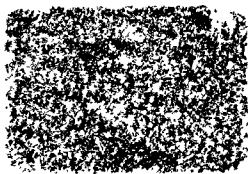




Environment
Canada

Environnement
Canada



Environmental
Protection
Service

Service de la
protection de
l'environnement

Technology for Controlling Emissions from Power Plants Fired with Fossil Fuel

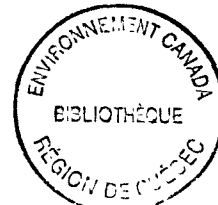
TD
182
R46
No. 3
AP-81-3

Rég. Québec Biblio. Env. Canada Library



Economic and Technical Review
Report EPS 3-AP-81-3

Air Pollution Control Directorate
April 1981



Canada

ENVIRONMENTAL PROTECTION SERVICE REPORT SERIES

Economic and Technical Review Reports relate to state-of-the-art reviews, library surveys, industrial inventories, and their associated recommendations where no experimental work is involved. These reports are undertaken either by an outside agency or by the staff of the Environmental Protection Service.

Other categories in the EPS series include such groups as Regulations, Codes, and Protocols; Policy and Planning; Technology Development; Surveillance; Training Manuals; Briefs and Submissions to Public Inquiries; and, Environmental Impact and Assessment.

Inquiries pertaining to Environmental Protection Service Reports should be directed to the Environmental Protection Service, Environment Canada, Ottawa, Ontario, Canada, K1A 1C8.

TECHNOLOGY FOR CONTROLLING EMISSIONS FROM POWER
PLANTS FIRED WITH FOSSIL FUEL *

TD

182

R46

ms. 3-AP-81-3

Combustion Sources Division
Engineering Assessment Branch
Air Pollution Control Directorate

Report EPS 3-AP-81-3
October 1981

* Prepared for Environment Canada by A.V. Slack, SAS Corporation, Wilson Lake
Shores, Sheffield, Alabama.

ABSTRACT

This report discusses the technologies available to control emissions of sulphur oxides, nitrogen oxides and particulate matter from thermal power plants. Advantages, disadvantages and costs of the individual technologies are assessed in relation to regulatory requirements and the desired level of control. Oil, natural gas and coal are the fuels covered, with the emphasis on coal, the one posing the greatest pollution problem.

The report is in support of the national emission guideline on thermal power plants and part of future efforts to control acid rain.

RÉSUMÉ

Le présent rapport traite des actuelles techniques de lutte contre la pollution par les oxydes de soufre, les oxydes d'azote et les particules émanant des centrales thermiques. Les avantages, les inconvénients et les coûts sont évalués pour chacune des techniques par rapport aux exigences des règlements et au degré de dépollution souhaité. Les combustibles étudiés sont le pétrole, le gaz naturel et le charbon, notamment ce dernier, car c'est celui qui pose le plus grand problème de pollution.

Les renseignements contenus dans le rapport viennent appuyer les lignes directrices nationales applicables aux émissions des centrales thermiques et les efforts futurs qui seront déployés pour lutter contre les pluies acides.

TABLE OF CONTENTS

	Page
ABSTRACT	i
RÉSUMÉ	ii
LIST OF FIGURES	v
LIST OF TABLES	v
EXECUTIVE SUMMARY	vi
1 INTRODUCTION	1
2 SULPHUR OXIDES	2
2.1 Low-Sulphur Fuel	2
2.2 Fuel Desulphurization	2
2.2.1 Physical Desulphurization of Coal	3
2.2.2 Chemical Desulphurization of Coal	9
2.2.3 Desulphurization of Oil	12
2.3 Coal Conversion	12
2.3.1 Applicability of the Technology	14
2.3.2 Cost Factors	15
2.4 Desulphurization during Combustion	15
2.4.1 Standard Boilers	16
2.4.2 Fluidized Bed Combustion	16
2.5 Flue Gas Desulphurization -- Dry Processes	20
2.5.1 Sodium Bicarbonate	20
2.5.2 Ammonia	20
2.6 Flue Gas Desulphurization -- Semi-Dry Processes	21
2.6.1 Process Evaluation	22
2.6.2 Cost	24
2.6.3 Status	24
2.7 Flue Gas Desulphurization -- Wet Scrubbing	25
2.7.1 Lime-Limestone	26
2.7.2 Double Alkali	32
2.8 Flue Gas Desulphurization -- Recovery Processes	33
2.8.1 Process Description	34
2.8.2 Cost	35
2.8.3 Status	35
2.9 Conclusions	36
3 NITROGEN OXIDES	38
3.1 Combustion Modification	38
3.1.1 Staged Combustion	39
3.1.2 Gas Recirculation	40

	Page	
3.1.3	Boiler Design Changes	40
3.1.4	Low-NO _x Burners	40
3.2	Fluidized Bed Combustion	40
3.3	Non-catalytic Reduction	41
3.4	Catalytic Reduction	42
3.4.1	Process Evaluation	42
3.4.2	Cost	45
3.4.3	Status	45
3.5	Conclusions	45
4	PARTICULATE MATTER	47
4.1	Electrostatic Precipitation	48
4.1.1	Design Problems	48
4.1.2	Cost	49
4.1.3	Status	50
4.2	Fabric Filtration	50
4.2.1	Design and Operation	50
4.2.2	Cost	51
4.2.3	Status	52
4.3	Wet Scrubbing	52
4.4	Conclusions	53
	REFERENCES	54

LIST OF FIGURES

		Page
1	GENERAL ARRANGEMENT OF EQUIPMENT FOR FLUE GAS CLEANING	vii
2	USE OF A COAL GASIFIER IN COMBINED-CYCLE POWER GENERATION	x
3	DESULPHURIZATION IN A FLUIDIZED BED BOILER	xii
4	ALTERNATE LOCATIONS FOR PRECIPITATORS	xviii
5	CONTROL OF NO _x BY CATALYTIC REDUCTION WITH AMMONIA	43

LIST OF TABLES

		Page
1	TYPICAL UNCONTROLLED EMISSIONS OF POLLUTANTS	viii

EXECUTIVE SUMMARY

The emphasis on controlling emissions from fossil-fuel-fired power plants is shifting from local considerations to regional concerns regarding problems such as acid rain, impaired visibility, and respirable particulates. In view of this, a reassessment of abatement methods is needed, aimed at determining which of the processes are most capable, from the standpoints of both control efficiency and cost effectiveness, of accomplishing the degree of control needed from the regional viewpoint. This report presents such a reassessment.

In general, the optimum process for controlling a given pollutant depends on the degree of control required. Processes that reduce emissions to an extreme degree are quite expensive and are usually not cost effective unless the high efficiency is essential. On the other hand, techniques that cost less normally are not capable of a high degree of control.

Another consideration is that processes differ in regard to operating reliability. If it is desired to meet a given emission standard continuously and without exception, then some methods are at a disadvantage because the spare equipment required for 100% reliability is more extensive and more costly than for other processes. On the other hand, if the rules allow averaging the emissions over an extended period (say a month), there can be periods of downtime for repair and maintenance without violating the standard or increasing overall emissions.

The pollutants covered in this review are sulphur oxides (SO_2 and SO_3), nitrogen oxides (NO and NO_2 , generally referred to as NO_x), and solid material carried in the gas stream (ash from the fuel, unburned carbon, and other non-gaseous particles -- all generally referred to as particulates). Most of these pollutants come from the fuel itself, by reaction of sulphur and nitrogen compounds with oxygen supplied by the combustion air and burning out the combustible compounds leaving the ash as small solid particles. In addition, some NO_x is formed by reaction of nitrogen and oxygen from the combustion air.

The amounts of these pollutants vary with type of fuel, size of boiler, and load factor (percentage of time operated). Typical data are given in Table 1. Although the tonnages listed are high, the concentrations in the flue gas are quite low because of the very large total tonnage of gas, which is composed mainly of nitrogen, carbon dioxide and water vapour; the 500-megawatt (MW) boiler referred to in Table 1 would produce a total of about 60 000 tons of gas per day or about 13 million tons per year.

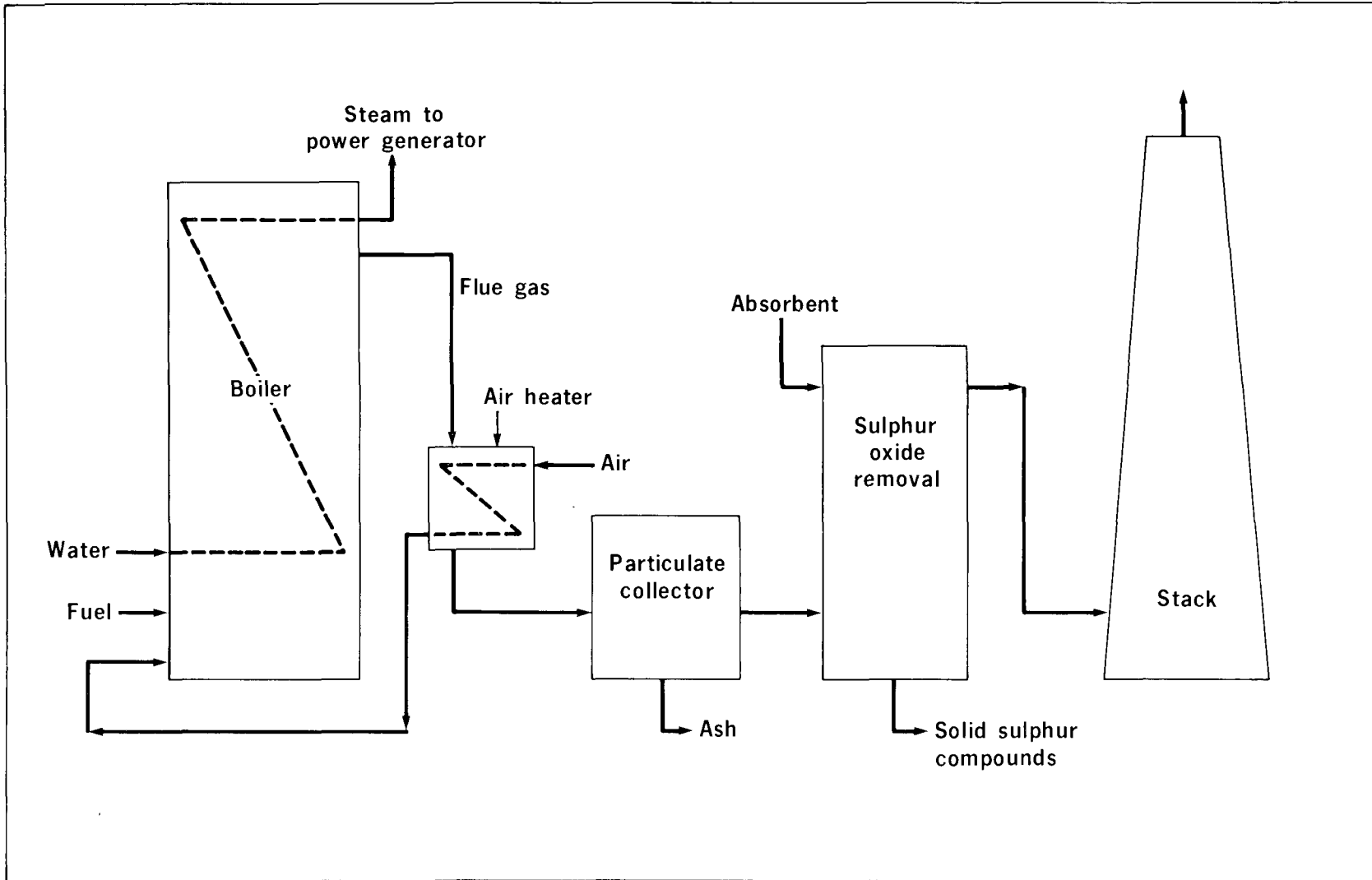


FIGURE 1 GENERAL ARRANGEMENT OF EQUIPMENT FOR FLUE GAS CLEANING

TABLE 1 TYPICAL UNCONTROLLED EMISSIONS OF POLLUTANTS*

Fuel	Pollutant, tons per year		
	Sulphur oxides	Nitrogen oxides	Particulates
Natural gas	**	4 000	**
Oil (1% sulphur)	15 000	4 800	1 100
Coal			
Low sulphur (0.7%)	15 000	8 200	110 000
High sulphur (4%)	88 000	8 200	110 000

* 500-megawatt boiler, 60% load factor. Levels given are fairly typical; in practice they vary over a wide range. Coal burned, about 1.1 million tons per year.

