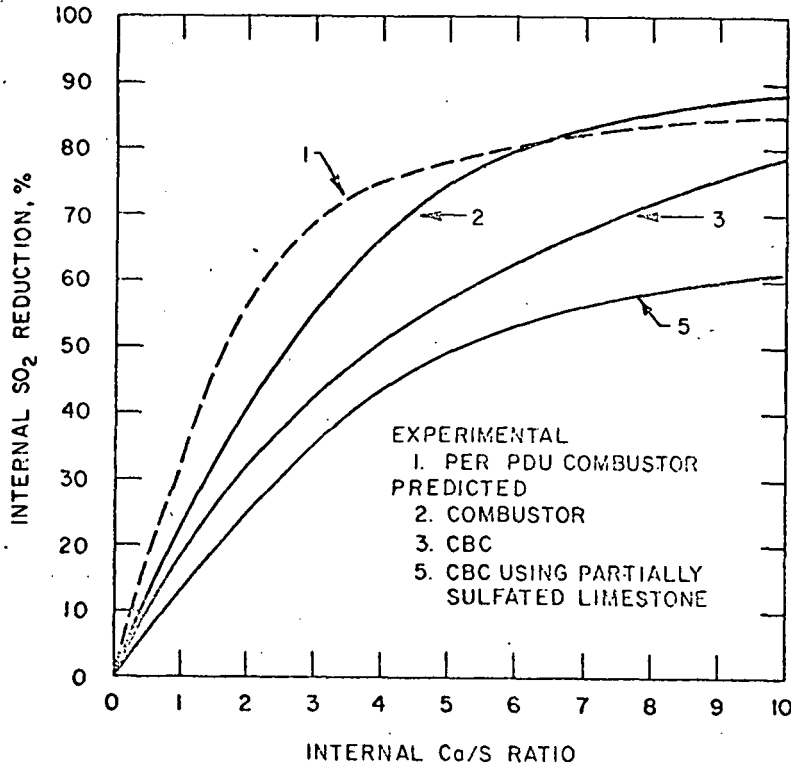


Figure 3. Predicted SO_2 Retention vs. Ca/S ratio for Germany Valley Limestone

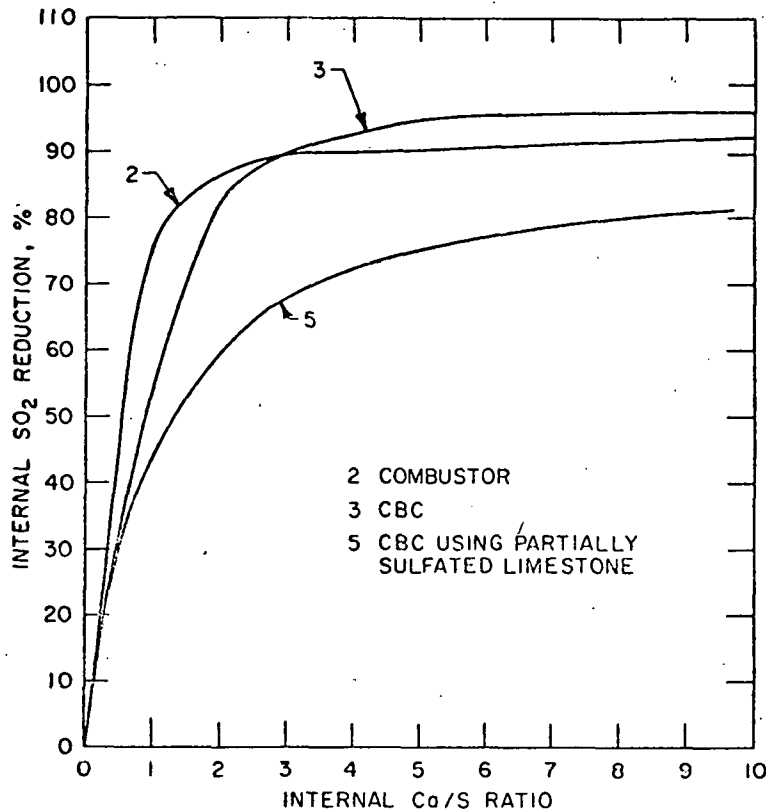


Figure 4. Predicted SO₂ Retention vs. Ca/S ratio for Tymochtee Dolomite

It was assumed in option 1 that no SO₂ was retained by limestone (fresh, elutriated, or otherwise) in the CBC. This assumption was used as a base case since it has been a prevailing assumption in FBC research. Since 7% of the total sulfur is assumed to be released from the CBC as SO₂,^{2,3} 89.3% SO₂ retention would be required in the combustor to give an overall SO₂ retention of 83%. This would require that the Ca/S ratio in the combustor be 4.8 (Curve 1, Fig. 2) and that the overall Ca/S ratio be 4.5 (Fig. 5).

The limestone requirements can be decreased by feeding fresh limestone to the CBC to remove SO₂ released in the CBC (option 2). A Ca/S ratio of 3.4 is required to achieve 83% SO₂ retention in the CBC (Fig. 2, Curve 3). A Ca/S ratio of 3.8 (for 83% SO₂ retention) is required in the combustor (Fig. 2, Curve 1). The overall Ca/S ratio is then 3.8 which is approximately 85% that for option 1.

