



Project Summary

Boiler Simulator Studies on Sorbent Utilization for SO₂ Control

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M. P. Heap

The objective of this program was to provide process design information for sorbent utilization as applied to the LIMB process. Specifically, the program was designed to investigate the role of boiler thermal history, sorbent injection location, calcium to sulfur molar ratio, and SO₂ partial pressure on capture effectiveness with limestones, dolomites, and slaked limes with and without metallic promoters. The experimental studies were supported by theoretical calculations using grain and pore models that considered both the heterogeneous chemical reaction and the relevant diffusional processes.

The experimental results and the sulfation model calculations indicate that the sorbent injection locations and the residence time within the sulfation temperature window can significantly influence overall sulfur capture for any particular sorbent. Unless the sorbent is promoted with a metal additive, downstream injection at about 2250 °F* results in optimum sorbent utilization. Increasing the gas-phase SO₂ concentration improves sorbent utilization, but the dependence is non-linear due to the combined effects of intrinsic chemistry and diffusion.

In general dolomitic sorbents perform better than calcitic sorbents and hydroxides are superior to carbonates. The true influence of pressure slaking is unclear; however, the best sorbents tested were the pressure slaked dolomites. The performance of all sorbents can be enhanced by adding appropriate metallic compounds in relatively small quantities.

This Project Summary was developed by EPA's Air and Energy Engineering Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

This report describes experimental and analytical work, the objective of which was to provide process design information for sorbent utilization as applied to the EPA's Limestone Injection Multistage Burner (LIMB) process. Specifically, the program was designed to investigate the role of boiler thermal history, sorbent injection location, calcium to sulfur molar ratio, and SO₂ partial pressure on capture effectiveness with limestones, dolomites, and slaked limes with and without metallic promoters. The experimental studies were supported by theoretical calculations using grain and pore models that considered both the heterogeneous chemical reaction and the relevant diffusional processes.

All of the data presented in this report were obtained with the 10⁶ Btu/hr* Boiler Simulator Furnace (BSF). The facility consisted of a refractory-lined vertical tower section 18 ft* tall with an ID of 1.8 ft, and a horizontal air-cooled convective section. The tower section consisted of eight modular units, each with multiple access ports for sampling, injection, or cooling. The BSF was used to simulate a wide range of time/temperature profiles by positioning water-

*To convert to the metric equivalent, use
 $T_K = (T_{°F} + 459.67)/1.8$.

*To convert to the metric equivalents, use
Btu/hr = 0.293 W, and ft = 0.305 m.

cooled panels or rods appropriately in the furnace. A low- NO_x distributed mixing burner was positioned at the top of the tower to down-fire either coal or natural gas.

The first portion of this study focused on the influence of the boiler design/sorbent injection parameters on overall SO_2 capture. These studies included consideration of overall excess air, burner stoichiometry, radiant zone heat removal rate, burner swirl, general sorbent injection location (burner versus downstream), importance of sorbent premixing with fuel, and impact of low NO_x operation. The results of these studies indicated that the parameters which were most critical for the optimization of sorbent utilization were sorbent injection location and the time/temperature history between injection and 1700 °F.

Combustion Parameters

Figure 1 summarizes results obtained on the impact of sorbent injection location with a typical limestone and slaked lime. These data indicate that the SO_2 capture increased approximately linearly with increasing calcium to sulfur ratio and higher capture was achieved with downstream injection at about 2250 °F for both sorbents. Figure 1 indicates that the slaked lime was more sensitive to injection location than the limestone, and that is typical of results with other calcium hydroxide materials. The desirability of downstream injection (compared to sorbent premixed with the fuel) shown in Figure 2 is typical of the trends obtained with a wide variety of other sorbents with both gas- and coal-firing under both favorable and highly quenched thermal conditions. Injection downstream of the burner greatly enhances the surface area available for subsequent sulfation due to increased surface area (reduced grain growth). However, if the injection is delayed beyond the beginning of the reaction window (about 2250 °F), the effective residence time in the sulfation zone decreases rapidly. In addition, a larger portion of the available residence time must be used for in situ calcination of the stone, and lower temperatures produce reduced diffusion and chemical rates. Therefore, the overall optimum injection temperature appears to be near the front of the sulfation window (about 2250 °F). Injection above this temperature results in decreased sorbent reactivity due to excessive grain growth; injection significantly below 2250 °F produces an even higher initial surface area, but this effect is more than

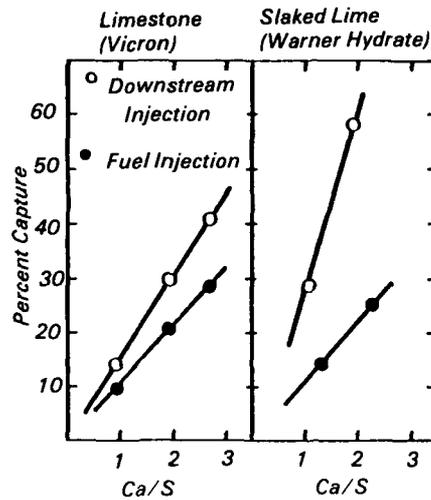


Figure 1. Influence of injection location on sulfur capture.