** Natural gas normally contains very little sulphur or ash.

The large amounts of pollutants emitted have led to regulations for reducing emissions. As might be expected from Table 1, the main emphasis in the past has been on particulate control where coal is the fuel because of the large amount involved; devices to collect and remove particulates from the gas stream have been required for a long time. Sulphur oxides have been receiving attention in recent years, especially for high-sulphur coal. Emphasis on nitrogen oxide control is just beginning, mainly in congested areas such as in Japan and southern California in the United States.

Sulphur oxides and particulates are removed from the gas stream by a variety of methods. (Sulphur oxide emissions can also be reduced by using low-sulphur fuel.) The general arrangement of the equipment is shown in Figure 1. For nitrogen oxides, the practice has been to reduce emissions by altering combustion conditions in the boiler in such a way as to reduce NO_x formation. Since this is only partially effective, there has been some use in Japan of a control system to remove NO_x from the gas. In Figure 1, the control system would be located between the boiler and the air heater.

Sulphur Oxides

In the past, one of the main approaches to sulphur oxide control in countries such as Japan and the U.S. has been to use naturally occurring low-sulphur fuel. This is still the practice in Japan, but the U.S. recently enacted federal regulations requiring a reduction in emissions for all new boilers -- and pressure is growing to require such a

reduction for existing units. Several approaches can be used to attain the reduction, including fuel desulphurization, coal conversion, desulphurization during combustion, and flue gas desulphurization (FGD).

Fuel Desulphurization. For coal, part of the sulphur can be removed at relatively low cost by physical methods, that is, the coal is subjected to a treatment based on gravity differences to separate the mineral matter from the coal. Frequently, the mineral matter contains much of the sulphur, and some 10-30% of this sulphur can be removed fairly easily. A variety of methods are used, including washing, shaking (on a special device that separates heavy from light material), and mineral concentration methods such as "sink-float" (based on differences in density of the coal and the impurities).

The problem is that if more than 10-30% removal of the sulphur is required, physical cleaning becomes expensive. It can be combined with other methods to advantage if an intermediate degree of removal is acceptable. For 90% and higher removal (10% or less left in the coal), however, as is now required in the U.S. for new plants, other methods are more cost effective.

Coal can also be cleaned by chemical methods; these are considerably more expensive than physical cleaning and do not seem capable of 90% and higher removal efficiencies. Oil desulphurization is in widespread use in Japan and California; high removal efficiency (90% or more) is possible but expensive.

Coal Conversion. Although not aimed primarily at desulphurization, coal conversion processes (production of liquid or gaseous fuels) accomplish a very high degree of sulphur reduction incidentally. If the resulting fuels are used in power boilers, the emission reduction goal is accomplished. This method has been commercialized in South Africa (SASOL project), but in North America, where economic conditions are different, coal conversion has not made much headway. A new method, called solvent refining of coal (liquid or solid product), has shown some promise, but the product is expensive and the process is unproven on a commercial scale.

Another approach is production of low-Btu gas by coal gasification, removing ash and sulphur, and burning the gas in a combined-cycle operation (Figure 2 shows the use of a gas turbine and boiler in series to improve energy recovery). In this case, an increase in energy efficiency is a major objective in addition to desulphurization, which

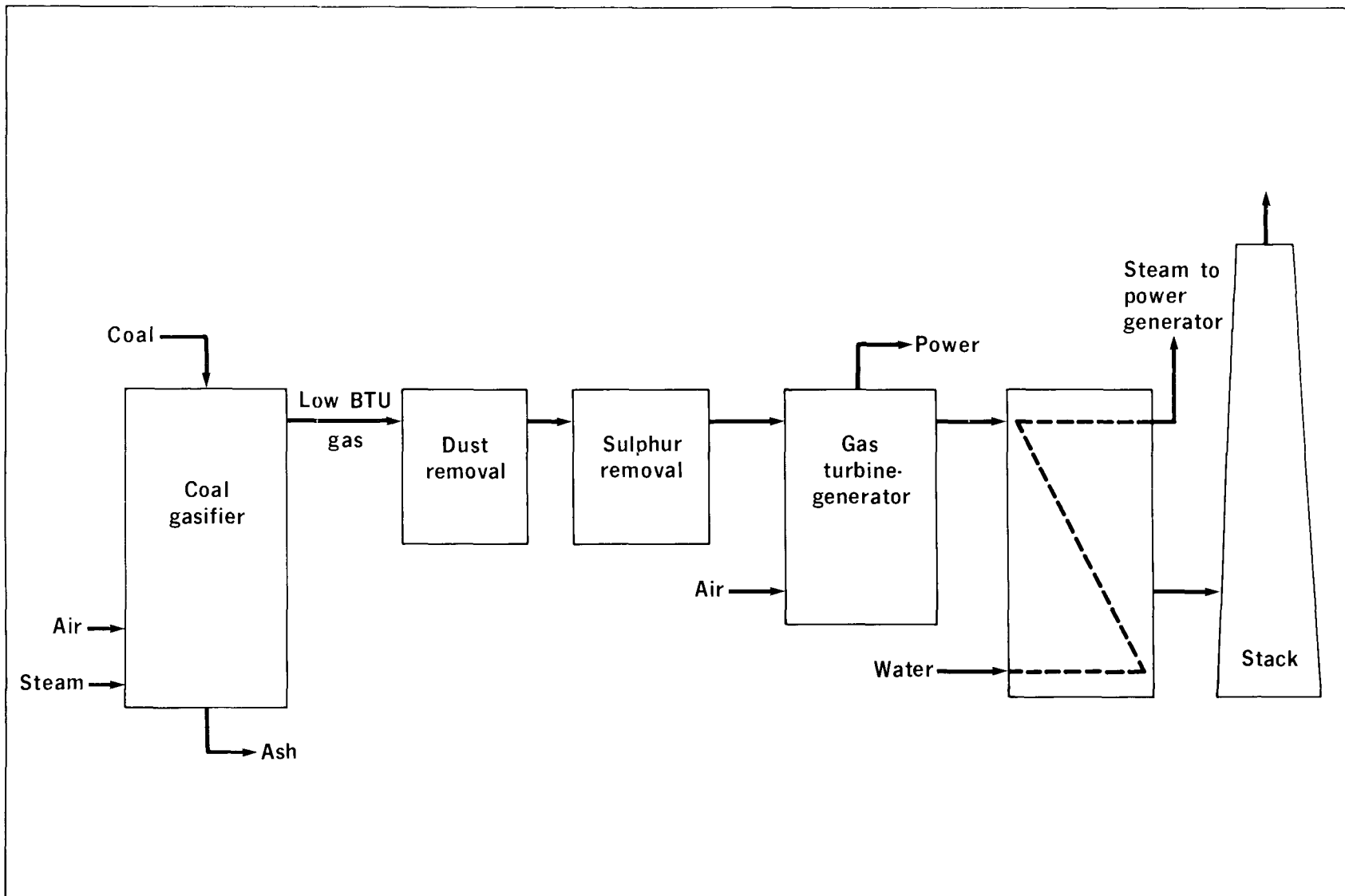


FIGURE 2 USE OF A COAL GASIFIER IN COMBINED-CYCLE POWER GENERATION

complicates estimation of the sulphur removal cost. Most estimates show a cost reduction of about 15% by the combined-cycle route (based on cost per kilowatt hour (kWh)) compared with a conventional boiler with FGD, but commercialization is probably 15 to 20 years away. Moreover, the cost of new processes tends to go up as development work progresses.

Desulphurization during Combustion. The most promising method in this area is fluidized bed combustion. In the fluid bed process, air blown up through a bed of fine coal and limestone burns the coal in a suspended state (see Figure 3) and produces steam in water tubes submerged in the bed. The limestone reacts with the SO_2 . The capital cost for SO_2 removal should be low because no separate reactor is needed. The main drawback is the difficulty in reaching a high level of SO_2 removal without using an inordinate amount of limestone. To get 90% removal, some two to four times as much limestone is required as for limestone wet scrubbing (gas washing with a limestone slurry, the standard process).

Estimation of sulphur control cost for fluidized bed combustion is complicated because reduced boiler cost is an objective as well as sulphur removal. Proponents' estimates generally show a saving of 10-15% per kilowatt hour compared with a conventional power plant equipped with wet scrubbing; other estimates show the two about even. Commercialization for use in power plants is probably about a decade away.

Flue Gas Desulphurization -- Dry Processes. One of the newer developments is injection of a lime slurry into a spray drier concurrently with the flue gas. The lime reacts with the SO_2 to form a dry, solid product that is collected downstream in an electrostatic precipitator or fabric filter (usually called a baghouse). The main advantages are relative simplicity of equipment, production of a dry waste material rather than a wet sludge, lower energy requirements, and possibly lower maintenance requirements and better reliability. The drawbacks are need for lime (more expensive than limestone) and difficulty in getting a high degree of removal. The latter effectively limits the process to low-sulphur coal (below 1%, assuming high-Btu eastern coal).

Only pilot-plant data are available but enthusiasm for the process has led utilities to contract for several installations in the U.S. There is some indication from bidding situations that the process does not have as much cost advantage as expected.