Option 3 was considered in the hope that the high temperature in the CBC would have a beneficial heat treatment effect and that the CBC would have two roles-- capture of SO₂ released in the CBC and pretreatment of limestone for limestone-SO₂ reactivity enhancement. As shown in Figs. 1 and 2, reactivity enhancement was not realized due to the detrimental effect of the CaSO₄ and high temperature (discussed above). Since in option 3, virgin limestone is fed to the CBC, the Ca/S ratio is 128 and a 95% SO₂ retention would be obtained in the CBC. This would require an internal SO₂ retention of 82% in the combustor. Since stones that have been partially sulfated at high temperature (~1100°C in the CBC) are less reactive at the lower temperature (Fig. 2, Curve 4), a high internal Ca/S ratio (9.5) would be required in the combustor. The overall Ca/S feed ratio for option 3 is 8.9.

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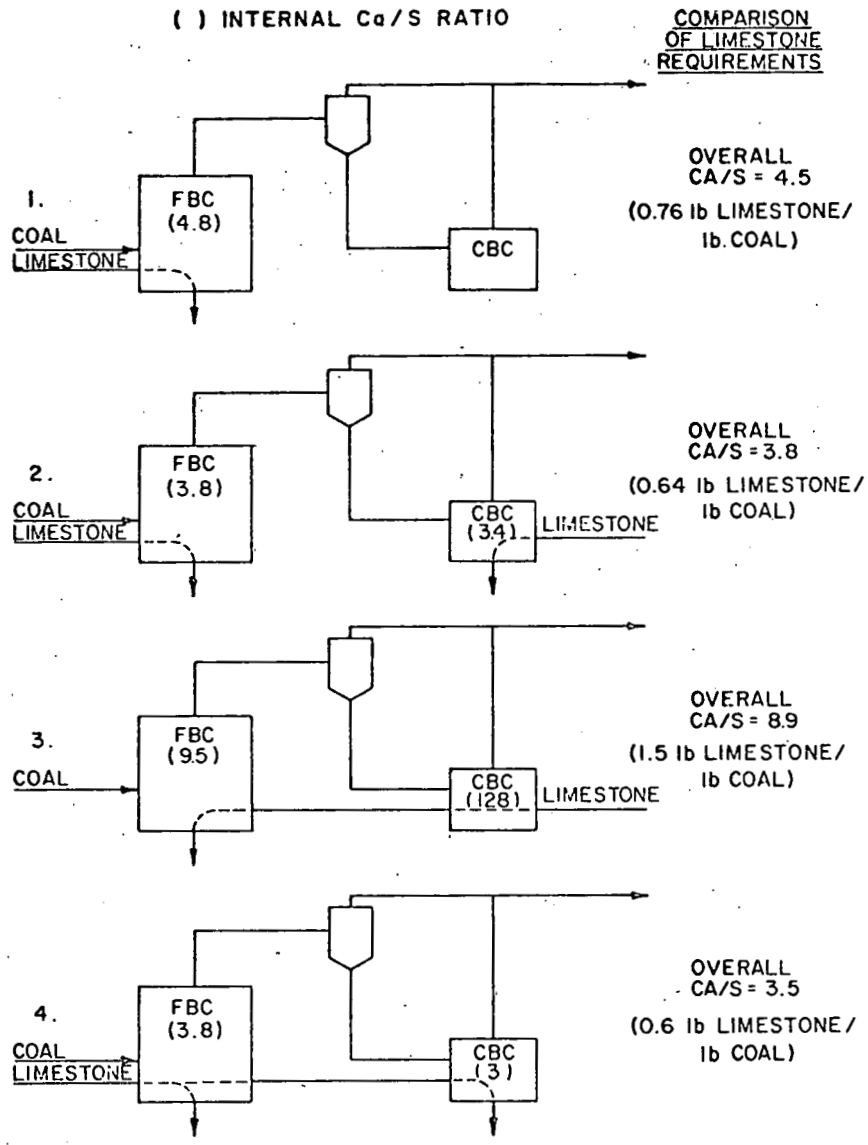