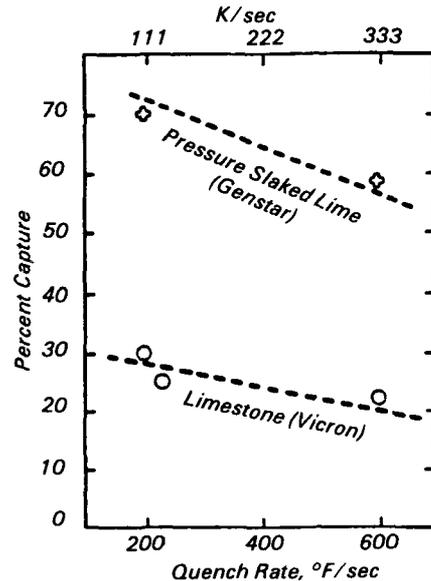


Figure 2. The effect of quench rate in sulfate zone.

compensated for by the decreased chemical reaction, product layer diffusion rates, and available residence time.

Detailed heat transfer calculations have indicated that the average quench rates within the sulfation zone (1700 to 2250 °F) of commercial boilers are in the range of 500-700 °F/sec, and in typical utility boilers the effective time available for sulfation may be as little as 1 sec. Figure 2 summarizes the influence of quench rate on SO_2 capture with downstream sorbent injection at a Ca/S ratio of 2.0. Two very different sorbents were used in these studies: Vicron

(representing a typical low surface area calcium carbonate) and Genstar pressure-slaked lime (typical of high surface area dolomitic hydroxides). The dashed lines represent capture predictions generated by EER's grain model. No model parameters were adjusted to improve the agreement between the experimental results and the model predictions. Overall, the agreement between the predictions and the data is excellent and, as expected, capture decreased with increasing quench rate because reaction time decreased. These results indicate that a particular sorbent injected under identical conditions in two separate boilers may produce different capture efficiencies because of variations in the thermal characteristics of the boilers. The influence of fuel sulfur concentration was studied by varying the amount of H_2S doped into the natural gas flames. SO_2 concentrations of 500, 1000, 1800, and 3000 ppm (0% O_2 , dry) were tested with Colton hydrated lime and Genstar pressure-slaked dolomitic lime (type S). The Ca/S ratio was held constant at 2.0, and the sorbent was injected at 2250 °F for all runs. These results are shown in Figure 3 along with model predictions. Both the data and the model showed an increase in sulfur capture with increasing SO_2 concentration. The curvature in the theoretical predictions, which is in agreement with the experimental results, is due to a coupling between the chemistry and the pore and product layer diffusion processes.

Sorbent Composition

Sorbent composition and other physical properties are probably the most important factors in determining overall capture performance. With all other variables held constant, a wide range of sulfur capture can be produced with different sorbent types. Capture with downstream sorbent injection ranged from 30 percent for Vicron to over 75 percent for pressure-slaked dolomitic sorbents at a Ca/S ratio of 2.0. The range in capture with high temperature injection (with the fuel) was not as broad (20 to 40 percent at Ca/S = 2.0), because the final surface area depends less on the initial sorbent characteristics when calcination occurs at flame zone temperatures. In general the capture results tend to be grouped primarily by general sorbent type. On a calcium molar basis, the three pressure-slaked dolomitic sorbents gave the best capture with downstream injection, although other studies with MgCO_3 indicate that the high capture with dolomitic sorbents was not due

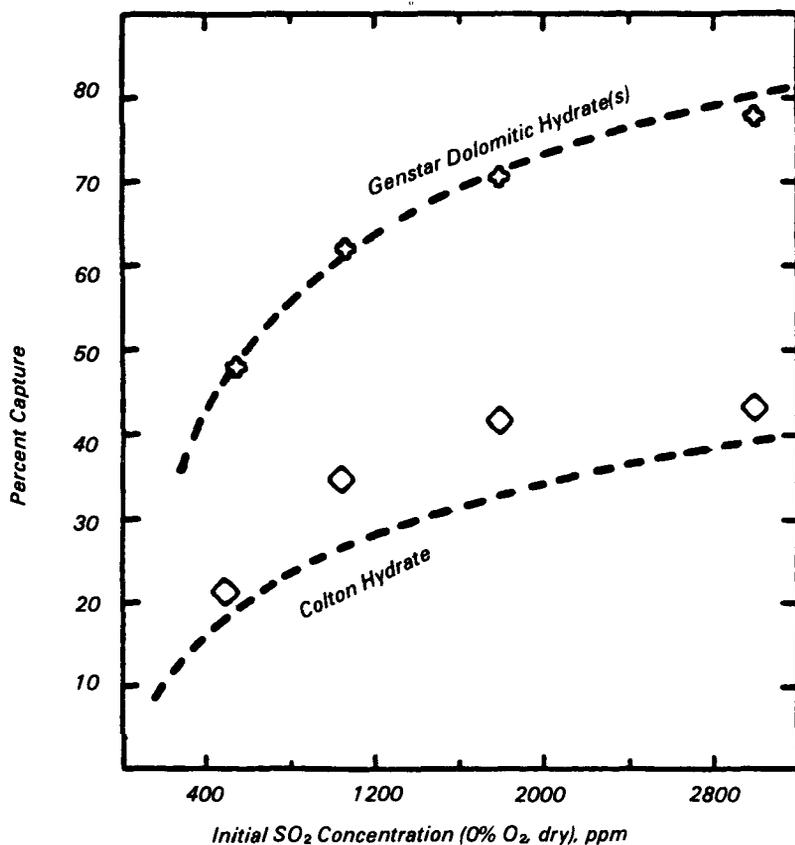


Figure 3. The effect of initial SO₂ concentrations.