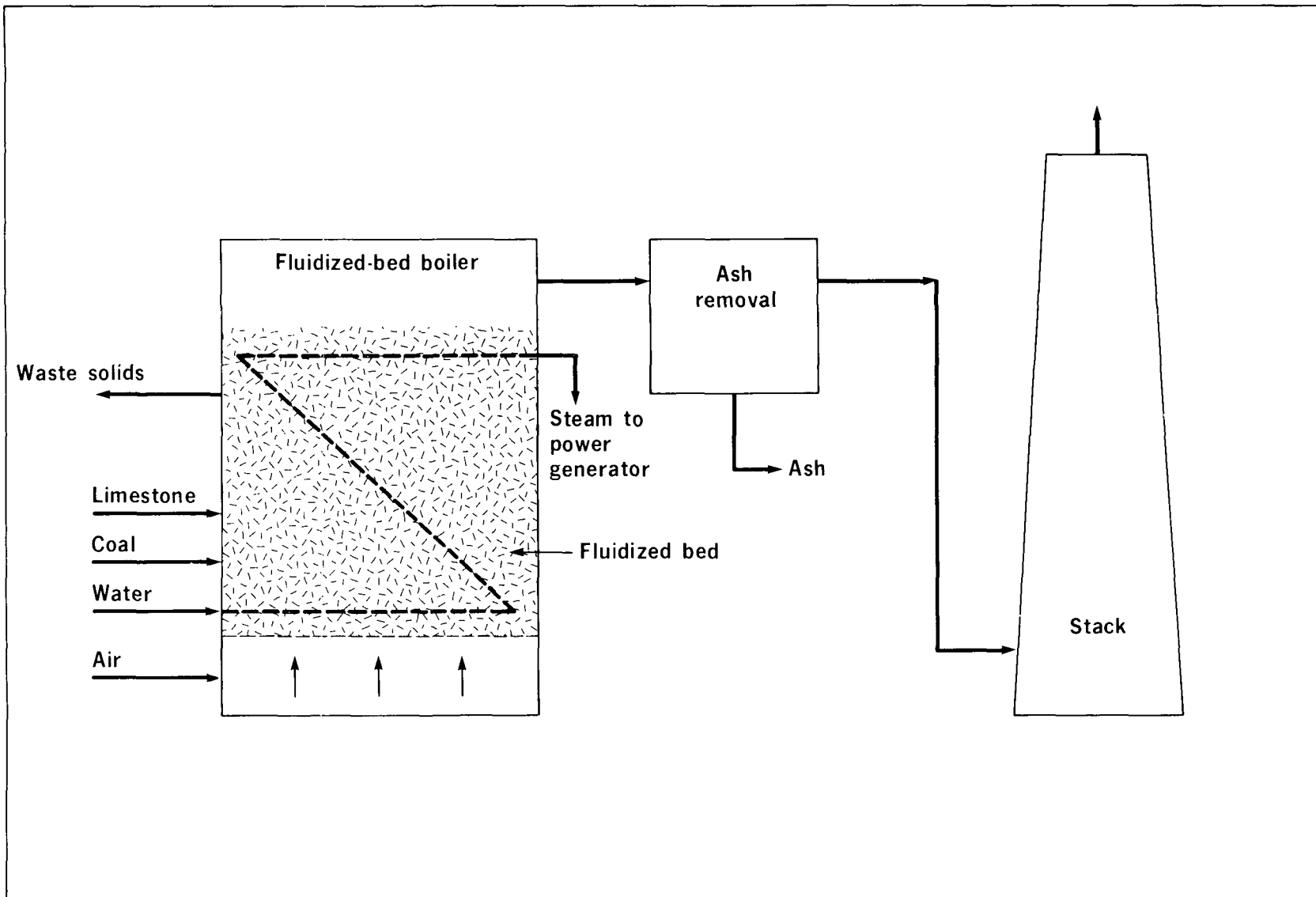


FIGURE 3 DESULPHURIZATION IN A FLUIDIZED-BED BOILER

Flue Gas Desulphurization -- Wet Scrubbing. Scrubbing the flue gas with a limestone slurry has become the basic FGD process. It has been commercial for over 10 years, limestone is the least expensive sulphur absorbent available, and no competitive process has been demonstrated to be more cost effective. The main drawbacks are corrosion/erosion in the scrubbers, stress on the very large slurry circulation pumps, and production of a wet, difficult-to-handle sludge. Moreover, there has been some trouble with reliability, particularly with high-sulphur coal.

Progress has been made in all these areas but the problems are still not completely resolved. One of the main questions is whether the 90% SO₂ removal now required in the U.S. for new boilers can be attained; such a degree of compliance has not been required in the U.S. before. It may be necessary to add small amounts of promoters such as magnesium compounds or adipic acid; tests have shown these to be quite effective in raising removal capability. Spare equipment is being generally installed as a means of ensuring reliability.

Limestone scrubbing systems are generally bid by suppliers at \$30-45 per kilowatt (vendor-supplied equipment only, erected).^{*} With owner costs added, the price range is approximately \$60-135/kW. The wide range is due to site-specific considerations and to variations in the bids from different vendors. The operating cost is in the order of 4-6 mills/kWh, or \$10-15 per ton of coal burned. (The typical cost for coal fed to the boiler is \$25-30 per ton.)

Lime has some operating advantages over limestone and is sometimes used, but the higher cost for lime makes limestone the usual choice.

Lime-limestone scrubbing is widely used in all countries -- Japan, the U.S., and West Germany -- where SO₂ emission reduction is required. In the U.S., the capacity currently operating on utility boilers is over 19 000 MW and another 53 000 MW is under construction or planned. It is estimated that nearly 160 000 MW will be in operation by 1990.

One of the scrubbing variations is the so-called double alkali process. The SO₂ is absorbed in a clear solution of a strong alkali and the resulting solution is then treated with lime to precipitate the sulphur as a solid compound that can be separated and discarded. The advantages of the process are very high removal efficiency and better scrubber operation because of the clear solution.

^{*}All costs in this report are expressed in U.S. dollars.

Processes for recovering the sulphur as a useful product are also mainly of the wet type. All are so expensive and energy intensive that viability is questionable, and none is in commercial operation.

Process Choice. The following recommendations are made for process choice at different required levels of emission reduction. These are only approximate, and site-specific conditions could well change the rankings. The rankings are judgmental, based on a subjective evaluation of factors such as cost, commercial viability, absorption efficiency, and process reliability. A more quantitative approach to ranking does not seem feasible in view of the uncertainties involved.

<u>Removal efficiency level, %</u>	<u>Process ranking</u>
Higher than 90%	<ol style="list-style-type: none"> 1. Double alkali 2. Limestone scrubbing with promoters 3. Coal gasification (combined cycle)^a 4. Recovery processes
90%	<ol style="list-style-type: none"> 1. Limestone scrubbing with promoters 2. Limestone scrubbing 3. Double alkali
50-90% (high-sulphur coal)	<ol style="list-style-type: none"> 1. Limestone scrubbing with physical coal cleaning 2. Fluidized bed combustion^a 3. Chemical coal cleaning^a
50-90% (low-sulphur coal)	<ol style="list-style-type: none"> 1. Spray drier process 2. Limestone scrubbing
Below 50%	<ol style="list-style-type: none"> 1. Physical coal cleaning 2. Low-sulphur coal substitution

^a When and if developed.

Nitrogen Oxides (NO_x)

The alternatives for nitrogen oxide control are combustion modification to reduce NO_x formation and flue gas treatment to remove it from the gas. Combustion modification, the much less expensive of the two, is used in the U.S. and Japan. In situations where the regulations have become so stringent that combustion modification is not capable of achieving the required emission reduction, flue gas treatment is used. This has occurred in Japan and California.

Combustion Modification. NO_x is formed both by oxidation of nitrogen compounds in the fuel (fuel NO_x) and by reaction of oxygen and nitrogen in the combustion air at high temperature (thermal NO_x). The principal modifications used for NO_x emission reduction are lowering the flame temperature to reduce thermal NO_x (mainly by recirculating combustion gas from a point downstream from the boiler proper and mixing it with the combustion air) and using staged combustion to reduce fuel NO_x. The latter involves injecting the combustion air in two stages, normally by reducing air flow to the burner and injecting the remainder through "overfire" air ports in the side of the boiler. Low-NO_x burners that accomplish staged conditions within the burner flame have also been developed.

Staged combustion is the most cost-effective method but normally only reduces emissions by 15-25%. Gas recirculation is more expensive but is quite effective for gas or oil, giving an emission reduction of up to 50%. Low-NO_x burners are effective (up to 30% reduction) and are often used in Japan in combination with the standard type of staged combustion and with gas recirculation. The combination has given very low emissions in Japanese tests, 100 ppm with coal and 50 ppm with oil -- a reduction of 75-80% compared with uncontrolled emissions. There is the problem, however, that an advanced degree of combustion modification can cause slagging in the boiler and corrosion of heat transfer surfaces.

Combustion modification is the least expensive of the NO_x control methods. In Japan, the capital cost for low-NO_x burners is about \$2/kW and for overfire air ports about \$4.50/kW.

Flue Gas Treatment. The leading method is injection of gaseous ammonia to reduce NO_x to nitrogen. Operation without a catalyst requires very high temperatures and removal is limited to about 35-40%. With a catalyst, 90% or higher is feasible but

80% gives much less operating difficulty and may be the upper practicable limit for high-sulphur coal.

In the presence of the catalyst, usually a mixture of vanadium and titanium oxides, the reaction of ammonia with NO_x proceeds much more rapidly at lower temperatures. The catalyst is placed in the flue gas stream between the boiler and air preheater where the temperature is about 400°C . The catalyst lasts about two years and its cost makes up about half the total cost of the installation.

Designing for very high removal efficiency (90% or higher) aggravates problems such as ammonia emission and air heater plugging by deposition of ammonium bisulphate (from reaction of ammonia and sulphur trioxide). At lower efficiency levels, such problems can be minimized by feeding less ammonia.

The air heater plugging problem can also be minimized by using gas or low-sulphur oil as fuel. For high-sulphur coal, however, the problem is serious and has not yet been resolved.

The capital cost for catalytic reduction is typically \$35/kW; non-catalytic reduction costs about half as much (but is much less effective). The operating cost for a catalytic system is between 1.5 and 5 mills/kWh.

Process Choice. The situation is similar to that for other pollutants -- process choice depends on the degree of control required.

<u>Removal efficiency level, %</u>	<u>Process ranking</u>
90% or higher	1. Catalytic reduction with more than the normal amount of catalyst, preceded by combustion modification
50-90%	1. As above, with a normal amount of catalyst 2. Combustion modification (all types) followed by non-catalytic reduction (ammonia injection without catalyst) 3. Combustion modification alone (for low part of range so as to minimize boiler problems)

<u>Removal efficiency level, %</u>	<u>Process ranking (cont'd)</u>
Below 50%	<ol style="list-style-type: none"> 1. Staged combustion^a 2. Low-NO_x burners^a 3. Gas recirculation (except for coal)^a

a Used in combination with others if necessary to achieve the required reduction level.

Particulate Matter

The basic method in the power industry for removing particulate matter originating as ash from the fuel is electrostatic precipitation. Relatively few wet scrubbers have been installed for this purpose in the U.S. and the practice is dying out because the new stringent regulations cannot be met with scrubbers except at high pressure drop in the gas stream, which makes them unacceptably expensive to operate.

There is a trend to the use of fabric filters (baghouses) in the U.S. as a means of attaining the stringent emission standards adopted recently for new boilers.