Figure 5. Greer Limestone Requirements for Four Limestone Feed Schemes

Option 4 provides a partially sulfated limestone feed stream from the combustor to the CBC. A CaO/S ratio of only 3 is required to obtain 83% SO₂ retention in the CBC, necessitating a Ca/S of 3.8 in the combustor. This option gives the lowest overall Ca/S feed ratio--3.5.

The limestone requirements presented above are based on Fig. 2. The projected limestone requirements are fairly insensitive to the SO₂-limestone reactivity in the CBC (Curves 3 and 5) since only 7% of the total sulfur is released as SO₂ in the CBC. Thus, if these SO₂ retention curves are over-optimistic by a factor of 2, the error in the overall estimated Ca/S ratio would be low by only 4%. The overall Ca/S

ratio is sensitive to the limestone-SO₂ reactivity in the combustor (Curve 2), which is why fresh limestone-SO₂ reactivity determined by PER was used as a base for comparing the various process options.

The results in Fig. 5 are for idealized limestone feed options, which were chosen to elucidate the role of the CBC in minimizing limestone requirements. In actuality, unreacted and partially sulfated limestone will be elutriated with the unburned coal, removed from the gas by the cyclones, and injected into the CBC. Most likely, the elutriated limestone will provide a CaO/S ratio in the CBC in excess of that shown in option 4 (CaO/S of 3), which is needed to obtain 83% SO₂ reduction. If this elutriated limestone has no SO₂ reactivity, a higher Ca/S ratio may be needed in order to meet EPA SO₂ standards, requiring feeding of larger amounts of Greer limestone to the combustor. However, as shown in Fig. 1, this is not the case; the elutriated limestone will have sufficient reactivity that excessive limestone quantities will not be required. If pilot plant testing shows these projections to be low (due to high elutriation rates in the CBC or other reasons), option 4 suggests that feeding of combustor bed material (larger particles) or fresh limestone (option 2) to the CBC would be beneficial.

Finally these estimated Ca/S ratios are only for a 4.3% sulfur coal. Obviously, a 3% sulfur coal would require less SO₂ retention and thus lower Ca/S ratios.

Germany Valley Limestone. The overall Ca/S ratio was determined for the various limestone feed options, using Germany Valley limestone and Sewickley coal. Option 3 was not considered due to the high Ca/S ratio requirement for this option with Greer limestone. For option 1 assuming that no sulfur capture occurs in the CBC (see Fig. 6), the SO₂ emission standard cannot be met, regardless of the amount of limestone fed to the combustor. This is due to the low reactivity of Germany Valley limestone with SO₂ and the high SO₂ reduction 89.3% needed in the combustor to meet the 83% SO₂ reduction requirements of an AFBC-CBC plant.