to sulfur capture by magnesium. They were followed by the natural dolomite, the two limes slaked at atmospheric pressure, and finally the Vicron limestone. For injection with the fuel, both the natural and the pressure-slaked dolomites gave the highest capture, while the calcitic carbonate (Vicron) and the calcitic hydroxide gave the lowest capture.

Since the sorbents studied include both calcitic and dolomitic materials, the total mass feed rate can vary substantially for a given Ca/S molar ratio: hence, both the initial sorbent cost and the amount of ash which must ultimately be disposed of are variable. In order to compare the different sorbents on a total mass output basis, the results are presented as a function of the mass parameter, MgO + CaO divided by the total inherent coal ash mass (assuming a 1 percent sulfur, 10 percent ash coal), as shown in Figure 4. Compiling the data on this basis indicates the capture that can be achieved relative to the amount of additional material that must be removed from the particulate collection devices. Even on

this basis the dolomitic pressure-slaked limes appear extremely attractive as does the Warner hydrated lime. Again the poorest performance was achieved with the Vicron limestone. These results suggest that capture in excess of 60 percent can be achieved with about a 50 percent increase in the dry ash handling requirements (i.e., sorbent addition is like switching from a 10 to a 15 percent ash coal with 1 percent sulfur).

Enhancement by Promoter Addition

The final primary area of study was the addition of various promoters to enhance sulfur capture. Initially, Cr₂O₃ was found to dramatically improve capture with Vicron, especially when the promoted sorbent was injected into the high temperature region at the burner. Many of the transition metal promoters were evaluated for possible capture enhancement; however, only molybdenum and chromium enhanced capture significantly relative to Vicron. Subsequently, it was found that alkali

metal components (lithium, sodium, potassium) gave positive results similar to those found with the chromium series materials. The most unusual thing about the chromium promoted limestone sorbent was that, in contrast to all previous results, the capture with high temperature injection was equivalent to that with downstream, low temperature injection. Chromium appears to have the ability to negate the effect of thermal sintering of the sorbent. Figure 5 shows the influence of Cr₂O₃ addition with three types of sorbents. The open bars represent the capture measured with the sorbents alone and the shaded bars indicate the increase in capture that resulted from five percent chromium addition. With all of the sorbents and with both burner zone and downstream sorbent injection, the capture increases with chromium promotion were significant. Even the performance of the Genstar pressure-slaked dolomite was improved: 70 to 85 percent capture for the downstream sorbent injection and 35 to 70 percent capture for injection with the fuel. In general the enhancement above the base line was greater when the promoted sorbents were injected into the high temperature region, although the absolute capture levels were generally higher for downstream injection.

The exact mechanism for the chromium and sodium enhancement is not clear; however, it appears likely that these materials promote capture by enhancing the product layer diffusion step since the model calculations indicate that product layer diffusion is the primary limitation to increased sulfation rates. Additional work is needed to optimize the method of promoter addition and clarify the controlling mechanisms.

Conclusions

The experimental results and the sulfation model calculations indicate that the sorbent injection locations and the residence time within the sulfation temperature window can significantly influence the overall sulfur capture for any sorbent. Unless the sorbent is promoted with a metal additive, downstream injection at about 2250 °F results in optimum sorbent utilization. Increasing the heat removal rate between about 1700 and 2250 °F results in decreased sulfur capture. Increasing the gas-phase SO₂ concentration (e.g., due to increased coal sulfur content) improves sorbent utilization, but the dependence is nonlinear due to the combined effects of intrinsic chemistry and diffusion.