Electrostatic Precipitators (ESP). The main problem in precipitator performance is high electrical resistivity of the ash. Low-sulphur coal gives the most difficulty; with high-sulphur coal there is enough sulphur trioxide in the gas to reduce resistivity by adsorption on the ash. The problem has been dealt with by adding SO₃ to the gas, changing the precipitator location to a point in the boiler train (see Figure 4) where the temperature is higher (which gives lower resistivity), and making the precipitator larger to give longer ash retention time.

Precipitator cost varies widely, depending on ash resistivity and the emission standard. The range is in the order of \$30-50/kW.

Fabric Filters. The use of baghouses is growing because they have an intrinsic capability for very high particulate removal. In the recent rule-making in the U.S. (June 1979), the particulate emission standard was in effect set at the level (0.03 lb/10⁶ Btu) that baghouses normally achieve. Precipitators can also meet this standard but they have to be made very large to do so, to the extent that baghouses are generally considered less expensive for such an extreme degree of control.

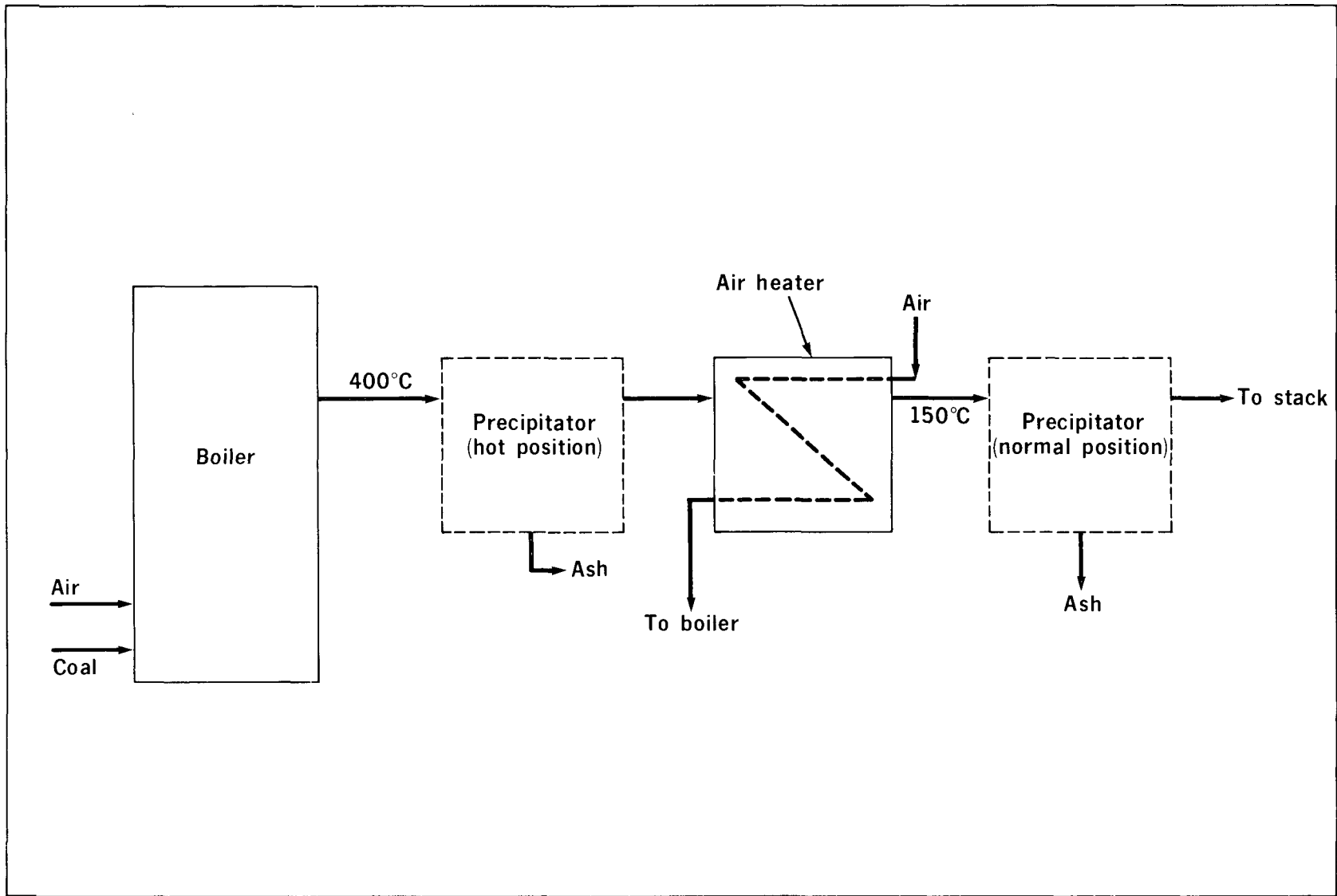


FIGURE 4 ALTERNATE LOCATIONS FOR PRECIPITATORS

The main problem is that there is little experience with baghouses for large utility boilers. Only two large installations are currently operating and both have had trouble. The principal difficulties are damage to the bags by various causes and pressure drop increase with time.

Baghouse cost does not depend on the ash characteristics or emission limits. Estimates vary but a recent bidding situation indicated a cost of about \$45/kW.

Wet Scrubbing. The limited ability of wet scrubbers to remove very fine particulate matter makes their use questionable for the present regulatory climate, an unfortunate situation because scrubbers can remove the bulk of the coarse particulate at very low cost. In a new development underway, a wet precipitator after the scrubber removes the fine particulate.

Process Choice. For the current new source standard for particulate emissions in the U.S. ($0.03 \text{ lb}/10^6 \text{ Btu}$), baghouses are probably superior for low-sulphur coal. For the high-sulphur type, the situation is not clear; more experience with baghouses is needed. For a standard such as $0.1 \text{ lb}/10^6 \text{ Btu}$, precipitators are more cost effective.

If the sequence of wet scrubbing followed by a precipitator is developed successfully, wet scrubbing could become the favoured method.

1 INTRODUCTION

The problem of emissions from power plants fired with fossil fuel, a matter of particular concern in the more industrialized areas of the world, is changing as the general levels of emission increase. In the past the main emphasis has been on maintaining air quality in the vicinity of the power station. The approach taken has been to model the dispersion pattern for a particular plant and thereby establish an emission limit low enough to prevent violation of the ambient standard in the area directly affected by the plant, particularly where the plume first reaches the ground.

The emphasis has been shifting recently to control on a regional or even global basis rather than the local approach, the reasoning being that direct effects of the pollutants (sulphur dioxide, nitrogen oxides, and particulate matter) are less harmful to health than the effects of secondary pollutants -- compounds or combinations that require some time to form and therefore appear far downstream from the point of origin.

The main adverse regional effects of power plant emissions are acid rain, impaired visibility, and health effects of respirable particulates. As yet there are few if any regulations that take such effects into account, but the sulphate standard in California and the visibility standards that the U.S. Environmental Protection Agency (EPA) is exploring may be indications of future trends.

The net effect of the regional emphasis is likely to be more stringent regulations for new fossil-fuel-fired power units and a wider application of controls to existing units (where the limited remaining life makes the cost/benefit ratio especially important). Such a possible tightening of the regulatory situation raises the question of whether the currently used technology is capable both of an extreme degree of control for new plants and of the cost effectiveness needed for existing plants -- and if not, whether new methods should be considered.

2 SULPHUR OXIDES

Sulphur oxide emissions can be decreased by several methods -- use of naturally occurring low-sulphur fuel; removal of the sulphur before combustion; reaction with an absorbent during combustion; or flue gas desulphurization.

2.1 Low-Sulphur Fuel

In the United States, passage of the Clean Air Act (1970) was the first major step toward reducing SO₂ emissions. New power boilers were limited by federal regulation to an SO₂ emission of 1.2 lb/10⁶ Btu, and state and local rules, especially State Implementation Plans (SIP's), limited emissions in some cases from existing boilers. Since the power industry considered the use of low-sulphur fuel to be the most cost-effective control method, there was a trend to low-sulphur oil and coal. Low-sulphur coal was especially popular during the 1970s, with eastern power companies shipping it from western states (mainly Montana and Wyoming).

The Clean Air Act Amendments of 1977 essentially ruled out low-sulphur coal as the sole control method for new boilers but it is still the standard strategy for existing ones. In the northeastern U.S., for example, the current projects for conversion from oil to coal will generally not require flue gas desulphurization because use of low- to medium-sulphur coal will be allowed as the means of SO₂ control.

In Japan, use of naturally occurring low-sulphur fuel (liquefied natural gas or low-sulphur crude oil) has become the main strategy for complying with the strict emission regulations in that country. In Europe, the European Economic Community (EEC) has been pushing for a limit of 1% sulphur in fuels used by the member countries. Although some of this would be desulphurized oil, the bulk would probably be naturally occurring low-sulphur oil or coal.

2.2 Fuel Desulphurization

In the U.S., the main use of desulphurized fuel has been in California and to some extent on the east coast, where desulphurized oil is generally used for oil-fired boilers. Coal desulphurization is not yet developed to the point where the desulphurized product is available for purchase, as is desulphurized oil.

In Japan and Europe, the situation is much the same. Desulphurized oil is used to some extent but not desulphurized coal; in contrast with the U.S., little or no development work is underway on coal cleaning.

2.2.1 Physical Desulphurization of Coal. Since much of the sulphur in coal is often concentrated in small mineral particles, some desulphurization can be accomplished by gravity separation methods based on differences in density. Such physical cleaning ranges from simple and relatively inexpensive methods (washing or shaking) to sophisticated "deep-cleaning" processes involving sink-float procedures that are relatively complicated and require a large investment. The complex methods remove more of the sulphur but they are costlier per unit of sulphur removed and are more difficult to operate.

In the U.S., some 50% of the domestic coal is physically cleaned to remove mineral matter and mining residue. Some sulphur is removed incidentally but seldom if ever enough to meet any sort of emission standard. Cleaning to remove sulphur has been limited in the past to metallurgical-grade coal. However, a coal-cleaning plant recently began operation to supply cleaned coal to the Homer City, Pennsylvania, generating station. This station has two 600-MW units, built in 1969, which must meet state SO₂ emission standards (4 lb/10⁶ Btu) for existing sources, and a new 650-MW unit that must meet federal new-source SO₂ emission standards (1.2 lb/10⁶ Btu). The purpose of the cleaning plant is to provide coal to these generating units that can meet the relevant federal or state SO₂ emission requirements without the need for flue gas desulphurization. Two grades of coal are required; 0.8% sulphur for the new unit and 2.25% sulphur for the older units. (To reach these levels, the feed coal (2.8% sulphur) is treated by a variety of well-established coal-cleaning techniques, including dense-media cyclones, hydroclones, and classifiers.)

The Homer City installation uses five distinct cleaning circuits. Overall energy recovery is about 95%.

- Crushing and classification. Much of the pyrite (iron sulphide) and non-combustible material is removed in this step.
- Fine-coal cleaning. The 2 mm x 100 mesh fraction is cleaned in dense-media cyclones at 1.3 specific gravity. Overflow material is washed of fines in special classifiers, giving a deep-cleaned product (0.88% sulphur) capable of meeting the SO₂ emission limit (1.2 lb/10⁶ Btu) for the new boiler.