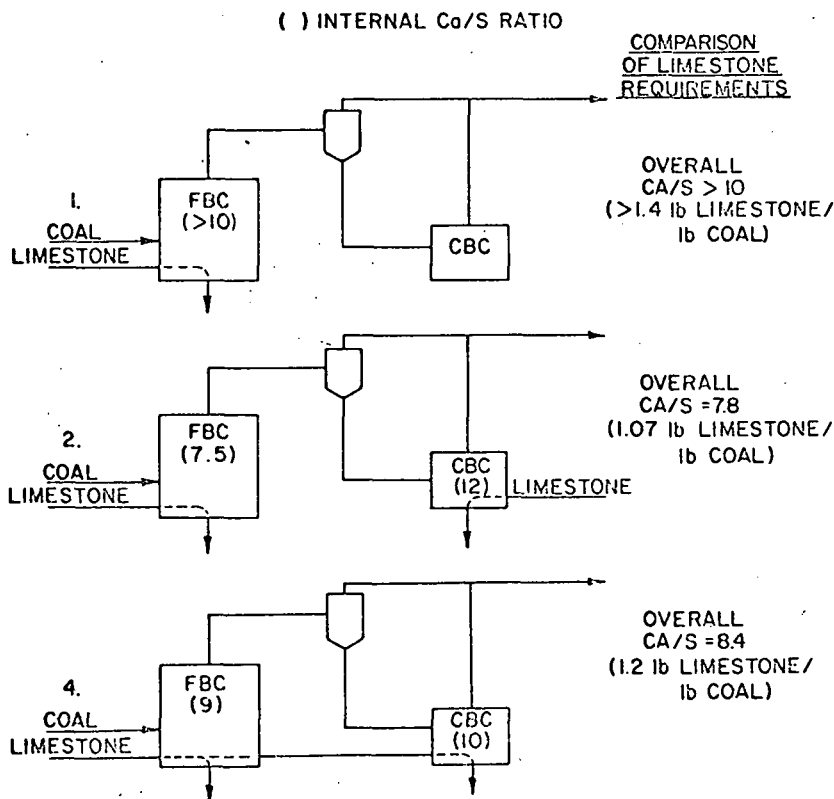


Figure 6. Germany Valley Limestone Requirements for FBC

Feeding fresh limestone to the CBC (option 2) results in 83% SO₂ reduction in the CBC. A Ca/S ratio of 7.5 in the combustor will produce a 83% SO₂ reduction, meeting emission standards. The overall limestone requirements is approximately 1.1 lb limestone per pound of coal (Ca/S = 7.8).

If a portion of the combustor bed material is fed to the CBC (option 4), approximately 60% of the SO₂ released in the CBC can be captured. Thus, 84.6% of the SO₂ in the combustor must be captured, requiring a Ca/S of 9. This feed option requires feeding of more limestone than coal on a weight basis (1.2 lb limestone/lb coal). Regardless of the feed option considered, the high calcium limestone, Germany Valley, is a poor choice for AFBC-CBC plants. It would require more limestone than coal.

Tymochtee Dolomite. Tymochtee reactivity with SO₂ is much higher than Germany Valley reactivity, and much lower Ca/S ratios are required to obtain 83% SO₂ reduction. Since there is no PER data for Tymochtee dolomite, all calculations are based on TGA kinetic results; sewickley coal was assumed for all the calculation of dolomite requirements for various feed options. In the absence of SO₂ capture in the CBC (Fig. 7, option 1), a Ca/S ratio of only 2.3 (0.6 lb limestone/lb coal) is needed in the combustor to obtain 90% SO₂ reduction, which is necessary to meet plant emission standards. Feeding fresh limestone to the CBC (option 2) reduces the Ca/S ratio to 1.5 (0.4 lb limestone/lb coal).

Feeding of "spent" combustor bed material to the CBC (option 4) can reduce the Ca/S ratio slightly; however, this requires a high Ca/S ratio of 10 in the CBC since the Tymochtee dolomite leaving the combustor has low reactivity. Achieving this high Ca/S ratio requires that 90% of the combustor bed material be fed to the CBC.