In general dolomitic sorbents perform better than calcitic sorbents, and hydrox-

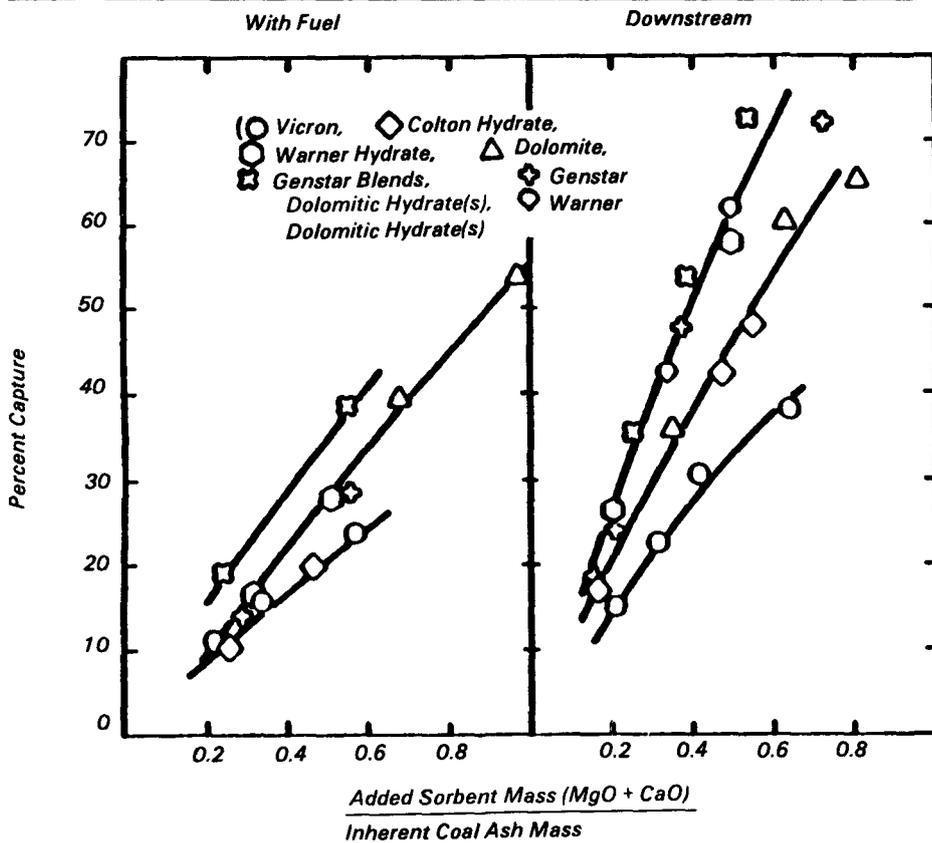


Figure 4. Capture comparison—mass basis.

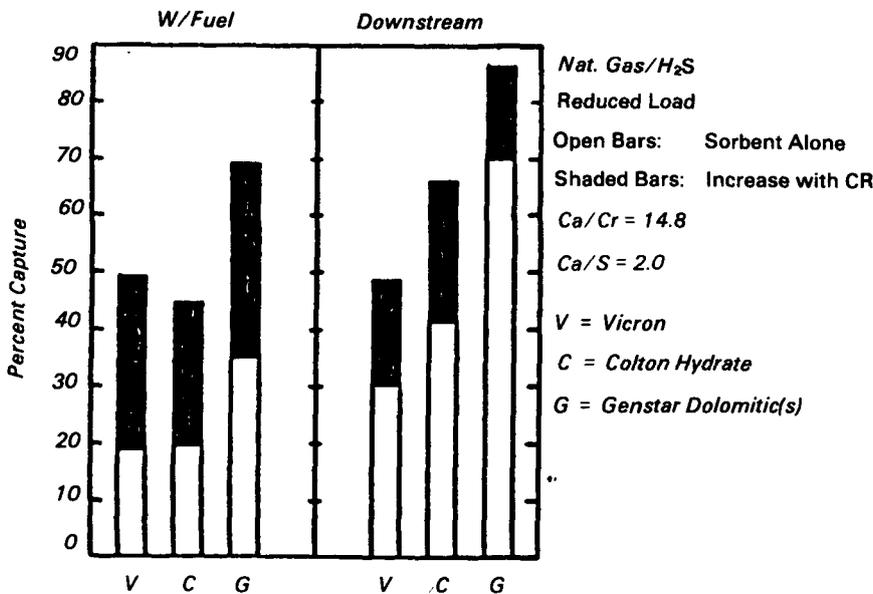


Figure 5. Capture enhancement with Cr_2O_3 .

ides are superior to carbonates. The true influence of pressure slaking is unclear; however, the best sorbents tested (on either a calcium molar or total mass basis) were pressure-slaked dolomites. The magnesium in the dolomite materials does not react to produce magnesium sulfate; it probably enhances product layer diffusion. The performance of all sorbents can be enhanced by adding appropriate metallic compounds in relatively small quantities.

Thus, the results of this study suggest that it is possible to achieve relatively high capture levels by at least two alternative methods: use of advanced sorbents (e.g. pressure-slaked dolomites) or promoted limestones. Clearly these two concepts can be combined to produce even higher capture levels.

In general the results of this study show that it is possible to exceed the current EPA performance goal of 50 percent at a Ca/S ratio of 2.0 with either an inexpensive limestone promoted with a material as simple as sodium carbonate or a pressure-slaked dolomite if the sorbent is injected downstream of the main heat release zone.

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The complete report, entitled "Boiler Simulator Studies on Sorbent Utilization for SO₂ Control," (Order No. PB 87-101 770/AS; Cost: \$16.95, subject to change) will be available only from:

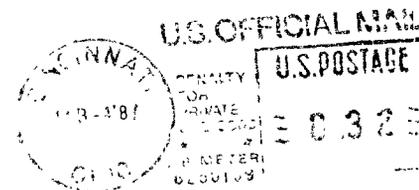
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Project Summary

Analysis of Utility Control Strategies Using the LIMB Technology

T. E. Emmel and B. A. Laseke

The report gives results of a study to evaluate the impact of proposed acid rain legislation on the potential application of limestone injection multistage burner (LIMB) technology to achieve sulfur dioxide (SO₂) and nitrogen oxide (NO_x) reductions at coal-fired utility power plants.