- Medium-coal cleaning. Medium-size coal (1/4 inch x 2 mm) is cleaned in two stages of dense-media cyclones (at 1.3 and 1.8 specific gravity). Some deep-cleaned coal is produced (partly used in the new boiler) but most of the product is middling coal suitable for use in the two older boilers for which the SO₂ emission limit is 4 lb/10⁶ Btu. The sulphur content of this fraction is about 2.25%.
- Coarse-coal cleaning. The coarse coal (1-1/4 x 1/4 inch) from the original crushing is cleaned in a dense-media cyclone at 1.8 specific gravity, giving a further quantity of middling coal.
- Fine-coal scavenging. Much of the heating value of the coal remains in the very fine (<100 mesh) and heavy reject fractions from the deep-cleaning step. These are treated in a somewhat complicated assembly of hydroclones, centrifuges, thickeners, and filters to produce more middling coal. Over half the coal needed for the older boilers is recovered in this step.

The Allegheny coals used at Homer City are particularly amenable to physical cleaning techniques; not only do they have a high pyritic sulphur content (ratio of pyritic to organic sulphur, 3:1) but the pyrite can also be relatively easily liberated from the coal matrix.

In addition to the Homer City project, the Tennessee Valley Authority (TVA) has two large coal-cleaning installations under construction, the objective of which is to meet the state-imposed emission limits for two existing stations (Paradise and Cumberland, 2 500 MW and 2 600 MW respectively). No deep cleaning is planned; only enough cleaning will be done to remove some 30% of the sulphur. In the Paradise station, this will be supplemented by limestone scrubbing to get the overall station emission down to the state standard.

2.2.1.1 Applicability of technology. The main drawback to physical coal cleaning is its limited capability for removing enough sulphur to meet SO₂ emission regulations. Since the regulations vary widely, analysis of the situation is difficult. The main considerations are as follows.

New Plants

In most countries where a significant degree of SO₂ emission control has been required (mainly the U.S., Japan, and West Germany), new power stations have been subject to stringent regulations. In the U.S., for example, the regulations for new plants

are 90% or higher removal for any coal containing more than about 3% sulphur, 70% for about 1.2% sulphur and less, and between 70% and 90% for sulphur levels between 1.2% and 3%. High-sulphur coals are usually the ones most amenable to physical coal cleaning because of their high percentage of relatively easily removable pyritic sulphur. This is also generally true for Japan and Germany, although their indigenous coals have a relatively low sulphur content.

It is doubtful that any combination of physical coal-cleaning techniques could accomplish 90% sulphur removal except at exorbitant cost or at low recovery of the heating value (which also gives high cost unless some use can be found for the reject fraction). At Homer City, the pyritic sulphur removal for the cleaner fraction is reported to be 91.8%, but corresponds to only about 25% of the total tonnage of coal cleaned. The installation would not have been practicable, but two older units also required some reduction in SO₂ emissions (about 50%) so that the partially cleaned middling coal could be burned in these units. This situation is not often encountered. If the older boilers had not been involved, the partially cleaned coal would have had to be shipped elsewhere, and the economics would therefore have been quite different.

However, neither the Homer City nor TVA installations use the most advanced technology available. Froth flotation, for example, an effective method for cleaning coal fines, is not used in either case. It is not clear whether the advanced beneficiation techniques used in the metallurgical industry would be capable of meeting the stringent emission regulations for new power boilers and if so whether the approach would be cost effective compared with other methods. The fact that there has been no move in this direction indicates that it probably would not be.

One of the main problems in physical coal cleaning is that a high degree of sulphur removal requires a reduction in particle size to the point that a major portion of the coal particles contain very little inorganic sulphur. For most coals this is very difficult to accomplish. Inorganic sulphur (pyritic) is usually present as such small inclusions that an impracticable degree of grinding would be required to separate them as discrete particles; in addition, organic sulphur cannot be removed by physical cleaning techniques.

For new plants burning low-sulphur coal, physical coal cleaning is even less applicable. In the first place, much of the sulphur in low-sulphur coals is in the organic form and therefore not amenable to removal by physical methods. Secondly, for the 70% removal regulation now in effect in the U.S. for low-sulphur coal, the usual approach is

to use flue gas desulphurization on part of the gas and leave the rest of the gas untreated. The portion scrubbed is desulphurized to an advanced degree, 90% or more as is feasible in scrubbing, thus giving an average removal of 70% or more. By this means, the capital cost is reduced. In contrast, coal cleaning would probably have to be applied to the entire coal supply.

Existing Plants

In the United States, emission regulations for existing plants are set by the states under SIP's approved by EPA. In most cases, especially in areas where naturally occurring low-sulphur coal is used, no emission reduction has been required. Where reductions have been required they have usually been between 25% and 50%, in which case it has normally been more economical to change to a coal with lower natural sulphur content than to clean the coal or install scrubbers. The Tennessee Valley Authority, which is the main example in the U.S. of state regulations being applied at existing stations, has found shifting to lower sulphur coal the best alternative at four or more of its stations.

Another consideration for existing plants is the "bubble concept" recently approved by EPA. Under this control philosophy, an entire station would be considered as an entity in regard to emissions and the company operating it would be allowed to use the most cost-effective mix of controls in meeting the overall emission limit. In such cases, the best course would probably be to install flue gas desulphurization on part of the individual units, desulphurizing the gas to a high degree, and leave the other units untreated. A less efficient method such as physical coal cleaning would not fit in as well with this approach.

There have been proposals recently to clean all coal above a certain sulphur content to remove some 10-20% of the sulphur. A bill to that effect has been introduced in the legislature of one of the Ohio River Valley states. The emphasis is on reducing the long-range transport of pollutants by a low-cost method.

Combined Technologies

A strategy that has received considerable attention is the combination of cleaning coal to a limited degree followed by flue gas desulphurization. Some have claimed that this combination is more cost effective than flue gas desulphurization alone.

In recent cost studies by TVA(1), however, the overall operating cost was found to be generally higher for the combination than for FGD alone.

The TVA studies were based on an 85% removal requirement, the level considered likely when the study was initiated (before the EPA standards of June 1979 were promulgated). For the current 90% standard, the combination approach would be penalized even more; the general indication from estimates made by other organizations is that the combination becomes less attractive as the removal requirement increases.

For existing plants, subject to an intermediate degree of required emission reduction, the combination could be cost effective. No generalizations can be made because site-specific considerations become important in the intermediate control area. Moreover, existing stations in the U.S. have not been under much pressure to install expensive measures such as flue gas desulphurization. This situation may be changing, however, because of increasing emphasis on problems such as Prevention of Significant Deterioration (PSD), visibility, conversion from oil to coal, and long-range transport problems. An example of the type of situation that can develop is that of TVA at the Paradise station. The original strategy was to use coal cleaning alone to meet the state regulation, which at that time only required an emission reduction of about 25%. After the plans were well underway, however, the state changed the regulation to require over 50% removal, making it necessary for TVA to install scrubbers on over 50% of the capacity. Thus the situation has ended up, without being planned that way, as a combination of coal cleaning and FGD.

2.2.1.2 Reliability. For situations where physical coal cleaning is applicable, reliability should be acceptable -- mainly because the cleaned product can be stockpiled and thus any impact on the power plant of problems in the cleaning operation is avoided. The main question is whether the process can meet a given objective in regard to sulphur removal with changing coal characteristics. Coal is a substance of variable composition, difficult to predict in regard to distribution of sulphur compounds and resulting amenability to physical cleaning methods.

This is a question, for example, at Homer City. The Electric Power Research Institute (EPRI), EPA, and the U.S. Department of Energy (DOE) have a major test program underway aimed at evaluating factors affecting performance.

2.2.1.3 Cost. Physical coal cleaning is probably the most cost-effective method available for reducing SO₂ emissions if a high degree of reduction is not required. The

TVA study (1) shows a cost of \$0.22 per lb of sulphur removed for cleaning and \$0.237 per lb for limestone scrubbing (2 000 MW, 3.5% sulphur coal, 29-32% removal for cleaning and 85% for FGD). Within the limits of accuracy of the estimates, the costs are thus about the same. There are certain intangible benefits to cleaning, however, that are not counted in this comparison and which should make cleaning the clear choice if 29-32% removal is acceptable.

For lower-sulphur coals, the cost of cleaning goes up rapidly with a decrease in coal sulphur content. For example, at 0.7% sulphur the cost of sulphur removal is \$1.88 per lb compared with \$0.89 for FGD. (The latter is for 85% removal; at the 70% level under current regulations the cost should be even lower.)

For the combination case (also for 2 000 MW and 3.5% sulphur coal), the estimated cost was \$0.258 per lb of sulphur removed for 85% removal compared with \$0.237 for limestone scrubbing alone -- again essentially the same. However, several cost factors that favour the combination but are difficult to quantify were cited.

- Transportation cost. Leaving the ash at the cleaning plant gives some advantage in freight cost. The saving will vary widely and is offset to some extent by the higher moisture level in cleaned coal.
- Pulverization cost. With much of the mineral matter removed and a lower tonnage required, the coal-grinding cost at the power plant is lower.
- Boiler fouling. The partial removal of mineral matter in coal cleaning reduces slagging and corrosion problems in the boiler. This is a major advantage but difficult to quantify.
- Ash handling and disposal cost is reduced because there is less ash. (However, this is offset by more refuse handling required at the cleaning facility.)
- FGD operation is generally improved by reducing the inlet SO₂ concentration. It is not clear that this is intrinsic to the process but it has been generally true so far.

Although these are important considerations, there are also some drawbacks to the cleaning/FGD combination.

- Precipitator operation. Removing the sulphur from the coal makes the ash less conductive and therefore more difficult to collect in an electrostatic precipitator.
- Waste disposal. The refuse from coal-cleaning plants is often thixotropic and difficult to dry. Moreover, exposure to air and moisture converts the pyrite to dilute sulphuric acid that can leach out metals and cause a trace element

contamination problem. The materials used to promote separation of refuse (e.g., magnetite and chemicals) may also cause disposal difficulty.

- Handling problems. Cleaned coal has smaller particle size than ordinary steam coal and thus may be more difficult to handle when dry (as on the surface of storage piles) or very wet.
- Energy requirement. Although the differences in energy consumption are counted in cost estimates, such as those prepared by TVA, the higher energy loss for the cleaning/FGD combination is a disadvantage. For the TVA and Homer City projects, it is calculated that about 5% of the coal energy will be lost in the rejects from physical coal cleaning; for the more usual types of cleaning the loss is much higher. Further losses occur in thermal drying of the coal before shipment and in evaporating the surface moisture (picked up in transit) in burning the coal. (The surface moisture is higher than for normal coal because of the larger surface area.)

2.2.2 Chemical Desulphurization of Coal. Much experimental effort has been expended on methods for desulphurizing coal by chemical means. The processes vary widely, ranging from simple leaching by chemical solutions to methods that involve dissolution of the coal and reconstitution of the solids. The latter, generally called solvent-refined coal (SRC), borders on a coal conversion process and is usually classed as such. However, it is also a process for cleaning the coal of ash and sulphur and producing a clean solid fuel with characteristics much like the original coal.