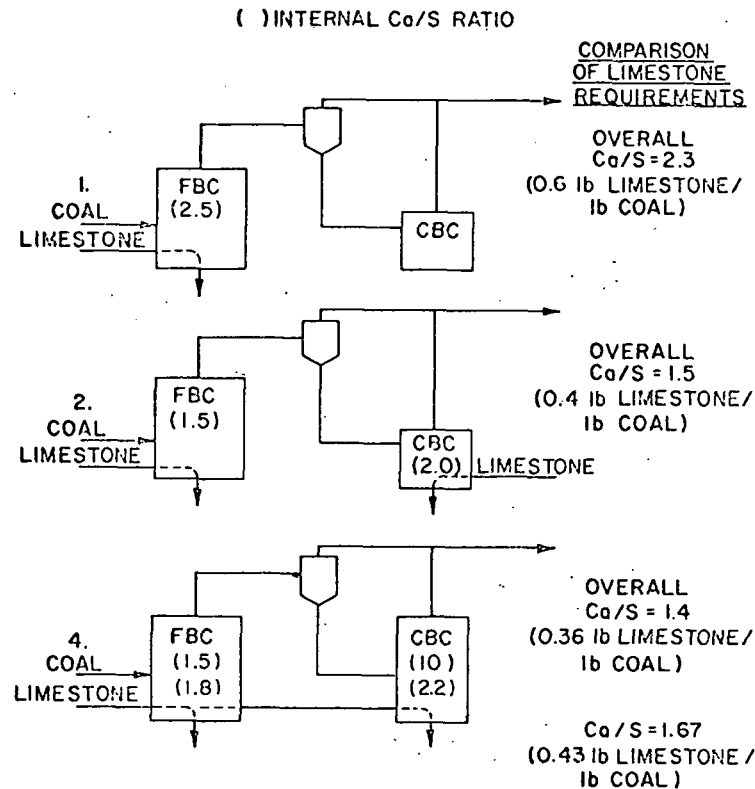


Figure 7. Tymochtee Dolomite Requirements for FBC

If feeding of combustor bed material to the CBC is not considered, 10-20% of the bed material could be elutriated and enter the CBC. It is estimated that this would produce a CaO/S ratio of 2.2 in the CBC, resulting in 62% SO₂ retention. Then, 84.6% SO₂ reduction would be required in the combustor, requiring that Ca/S ratio be 1.8. An overall Ca/S ratio of 1.67, or 0.43 lb of Tymochtee per lb of coal, would be required.

Comparison of Greer, Germany Valley, Tymochtee. Greer, Germany Valley, and Tymochtee are compared in Fig. 8 for option 4 since introduction of elutriated limestone into the CBC is the only feed system presently in operation.¹¹ Tymochtee dolomite would require only 0.36 or 0.43 lb of limestone/lb of coal, depending upon how much spent combustor bed material is fed to the CBC. Greer limestone would require 50% more material and three times as much Germany Valley would be required as Tymochtee. For a 635-MW plant, a 70% capacity factor, and a limestone cost of \$8/ton, the use of Tymochtee dolomite would result in a \$2-3 million/yr savings in comparison to Greer limestone and a \$9-10 million/yr savings in comparison to Germany Valley. These are savings in the cost of the limestone only and do not include savings due to smaller size of equipment.