The study found that proposed acid rain legislation, which mandates the retrofit of high efficiency control technologies such as flue gas desulfurization (FGD) or which requires national SO₂/NO_x reduction levels greater than 10 million tons per year, would significantly reduce the application of LIMB. For regulatory strategies which do not mandate the use of FGD and which require emission reductions of 8 to 10 million tons per year, the potential LIMB application ranges from 15,000 to 100,000 MW of coal-fired boiler capacity in the 31 eastern state acid rain region.

This Project Summary was developed by EPA's Air and Energy Engineering Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

A number of bills have been proposed by Congress that would require reductions of acid rain precursor emissions. These congressional bills would require different mixes of emission control technologies to achieve SO₂ and NO_x reductions at coal-fired utility power plants. The objective of this research program was to evaluate the impact of proposed acid rain legislation on the potential application of LIMB technology incorporating recent LIMB research and development findings.

A number of regulatory strategies and emission reduction targets were developed by reviewing acid rain legislation proposed in the 97th and 98th congressional sessions. For each regulatory strategy developed, the control technology mix of LIMB, FGD, and coal switching required to achieve the selected emission reduction level was determined. Next, the maximum number of boilers to which LIMB technology could be applied was determined by examining technical and regulatory constraints and emission reduction targets. The cost effectiveness of each regulatory case and control technology mix was estimated to evaluate the cost of each control technology mix.

Regulatory Case Development

The primary differences in the congressional bills are a result of the level of SO₂ reductions that is required at each plant due to plant/boiler specific emission limits or due to requiring high overall SO₂ reduction levels. All of the bills use 1980 as the base year for which emission reduction levels apply. Differences in the method of calculating excess emissions, implementation years, financing methods, and state reduction allocation and implementation were not considered important for the purposes of this study. The following three legislative/regulatory cases were analyzed:

Regulatory Case	SO ₂ Reductions, million tons/yr
Boiler Performance Standard	10
Regional Reduction Levels	10
Regional Reduction Levels	8

For this study, a regulatory strategy was developed based on bills which base reductions on boiler and/or state reduction performance standards (S 1709, HR 4816, HR 3400). These bills require that existing boilers must comply with New Source Performance Standards (1971 or 1979) if their emissions are greater than a specified amount of SO₂ per million Btu of fuel. These bills also require state wide reductions. Because these bills require very high levels of SO₂ reduction at individual plants/boilers, the use of wet FGD will be required at most affected plants. These legislative cases are entitled "Boiler Performance" cases, and FGD is applied to boilers at the largest emitting utility power plants.

The other major type of bill introduced in Congress (HR 4829, S 3041) allocates state level emission reductions generally based on the portion of emissions from facilities with emission rates greater than 1.2 lb SO₂ per million Btu fuel input. These bills allow the states to determine how the allocated emission reductions for that state are to be achieved and in some cases allow trading of emission reductions. Because these bills provide much greater flexibility in how emission reductions are achieved on a plant/boiler basis, they do not require the use of certain types of SO₂/NO_x control technologies. Study cases based on this type of legislative scenario are entitled "Regional Reduction" cases.

The other major difference between bills that would impact the mix of control technologies used by utilities is the amount of emission reduction required because, as the SO₂ reduction target increases, the average emission reduction needed to be achieved at each coal-fired boiler increases. For this study three SO₂ emission reduction levels were evaluated: 8 and 10 million tons per year, consistent with the different levels proposed by the congressional bills reviewed; and 12 million tons per year, a sensitivity case to evaluate the impact that this level of reduction would have on the control technology mix needed to achieve this high level of reduction.

The Congressional bills differ in the amount of credit given for NO_x reductions. For this study half credit was given for NO_x reductions; e.g., 1.0 ton of NO_x removed equals 0.5 ton of SO₂ reduction. Thus, for this study, a NO_x credit was included for low NO_x combustion modification assumed to be made with furnace sorbent injection.

Region and Boiler Specific Data Base

A major part of the study was development of a boiler specific data base and boiler specific control costs for LIMB, FGD, and coal switching. Developing an accurate data base for all coal-fired boilers in the 31 eastern states was not feasible. However, an accurate data base was easily developed for the top 100 SO₂ emitting coal-fired utility power plants. These top 100 plants accounted for over 72% of total U.S. utility power plant SO₂ emissions in 1980. Results of the applicability study for the top 100 plants were then extrapolated to the boilers in the 31 eastern state region. SO₂ emission reduction targets used for each regulatory case, based on allocating 72% of the emission reduction target to the top 100 coal-fired boiler population, are

Regulatory Strategy	SO ₂ Emission Reduction From Top 100 Plants, 10 ⁶ tons per year	Total Required SO ₂ Reduction, 10 ⁶ tons per year
Boiler Performance Standard	7.2	10
Regional Reduction	7.2	10
Regional Reduction	5.8	8
Regional Reduction	8.6	12

Control Technology Performance/Cost

Three coal-fired boiler SO₂ reduction technologies were examined: (1) limestone FGD with 90% SO₂ control; (2) LIMB with 50-60% SO₂ control and 50% NO_x control; and (3) switching to 2.5 lb SO₂ per million Btu eastern bituminous coal.