The principal chemical-coal-cleaning (CCC) processes are as follows.

1. Kennecott. Oxidative leaching using oxygen and water at moderate temperature and pressure.
2. TRW. Oxidative leaching using ferric sulphate and oxygen in water.
3. KVB. Oxidation in NO₂-containing atmosphere; sulphates are washed out.
4. Institute of Gas Technology. Oxidative pretreatment followed by hydrodesulphurization at 800°C.
5. Jet Propulsion Laboratory. Chlorinolysis in organic solvent.
6. Battelle. Mixed alkali leaching.
7. General Electric. Microwave treatment of coal permeated with NaOH solution produces soluble sulphides.
8. DOE. Air oxidation and water leaching at high temperature and pressure.

9. Syracuse Research. Coal comminution by exposure to ammonia vapour; conventional physical cleaning then separates mineral matter.
10. Hazen Research. Pyrite is magnetized by treating dry, fine coal with $\text{Fe}(\text{CO})_5$; the pyrite is then separated magnetically.
11. SRC. Coal is dissolved in an organic solvent (produced in the process). The addition of hydrogen causes the sulphur to be evolved as H_2S , from which elemental sulphur is produced. The ash is separated by filtration, the solvent evaporated, and the coal resolidified by cooling.

Although much development work has been done on these methods, there has been no commercial application yet. The solvent-refined coal, sometimes called a synthetic fuel, is probably the closest to commercialization. One module of a commercial-size plant is to be funded by DOE, with final designs due by mid-1980 and startup planned for 1984. The developers plan to expand the facility to commercial size and have it operating by 1990; the capacity would be 30 000 tons per day (five 6 000-tpd modules).

All the other CCC methods listed above are only at the bench or pilot-plant scale of development.

2.2.2.1 Process evaluation. Chemical coal cleaning has the same problem as physical cleaning -- difficulty in getting a high degree of removal without high cost. Although most of the methods will do better than the physical cleaning processes in removing both pyritic and organic sulphur, especially the latter, overall removal is usually considered to be between 60% and 75%. The solvent-refined coal process does better than the others because the hydrogenation promotes sulphur removal; the process probably can achieve 85% removal at a cost competitive with wet scrubbing but the 90% or higher removal required under the current U.S. New Source Performance Standards (NSPS) is a difficult objective.

Thus, like physically cleaned coal, the chemically cleaned product may be more applicable to existing plants where more SO_2 can be emitted. There is some question, however, as to the effect of future regulations on the business risk in building CCC plants. To be economical, the cleaning plant must be very large and thus a heavy capital outlay is required (in contrast with physical cleaning plants). The current trends in SIP's, due to pressure from present and pending regulations on PSD, non-attainment, visibility, acid rain, and respirable particulates, are in the direction of tighter regulations

on existing plants -- with emission limits in the future that CCC may not be able to meet. Moreover, by the time a new technology such as SRC becomes commercially available, which may be 1990 at the earliest, many of the existing plants will have been replaced by new ones. This prospect is not attractive for new ventures requiring a large amount of capital.

One of the plus factors for chemically cleaned coal is that the load factor for existing plants declines with age, making the capital charges for flue gas scrubbing an increasingly expensive item. Buying clean coal from a central facility thus becomes more attractive.

2.2.2.2 Cost. Because of the chemical steps involved, chemical coal cleaning costs considerably more than physical cleaning. In the TVA study referred to earlier, the cost per lb of sulphur removed ranged from \$0.253 to \$0.44 for the KVB, TRW, and Kennecott processes (same conditions as before -- 2 000 MW, 3.5% sulphur coal). In contrast, the estimated cost for FGD, which was assumed to remove 85% of the SO₂ as compared with 59-73% for the CCC processes, was \$0.237 per lb of sulphur removed.

Various cost estimates have been published for solvent-refined coal, some optimistic and some less so. The Electric Power Research Institute estimates a cost of about \$4.50/10⁶ Btu for SRC, which corresponds to about \$113 per ton of eastern coal (at 12 500 Btu/lb). This is considerably higher than the levels estimated for the use of raw coal plus scrubbing, which were \$25-30 per ton for the coal and \$10-15 per ton for the scrubbing. However, SRC has several advantages such as low ash content that give other savings, thus complicating the cost comparison. At best, the process does not seem likely to be competitive with flue gas scrubbing at 90% and higher removal requirement.

The cost of a new process almost always increases as more is learned about it. Thus the new approach usually needs a significant advantage in initial cost estimates to have any chance of being competitive when the development is complete. None of the CCC processes, including SRC, seems to have such an initial advantage.

Adverse cost factors for CCC include disposal problems resulting from chemical residues and the acidic nature of the waste solids; corrosion at the high temperatures encountered in some processes; explosion hazard in the KVB method; and residual sodium in the coal (adverse to boiler operation) in processes that use caustic.

2.2.2.3 Reliability. The cost comparison between CCC and FGD varies considerably with how the reliability problem is handled. Chemical coal cleaning can be considered completely reliable to the power plant operator on the basis that the CCC plant will

maintain a stockpile of the product to assure an uninterrupted supply. For FGD, however, full reliability cannot be assumed, and has not been attained in most operating systems. Most cost estimates have ignored this but some have assessed a penalty on the basis that operating interruptions due to lack of reliability will incur a cost assignable to the FGD system -- either as a penalty assessed by the regulatory agency or a cost differential for power purchased to avoid violation of the standard. If a very high penalty is assessed for lack of reliability, then a CCC method such as solvent-refined coal could well be more cost effective. The question again is how to get the removal by the CCC approach up to the level required by the regulations without incurring inordinate cost. For FGD, reliability can be improved by spending more money for spare equipment, the cost for which can be estimated fairly well, but it is not clear what the cost would be for raising CCC efficiency to the 90% or higher level.

2.2.3 Desulphurization of Oil. Oil desulphurization is a well-developed, complex technology, used widely in several countries. Ando (2) has reported on the situation in Japan, where the method has been used extensively to reduce SO₂ emissions.

Most oil desulphurization processes involve the use of hydrogen and a catalyst to decompose complex sulphur compounds and evolve the sulphur as H₂S, which is recovered as elemental sulphur. Thermal decomposition is also used.

The residual sulphur in the cleaned oil is usually between 0.2% and 0.5% but a higher degree of desulphurization is feasible. The situation is similar to coal cleaning in that the cost increases with the degree of desulphurization. Ando shows an increase from \$16/kL to \$27/kL in going from 70% to 97% sulphur removal, compared with an equivalent increase from \$16/kL to \$19/kL for a similar improvement in efficiency for flue gas desulphurization.

Since new oil-fired boilers are not being built in the U.S. and are on the decline in Japan, the main role for desulphurized oil is for existing plants in congested areas (e.g., Tokyo and Los Angeles) where local regulations are stringent.

2.3 Coal Conversion

Although not aimed directly at SO₂ abatement, coal conversion must be considered in assessing the technology for controlling emissions from power stations. (The usual definition of coal conversion is production of clean gaseous or liquid fuels, which indicates that sulphur is removed in getting the "clean" product.) Much of the coal conversion technology is also not aimed at power production but rather at the

manufacture of pipeline gas, synthesis gas, liquid hydrocarbons (substitutes for their petroleum-derived counterparts), and chemicals. However, some of the methods have been proposed as a means of producing clean fuel for use in power plants. SRC-II, for example, the "liquid product" version of the solvent-refined coal process, would make a fuel suitable for direct use in oil-fired boilers. Coal gasification has been considered seriously for making a low-Btu gas suitable for firing in a standard boiler or in a combined-cycle power system (see Figure 2). Finally, if any of the coal liquefaction processes are successful and are adopted commercially, the product could be burned in oil-fired power plants as petroleum-based products are now.

Liquid fuels have been made from coal on a commercial basis by SASOL in South Africa but the different economic conditions in North America have so far prevented commercial production by the SASOL process. SRC-II may have a better chance.

The initial emphasis in coal gasification was on making gas for firing directly in a standard boiler; the sulphur (present after coal gasification as H_2S) would be removed from the gas by standard methods. The main effort was at Commonwealth Edison's Powerton station, where plans were developed for testing a Lurgi gasifier to produce gas for a 75-MW boiler. When detailed cost estimates were developed, however, the projected cost for large boiler retrofits was found to be nearly \$600/kW just to control sulphur. Estimates by TVA generally supported this high figure. The project was dropped.

Since the production of gas for use in combined-cycle operation appeared more attractive, development turned in that direction. The main test of this approach has been at STEAG's Kellerman station in Lunen, West Germany (started in 1972), where five Lurgi gasifiers were installed to produce gas for firing in a turbine-pressurized-boiler assembly (170 MW total). The test program was carried on for several years and much improvement was made. Several problems remain, however, including the need to agglomerate coal fines before gasification; dust removal at 600°C; high cost of H_2S removal (Stretford unit required in series with hot carbonate); and inadequate load following (poor bed temperature control during load increase). A pilot plant is under construction to test some new concepts for minimizing these problems; startup is planned for late 1980.

Several other projects, using other types of gasifiers, are underway in the U.S. The main ones are as follows.

- Texaco - Southern California Edison. The Cool Water combined-cycle power plant (100 MW) uses a Texaco coal gasification system. The plant is scheduled for completion in late 1983.
- Westinghouse. A 15-tpd pilot plant has been operating for some time.
- Central Power and Light (Texas). A 17-MW demonstration unit was started up in 1979, using the "chemically active fluidized bed" gasification concept developed in England by Esso Research. The system is designed for oil but is to be tested on coal. Another project to use the process is being negotiated, a 90-MW test program at American Electric Power's Cabin Creek station.

2.3.1 Applicability of the Technology. Unlike the fuel desulphurization processes, coal conversion is capable of attaining a high degree of sulphur removal. This is particularly true for coal gasification; sulphur compounds are converted to H_2S almost completely, which can be removed efficiently by well-developed commercial processes.

The main question is the general applicability of coal conversion to power production. For combined-cycle operation, the principal objective is not SO_2 control but rather an increase in energy efficiency, which proponents have claimed can amount to several percentage points. It remains to be seen whether such an improvement can be attained in practice in view of several unresolved problems. One is the need for a turbine that can operate at 2500-3000°F, much above the current level of about 2000°F. At the higher temperature, impurities in the gas become a serious problem because of turbine blade corrosion and erosion. Gas purity thus becomes less of an environmental consideration and more of an operating necessity.

At present, the best hope for near-term commercialization appears to be for coal-derived liquid fuels such as SRC-II as a substitute for petroleum-based products. At best, however, 10 years or so will probably elapse before such fuels are commercially available. By that time the oil supply-demand situation may well have changed in the direction of less need for liquid fuels. For example, the currently planned conversions from oil to coal in the U.S. may essentially eliminate any need for synthetic liquid fuels in the power industry.

For coal gasification, the prospect for near-term use is even more remote. Since combined-cycle operation seems to be the only practicable way to use gasification,

the power industry would have to make a major change in baseload technology for this method of SO₂ control to have much impact. This is not likely to happen soon. The STEAG test work was started in 1972 but the utility has gone back to the pilot plant studies to resolve problems. Assuming that the pilot-plant program is successful, another demonstration program would be needed. Meanwhile it would be necessary to develop higher temperature turbines for the combined-cycle system, even if it were successfully demonstrated. Finally, environmental restrictions will probably become more stringent in regard to the organic waste materials produced in the process. High removal efficiency for these is likely to be more difficult than for the pollutants in standard power plant operation.