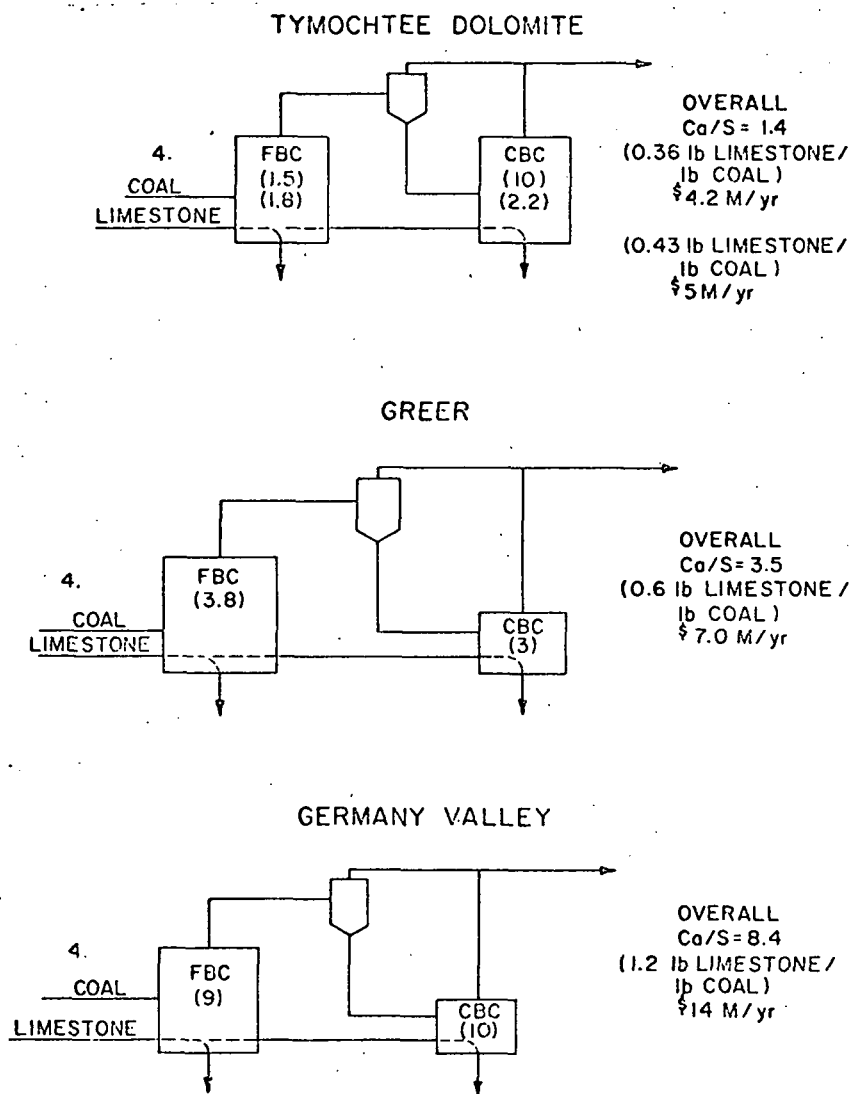


Figure 8. Comparison of Limestone Requirements for 635-MW AFBC, 70% Cap., \$8/Ton Limestone

The estimates are based on laboratory experiments which indicate that the use of dolomites instead of limestone may produce a significant cost and environmental savings. It should be cautioned that estimated limestone requirements were based on laboratory TGA data and on only three calcium-based stones. Nevertheless, the testing of limestone dolomites in AFBC-CBC pilot plants should be performed to determine actual limestone and dolomite requirements.

CONCLUSIONS

The quantities of limestone required for AFBC-CBC plants to meet SO₂ emission standards have been estimated. Four different limestone feed options were considered. It was found that virgin limestones and partially sulfated (in the combustor) limestones do react with SO₂ at the conditions being considered for operation of the CBC. Feeding virgin limestone or limestone that has been partially sulfated in the combustor to the CBC minimizes limestone requirements. The results indicate that highly reactive dolomite requires less limestone (on a weight basis) than does limestone to meet SO₂ emission standards for an AFBC-CBC plant.

It should be cautioned, that these estimates are made from experimental laboratory data and need confirmation in pilot plant operations. Also these data are valid only for fluidized-bed combustor which incorporates a carbon burnup cell and are not valid for a fluidized-bed system that recycles the elutriated bed material back to the main combustor.

ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. Energy Research and Development Administration. We thank A. A. Jonke, D. Webster, and L. Burris for their support and guidance. We also thank John Stockbar for his laboratory assistance and Ken Jensen for analytical assistance. Finally we would like to thank J. Simmons, the technical editor.

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