Boiler specific costs for FGD and LIMB were provided, using the IAPCS-2 computer model. Table 1 summarizes the cost/performance assumptions used to make the computer runs.

The cost of coal switching was based on a coal cost differential of \$1.00 per million Btu above the current higher sulfur coal. Although boiler specific costs for high and low sulfur coals were available, due to the current soft market, several plants are actually obtaining low sulfur coal at prices below high sulfur coal. This is not anticipated if many plants were required to switch coals because the added demand for low sulfur coal would drive up its price relative to high sulfur coals.

Discussion of Results

Figures 1 and 2 summarize the results of the 10 million ton per year SO₂ reduc-

tion cases. Figures 3 and 4 summarize the results of the 8, 10, and 12 million ton per year SO₂ reduction cases.

10 Million Ton Per Year SO₂ Reduction Cases

Figure 1 summarizes the results of the 10 million ton per year SO₂ reduction cases. Two cases were run for the Boiler Performance Standard strategy to provide an upper and lower bound on the amount of LIMB which would be used to achieve the desired SO₂ reductions. In both cases, FGD was applied to the boilers in the top 50 SO₂ emitting power plants with post 1965 service year achieving over 5.5 million tons per year of SO₂ reduction. In the first case LIMB was applied to the remaining boilers which were considered technically applicable (post 1960

wall/tangential fired boilers with sulfur emissions between 1.2 and 6.0 lb/million Btu). This case results in 69,000 MW of FGD application, 13,000 MW of LIMB application and 3,000 MW of coal switching. For the second Boiler Performance Standard case, coal switching (MAX CS) was applied before LIMB resulting in 8,400 MW of coal switching. Because coal switching can be achieved on the 1950's boiler to meet the required emission reduction target, no LIMB was applied.

Three different cases were run for the 10 million ton per year regional allocation scenario. The first two cases provided an upper and lower bound on the amount of LIMB which would be used versus coal switching. The other case looks at the impact of high performance (HP) LIMB (60% SO₂ reduction). For the maximum (MAX) LIMB case, LIMB was applied first to the applicable boilers resulting in half of the boiler population (71,000 MW) being controlled with the LIMB technology, 15,000 MW of FGD, and 11,000 MW of coal switching. For the second 10 million ton reduction case, coal switching was maximized (MAX CS) by applying it first to all the 1950's boilers. This reduces LIMB application to 65,000 MW and increases

Table 1. Performance and Cost Parameters Used to Estimate FGD and LIMB Annualized Costs and Emission Reductions

LIMB Performance Parameters	
50% LIMB Cases	60% LIMB Cases
50% SO ₂ Reduction	60% SO ₂ Reduction
50% NO _x Reduction	50% NO _x Reduction
Calcitic Hydrate	Calcitic Hydrate
2.5:1 Ca/S Ratio	3:1 Ca/S Ratio
700°F Quench Rate	700°F Quench Rate
ESP upgrade and SO ₃ conditioning for control of additional particulate matter.	
FGD Performance Parameters	
90% SO ₂ Reduction and No NO _x Reduction	
Limestone Slurry Sorbent	
No Spare Absorbers	
Number of Absorber Towers Based on Boiler Size:	
Boiler Size, MW	No. of Towers
<100	1
100-250	2
250-500	3
500-750	4
>750	5
General Cost Bases	
EPRI Cost Premises Used	
Costs are in 1995 Dollars	
Equipment Book Life of 15 Years	
FGD Retrofit Difficulty Factor: 1.2 Times New Plant Cost	

coal switching to 25,000 MW of application. For the third 10 million ton per year reduction case, high performance (HP LIMB) LIMB was applied, followed by FGD and coal switching as in the MAX LIMB case. This case decreases the penetration of FDG due to the greater SO₂ reduction achieved by high performance (60%) LIMB technology.

Figure 2 summarizes the cost results in the five 10 million ton per year SO₂ reduction cases. The boiler performance standard cases have the highest annual control cost of \$13-\$14 billion per year due to the large number of boilers which must apply FGD. The regional annual costs of the regional reduction level cases are significantly lower and range from \$9.9 to \$11.7 billion per year.

8, 10, and 12 Million Ton Per Year Cases

Figure 3 presents the results analyzing the impact of various emission reduction scenarios on the application of LIMB. The 10 million ton per year SO₂ reduction case is the same as for the Max LIMB regional allocation case discussed above. For this case, 71,000 MW of LIMB was applied to achieve the emission reduction target.

For the 8 million ton per year reduction case, coal switching to the 1950's boilers was applied first (lowest unit cost), followed by LIMB and FGD to achieve the emission reduction target. This results in boiler application of 71,000 MW of LIMB, 25,000 MW of coal switching, and 3,200 MW of FGD.

For the 12 million ton per year emission reduction case, the application of LIMB cannot be maximized if the emission reduction target is to be achieved. For this case, LIMB application was reduced by increasing the use of FGD and allowing all boilers where FGD and LIMB were not applied to switch coal. This results in the following boiler applications: 38,000 MW of LIMB, 50,000 MW of FGD, and 25,000 MW of coal switching.