Thus gasification appears to be 15 to 20 years away if it is ever adopted in power plants at all.

2.3.2 Cost Factors. Gasification is difficult to evaluate on a cost basis as an SO₂ control method because the main objective is better energy efficiency. Because of this, the estimate must cover the cost of operating the entire power plant rather than just the SO₂ control system. The numerous published estimates in which combined-cycle operation is compared with a conventional power plant fitted with FGD nearly all conclude that combined cycle is more cost effective. For example, a study by Fluor Engineers for EPRI (3) indicated a cost of \$0.0512/kWh for a conventional boiler with FGD and a range of \$0.0413/kWh to \$0.0514/kWh for various gasification/combined-cycle combinations.

The general power utility attitude seems to be that this remains to be proven. In most cases the costs of the demonstration units have been much higher than those projected on the basis of cost estimates.

Compared with other coal conversion processes, gasification has the disadvantage that it must be sized for the power-producing capacity with which it is associated. In contrast, coal liquefaction processes (or coal cleaning) can be built in an economically large size at a central location.

2.4 Desulphurization during Combustion. One of the earlier concepts in SO₂ emission control was the injection of an alkali into the boiler to react with the SO₂ to form an alkali-sulphate particulate that could be collected downstream and removed from the gas. The main advantage would be lower cost because the boiler serves as the reaction vessel, the gas is not cooled to the point that reheating is necessary to avoid

downstream condensation (as in wet scrubbing), and the only significant operating cost is that for the absorbent.

2.4.1 Standard Boilers. The early work concentrated on the injection of limestone to form calcium sulphate. Fairly good results were reported in Germany and Japan for tests in small boilers but when TVA tested the method on a large boiler in 1969-71 the performance was very poor. The main problem was getting the limestone distributed adequately over the boiler cross-section. (Injection with the coal was not feasible because the absorbent was "dead-burned", or inactivated, at the high temperature.) It was found that both SO_2 concentration and gas velocity varied over the cross-section, to the extent that it was virtually impossible to get the right amount of limestone to the right place at the right time so that adequate reaction with SO_2 would take place in the short time available (a second or two) before the gas cooled to a temperature at which the reaction became too slow. About the best that could be done was 40% removal at three times the stoichiometric (theoretical) amount of limestone. Moreover, the excess limestone (calcined to lime in the boiler) had a major adverse effect on dust collection efficiency in the precipitator.

There has been little activity in the injection of absorbents into standard boilers since the TVA tests. The injection of sodium and potassium carbonates has been proposed but boiler corrosion is a major problem with such materials. In recent tests by both EPA and the University of West Virginia, intimate mixing or pelletizing the limestone with the coal has shown some promise; presumably the SO_2 reacts with the limestone before it becomes inactive. It does not appear, however, that the method could give a very high degree of removal.

2.4.2 Fluidized Bed Combustion. The most promising development in the boiler injection area is fluidized bed combustion (FBC), in which the coal is burned in a bed of solids (limestone and ash) fluidized (suspended) by the upward flow of combustion air. Since the limestone particles are held in suspension for a relatively long time in the gas stream, both removal efficiency and limestone utilization are improved compared with injection into a standard boiler.

For electrical-power-plant application, the status of fluidized bed combustion is about the same as that for coal gasification. An intense developmental effort is underway in several countries but there is no commercial use yet. The main developmental efforts are as follows:

- Bergbau Forschung (West Germany). Basic laboratory work, a small pilot plant (0.25 MW), and an 8-MW pilot plant under construction.
- Ruhrkohle (West Germany). Several projects, including conversion of a grate-type boiler to FBC.
- Grimethorpe (England). A major project (about 30 MW) should be currently in the startup phases.
- Woodall-Duckham (England). A stoker-fired boiler in Renfrew, Scotland, was converted to FBC in 1975.
- EPRI-Babcock and Wilcox. A 2-MW (6 x 6 ft) combustor at Alliance, Ohio.
- DOE (Rivesville). A 30-MW demonstration unit, designed by Pope, Evans and Robbins and built by Foster-Wheeler, was installed at Rivesville, West Virginia, in 1976. There have been severe operating problems.
- TVA. A 20-MW test unit is planned.

Several other test facilities are in preliminary planning stages in both the U.S. and West Germany.

2.4.2.1 Process evaluation. The main advantages claimed for fluidized bed combustion are as follows.

1. The boiler can be smaller because of the better heat transfer from the burning coal to steam tubes submerged in the fluidized bed.
2. There is less boiler fouling and corrosion because the ash does not melt as in a conventional boiler. The temperature (1500-1600°F, 815-870°C) is below the melting point of the ash, whereas in a conventional pulverized fuel boiler the temperature is about 3000°F (1650°C) and much of the ash melts.
3. Low grades of coal and other solid fuels (high ash and moisture) can be burned without difficulty whereas in standard boilers such fuels cause severe operating problems.
4. Injection of limestone removes SO₂ without the need for an additional reactor or scrubber and without the need for reheat.
5. The low operating temperature reduces NO_x formation.
6. By operating the fluidized boiler under pressure, an energy saving can be attained by using a turbine on the exit gas.

These are impressive advantages but there are some offsetting problems that are as yet unresolved.

1. It is difficult to get as complete combustion as in conventional boilers. Studies in England indicate about 97.5% combustion efficiency as compared with 99% for standard operation.
2. It is also difficult to get as high a steam temperature as in conventional firing; any reduction in temperature would have an adverse effect on energy efficiency.
3. The limestone requirement is much higher than in wet scrubbing. To get 90% SO₂ removal, some 3-6 times the stoichiometric amount of limestone is required, compared with 1.05-1.1 times the stoichiometric amount in the scrubbing process. As a result, the amount of waste solids is increased. For high-sulphur coal, the tonnage of limestone required could be over half that of the coal.
4. SO₂ removal efficiency varies with the type of coal and limestone. In English work, for example, one limestone gave 90% SO₂ retention at a stoichiometric ratio of 2.25 but another limestone type required 5.25. For the latter, the limestone tonnage was 70% of that for the coal.
5. The excess lime carried downstream has an adverse effect on dust collection in precipitators because the SO₃, which otherwise would make the dust particles conductive, is removed by the limestone. This could be a severe cost penalty for high-sulphur coal since the precipitator would have to be much larger than for conventional boilers. A baghouse could be used but would also incur a cost penalty if used with high-sulphur coal.
6. Cyclones are used for collecting and recycling most of the solids elutriated from the bed; the material is returned to a carbon "burn-out" cell. Cyclones were used in conventional systems to recover dust but were generally abandoned because of high maintenance requirements.
7. The waste product is a mixture of ash, gypsum, lime, and unreacted limestone. Dry handling is recommended (4) but is expensive. Utilities normally wet the ash before disposal; fugitive dust is a major problem when handled dry.
8. Many utilities sell ash, which may not be feasible if mixed with lime and gypsum.

2.4.2.2 Cost factors. Like coal gasification, most estimates for FBC show a cost 10-15% lower than a conventional system fitted with FGD. Since most of these estimates have been published by proponents of the method, the perspective may be optimistic. In

one of the better cost comparisons (4), prepared for EPA by TVA, it was concluded that although FBC (atmospheric and pressurized operation) shows a potential saving of 9-14%, "when uncertainties are included, the estimated cost of electricity for the three alternatives is so close that all are considered to be within the competitive range for further consideration."

The FBC approach was favoured by some of the assumptions in the TVA study, mainly the higher energy efficiency for FBC and the relatively high energy penalties assigned to a conventional system plus FGD. For example, it was assumed that atmospheric FBC has an energy efficiency of 35.8% compared with 31.8% for conventional boilers. In contrast, the English study mentioned earlier shows 36.6% for FBC and 37.1% for conventional boilers.

It must be concluded that the comparative cost of FBC and conventional systems cannot be calculated at the present state-of-the-art. A relatively minor change in assumptions can throw the conclusion either way.

2.4.2.3 Status of the technology. Various predictions have been published as to when fluidized bed combustion will be commercially available, ranging from the late 1980s to the late 1990s. In view of the still unresolved problems and the usual slow pace of commercial acceptance, the latter is probably a better prediction.

Although the method has some outstanding potential advantages, it has the problem, like coal cleaning, that regulatory trends are against it. Increasing environmental pressure will produce more stringent emission standards, requiring more limestone, or other expensive measures, if FBC is the method used for SO₂ control. In contrast, wet scrubbing has the advantage of being able to attain very high removal efficiencies at lower incremental cost. The last few percentage points of SO₂ removal are much more easily accomplished when the absorbent is in solution compared with the solid state, where the SO₂ has to diffuse into the pores of the solid.

As with coal gasification, FBC also has the problem that as the technology matures, more problems will be encountered and the cost therefore will probably increase compared with current estimates. This was the situation for FGD, for which early estimates were much lower than current prices (after allowance for inflation).

Nevertheless, FBC has some important intrinsic advantages, one of which is the ability to burn low-grade fuel. This could be the main role for the process in the future.

2.5 Flue Gas Desulphurization -- Dry Processes

Considerable research has been aimed at dry absorption -- injecting an absorbent into the combustion gas and collecting the resulting SO_2 reaction product in dry particulate collection equipment downstream. Most of these processes involve the collection of fly ash and the SO_2 product together in a precipitator or fabric filter -- an arrangement that makes SO_2 removal relatively inexpensive because the dust collector is needed anyway for the fly ash.

The main problem is the difficulty in getting a high degree of SO_2 removal without having to use so much absorbent that the cost becomes unattractive. The only absorbents that have shown any promise are sodium bicarbonate and ammonia.

2.5.1 Sodium Bicarbonate. Sodium bicarbonate has the advantage that evolution of CO_2 and H_2O resulting from injection at relatively high temperature provides a high degree of porosity. The porosity makes the material a much more effective absorbent than soda ash (Na_2CO_3), a chemical that is more readily available commercially than the bicarbonate.

The only economical source of the bicarbonate appears to be nahcolite, a naturally occurring form of sodium bicarbonate. Unfortunately, nahcolite is relatively rare in comparison with Na_2CO_3 . The main nahcolite mineral deposits are in the western part of the U.S., for example, in Colorado. Use of the material is therefore limited to areas that are close enough to the deposits to avoid high shipping costs.

Another problem is limited removal efficiency. Although nahcolite is a reactive material, removal of more than 90% of the SO_2 is difficult. The 70% removal allowed under current federal U.S. regulations for low-sulphur coal can probably be attained; state regulations, however, may require a higher removal efficiency.

The availability of nahcolite is also a problem. It is not being mined currently and will probably not become commercially available unless the regulatory situation makes it more attractive. This does not seem likely.