Figure 4 presents the annual cost for the three cases. The annual costs and unit costs increase significantly as the emission reduction levels increase over 10 million tons per year:

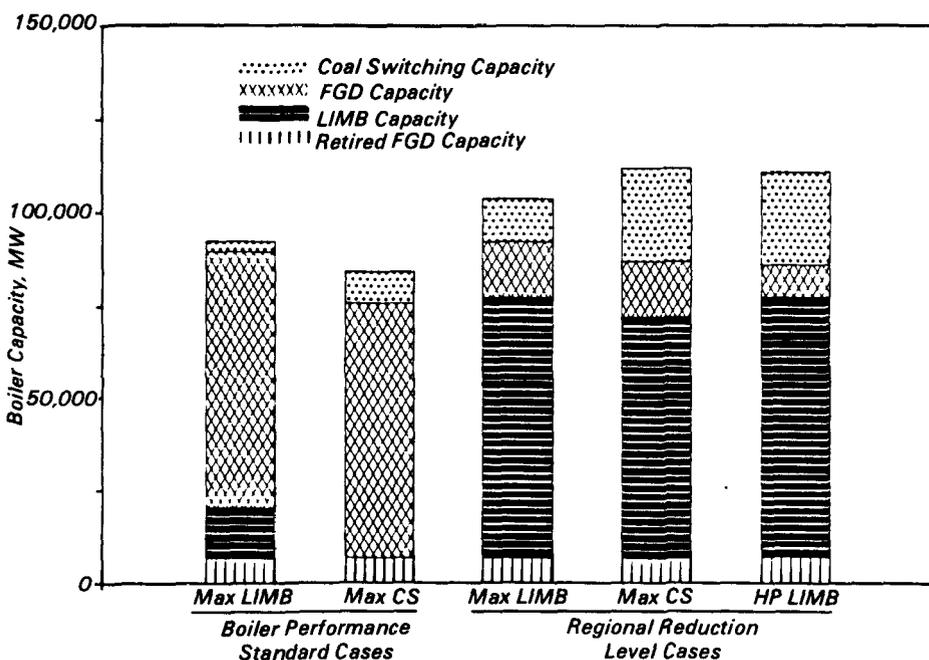


Figure 1. Boiler application results for 10 million ton per year of SO₂ reductions.

Annual Reduction, 10 ⁶ tons per year	Increase Emission Reduction, %	Increase in Cost, %	Average Unit Cost, \$/ton
8	—	—	1381
10	25	23	1397
12	50	73	1678

These cost increases are due to the significantly increased application of FGD needed to obtain the very high overall average emission reductions per boiler/plant.

31 Eastern State Region

To estimate the potential LIMB applicability for all of the coal-fired boilers in the 31 eastern state region, the number of boilers in that region that fit the LIMB and FGD technical applicability was determined from the 31 eastern state utility boiler data base. The amount of capacity for which LIMB was applicable was 103,000 MW. The amount of FGD capacity for this boiler population was 108,000 MW.

The average unit cost of applying FGD to the applicable boilers not in the top 100 plants is significantly greater due to the smaller boiler sizes and lower coal sulfur contents. This means that LIMB technology would be favored over FGD, and the LIMB applicability potential for the 10 million ton per year SO₂ reduction strategy not mandating the use of FGD could be as high as 100,000 MW of boiler capacity.

Conclusions

This study indicates that up to 100,000 MW of boiler capacity of LIMB application is possible depending on the type of acid rain legislation adopted and the amount of coal switching that is economically and politically practical. Currently proposed legislative strategies requiring SO₂ reductions of 8-10 million tons per year will maximize the application of LIMB because it is anticipated to be more cost effective than FGD. Control strategies requiring SO₂ reductions greater than 10 million tons per year will decrease the application of LIMB, because the average level of SO₂ control required at each boiler would exceed that available with a broad application of LIMB. Legislative strategies which would require high levels of control (>60%) at each boiler would also reduce the application of LIMB unless combined with fuel substitution.

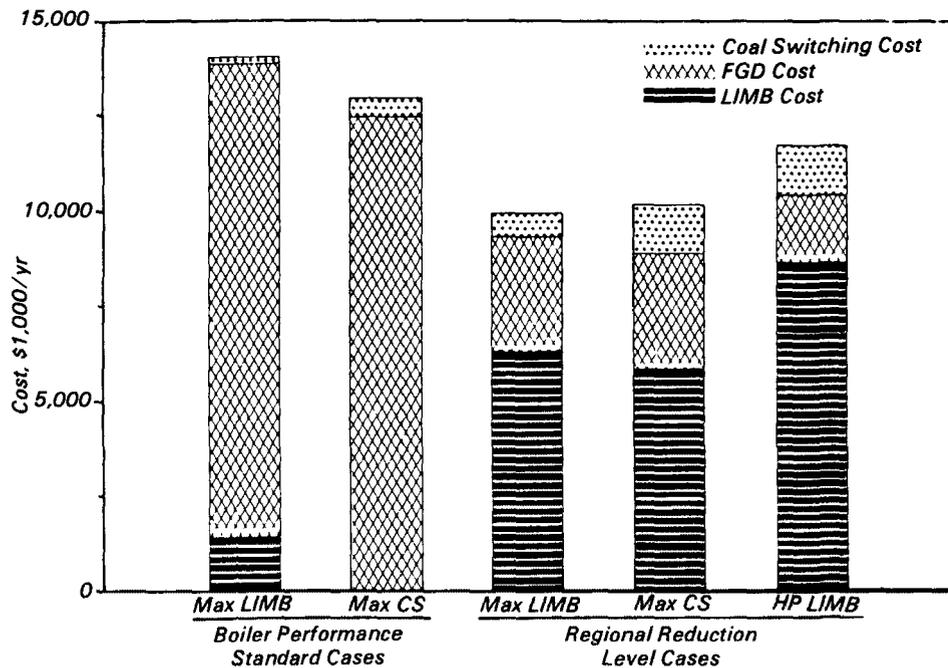


Figure 2. Levelized annual cost of control (1995 \$).

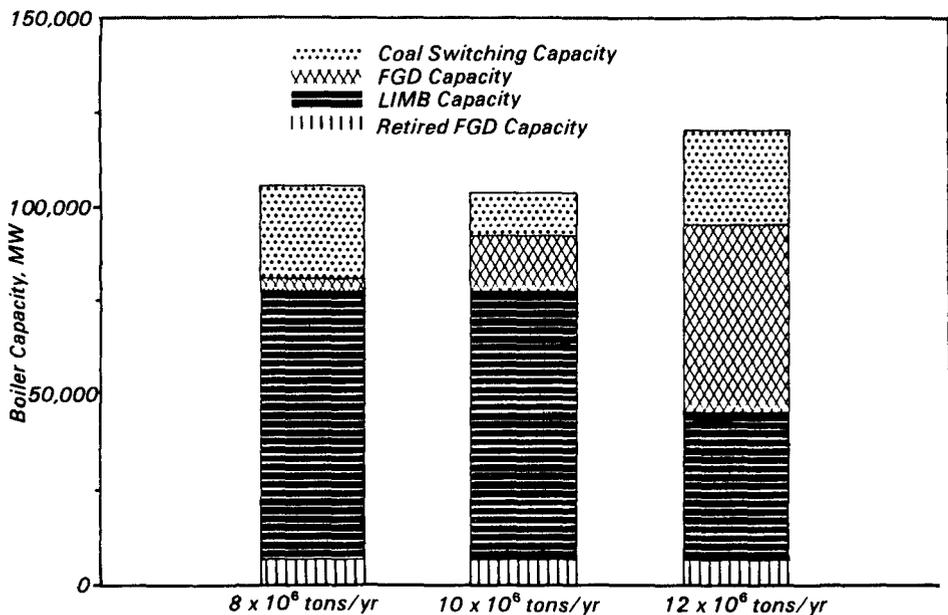


Figure 3. Boiler application results for 8, 10, and 12 million ton per year cases.

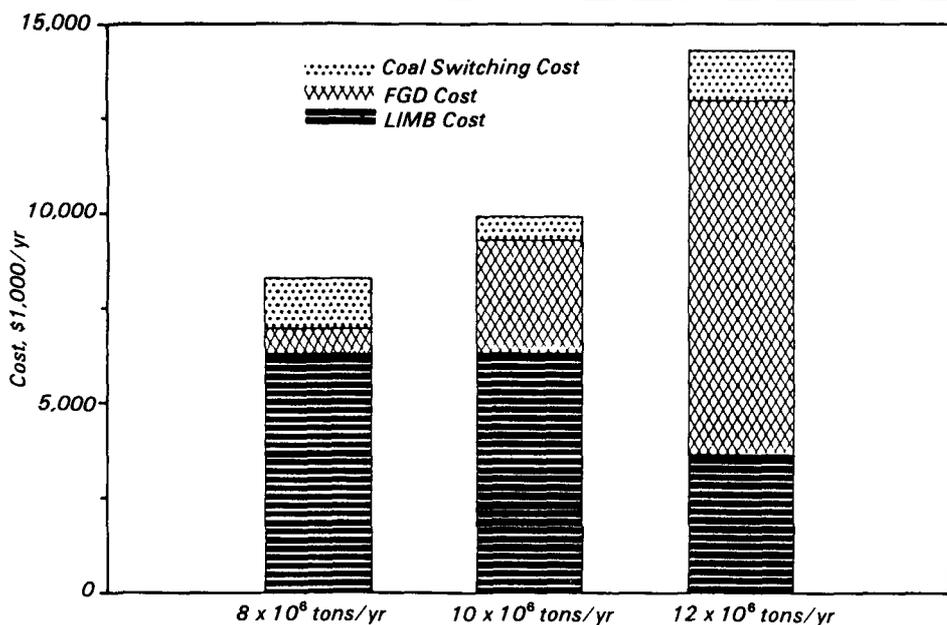


Figure 4. Levelized annual cost of control for 8, 10, and 12 million ton per year cases (1995 \$).

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The complete report, entitled "Analysis of Utility Control Strategies Using the LIMB Technology," (Order No. PB 87-100 574/AS; Cost: \$9.95, subject to change) will be available only from:

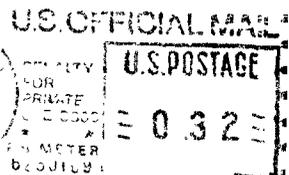
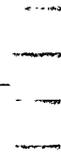
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