2.5.2 Ammonia. Of the various absorbents, ammonia is easiest to use because it can be handled and injected as a gas. Unfortunately, the main reaction product, ammonium sulphite ($(\text{NH}_4)_2 \text{SO}_3$), is relatively unstable and will not form until the temperature has been reduced to a level at which a solid product with low vapour pressure precipitates

from the gas. This may be too low a temperature for the operation of particulate collection equipment such as precipitators and bag filters.

The use of ammonia as the absorbent in a dry type of FGD process has not been studied extensively. The most promising results have been obtained in a pilot-plant study at the University of Tennessee (5), using a bag filter to collect the reaction product and the ash. By injecting the ammonia and then cooling the gas to about 180°F (82°C) before the bag filter, an SO₂ removal efficiency of over 90% was attained. These were limited tests, however, carried out with only one type of coal and with little exploration of operating variables.

Disposal of the reaction product would be a problem in the use of ammonia (because of the high solubility), but this is true for any alkali injected into the gas.

2.6 Flue Gas Desulphurization -- Semi-Dry Processes

Because of the problems in getting high removal with dry absorbents injected directly into the gas, a process has been developed in which the material is injected as a solution or slurry into a spray drier through which the gas passes and serves as the drying medium. Since the absorbent is in contact with the gas as a solution until the spray droplets evaporate, the absorption efficiency is much higher than for dry injection. The effect is much like that in a spray scrubber except that in the latter the spray droplets do not evaporate and therefore there is a longer time of contact with the gas.

Because of the limited contact time, a reactive type of absorbent is required. Sodium carbonate (Na₂CO₃) and lime have been the main choices; limestone is not considered reactive enough for use in the process.

The semi-dry processes are a relatively recent development; the first pilot-plant work of any consequence in the U.S. was done only two or three years ago. (There was earlier work in Japan but the process never gained wide acceptance.) Since then there has been considerable enthusiasm for the method and several full-scale installations have been contracted by utilities on the basis of the pilot-plant data. None of these is yet in operation.

The main projects, both full-scale and developmental, are as follows.

- Otter Tail Power Company (Coyote station, North Dakota). New boiler (unit 1), 440 MW. Startup planned for 1981. Soda ash is the absorbent; the designer is Rockwell (with Wheelabrator-Frye).

- Basin Electric Power Cooperative (Antelope Valley 1 and 2, North Dakota). New boilers, 440 MW each. Startup planned for 1981 and 1983. Lime-based system supplied by Joy-Niro.
- Basin Electric Power Cooperative (Laramie River 3, Wyoming). New boiler, 600 MW. Startup planned for 1981. Lime-based system supplied by Babcock and Wilcox.
- Tucson Gas and Electric (Springerville 1 and 2, Arizona). New boilers, 350 MW each. Startup in 1984-5. Lime-based system supplied by Joy-Niro.
- United Power Association (Stanton, North Dakota). New boiler, 63 MW. Lime-based system supplied by Research-Cottrell.
- Babcock and Wilcox. 20-30 MW test facility operating at Pacific Power and Light's Jim Bridger station in Wyoming. Design changes have been necessary.
- Rockwell-Wheelabrator Frye. 25-MW test facility started up recently at a Celanese plant in Cumberland, Maryland.
- Joy-Niro. Full-scale test module (100 MW) under construction at Northern States Power Company's Riverside station in Minnesota. Startup planned for late 1981.
- Mikropul. Small unit installed at a plant of the Hammermill Paper Company. Severe operating problems have been encountered.
- Other. Several other companies, including Combustion Engineering, Research-Cottrell, and Envirotech, have pilot plants in operation or planned.

2.6.1 Process Evaluation. Spray drier scrubbing has several advantages, some obvious and some claimed by proponents.

- Process simplicity. There is no need (as in wet scrubbing) for large slurry pumps, mist eliminators, reheaters, and sludge dewatering equipment.
- Lower cost. Proponents claim that less operating and maintenance labour will be required and that equipment can be constructed of unlined carbon steel.
- Lower energy requirement, said to be only 25-50% of that for wet scrubbing.
- Lower water consumption, said to be about 50% of that for wet scrubbing.
- No scaling. The reaction product is dry before it reaches the wall of the spray drier.
- Better reliability. The relative simplicity is claimed to give higher reliability.

A detailed analysis of these advantages indicates that although there are some significant advantages for the semi-dry route, they are not as overwhelming as proponents claim and they may be offset by some of the disadvantages. The main difficulty, as is the

situation for several of the other processes that compete with wet scrubbing, is in getting the high degree of removal required by current regulations. The limited contact time between the gas and the absorbent, before the latter becomes too dry to be efficient, makes it difficult to attain the high removal (as high as 98-99%) feasible in wet scrubbing -- unless a large excess of absorbent is used. There are also several other drawbacks.

- Process control. The amount of water injected must be limited to that which the available heat in the gas will evaporate. Otherwise some would remain unevaporated and the system would become "wet" with all the resulting disadvantages. Moreover, to be safe, it is desirable to use somewhat less water than that required to saturate the gas and cool it to the water dewpoint, which requires additional control compared with wet scrubbing. The usual practice is to control the water addition on the basis of the outlet temperature, with the latter held at a safe level by cutting back on the water if the temperature drops too low. Lime addition must also be controlled but pH cannot be used as in a wet system. The usual control is by a combination of inlet SO₂ concentration, inlet gas flow, and outlet SO₂ concentration -- a somewhat unreliable control system because SO₂ is difficult to measure continuously.
- Absorption efficiency can be improved by using more water, that is, by running very close to the water dewpoint. The difficulty is that condensation may take place in the dust collector; some designers avoid this by bypassing a small amount of gas around the spray drier to raise the temperature of the gas entering the dust collector. The gas is usually taken from before the air heater to get more heat with less gas since only a small amount of gas can be bypassed if high removal efficiency is required. Bypassing gas from before the air heater reduces overall energy efficiency.
- Low-load problems. At reduced load, less heat is available but regulations require the same removal efficiency as at full load. Since less water can be used, low load requires either more gas bypassing or use of excess lime to maintain removal efficiency.
- Cost of absorbent. The cost of lime for the semi-dry process is usually much higher than for the limestone generally used in wet scrubbing.
- High-sulphur coal. The process is much more applicable to low-sulphur coal. For high-sulphur coal, the ratio of lime to water becomes so high that the feed slurry

may be too thick to feed properly. The higher amount of mass transfer required makes it more difficult to attain high removal efficiency, and the absorbent cost differential becomes more important because of the larger lime requirement.

2.6.2 Cost. Published estimates indicate a lower cost for the spray drier process, about 15%. Basin Electric, for example, estimated the capital cost at Antelope Valley to be \$129/kW (including particulate removal) for a dry system compared with \$145/kW for wet scrubbing (limestone). At Laramie River, the estimates were \$100/kW and \$121/kW respectively.

The power companies that have bought dry systems apparently assumed that the dry route was better and did not take bids on wet processes, a good way of finding out what systems really cost. Other utilities have taken bids on both and ended up buying the wet system. In one instance, the dry-process bid was considerably higher than that for wet scrubbing.

Thus the capital-cost status of dry processes is quite uncertain. Because of the relative simplicity there should be some cost advantage but this has not yet been substantiated in competitive bids.

One of the cost considerations is that where the removal requirement is relatively low, e.g., the 70% under the federal standard for low-sulphur coal, a considerable saving can be attained in wet scrubbing by scrubbing only part of the gas and bypassing the rest. For example, if the scrubbers are designed to remove 95% of the SO₂ then only about 74% of the gas needs to be scrubbed to achieve the 70% requirement. The resulting saving gives a considerable advantage compared with dry processes, for which high removal is more difficult and bypassing is not as feasible.

The operating cost depends mainly on what is assumed for operating labour and maintenance, plus the amount of lime required and the price margin over limestone. TVA estimates show lower direct costs (including absorbent) for wet scrubbing, but when items such as overheads and capital charges which depend on capital cost are added, the cost becomes 4.68 mills/kWh for the dry process and 5.47 mills/kWh for the wet process.

2.6.3 Status. The rush to adopt dry processes seems to have slowed. It is perhaps significant that none of the utilities buying the dry process, without taking bids on wet processes, has had any experience in FGD. Moreover, all those bought so far are in western areas of the U.S. where the sulphur content of coal is quite low and the cost differential between lime and limestone is not very great.

Other utilities planning to use low-sulphur coal have gone to the other extreme -- taking bids on wet scrubbing but not on dry processes. This confusing situation will presumably settle down soon.

Future use of the dry method will depend to some extent on reliability. If the claims of proponents as to higher reliability than for wet scrubbing are borne out, a major advantage for the process would be realized. This will not be known, however, until full-scale systems have operated long enough to establish performance.

Suppliers of dry systems predict widespread adoption, even for high-sulphur coal. This seems unlikely. Although the dry process will find a place, it will probably be limited to low-sulphur coal and to site-specific conditions unsuitable to wet scrubbing.

2.7 Flue Gas Desulphurization -- Wet Scrubbing

The oldest and most used of the FGD processes is wet scrubbing. It has had a history of failures and few examples of successful development. The main problems have been the complexity and size of the equipment required to scrub some 125 000 tons of gas per day (for a 1 000-MW power boiler) and to pump the 200 000 gallons of slurry per minute through the scrubbers.

A variety of absorbents have been tested and some used commercially, including water (actually weak sulphuric acid at steady state), lime or limestone, magnesia, and alkali compounds such as sodium and ammonium sulphites. The technology is divided into two classifications -- throwaway and recovery -- on the basis of whether the reaction product is discarded or recovered as a useful and saleable material. Of the two, throwaway is by far the more popular, mainly because it is simpler, usually more economical, and does not put the power company in the business of selling a chemical product.

There has been little commercial use of any absorbent other than lime or limestone, which are usually classed together since there is little difference between them in regard to the equipment used and the waste material produced. The choice is usually based on absorbent cost; limestone is currently favoured because of ready availability, relatively low cost (unless a long shipping distance is involved), and the energy cost in converting limestone to lime. Lime has some operating advantages, however, that have made it the choice in some situations.

The main disadvantage of lime and limestone scrubbing is the low solubility of both absorbent and product, resulting in a slurry in the scrubber circulation loop that

produces some operating problems. An alternative process, called double alkali, has been developed in which the absorbent is a soluble material such as sodium sulphite and the scrubber effluent solution is regenerated by treating with lime. The final product is the same as for lime scrubbing. The main advantage of the process is use of a clear solution in the scrubber.

The main emphasis in this report is on limestone scrubbing because of its current pre-eminent position. Lime scrubbing is discussed along with limestone because of the many similarities.

2.7.1 Lime-Limestone. Tests of lime-limestone scrubbing were carried out in England as early as 1925 and a full-scale scrubbing system was installed in the 1925-35 period. Since the once-through operation (scrubber effluent discharged to the Thames River) was an undesirable feature, a closed loop process (no liquid effluent) was developed in the 1930s and was installed on a small boiler at a power station. There was little activity thereafter in scrubber development; the utility (CEGB) turned instead to the tall stack method of controlling ambient SO₂ concentration.

In Japan, development work began in the late 1960s, leading to the installation of numerous scrubbing systems. The total equivalent capacity is about 35 000 MW, about half of which is on utility boilers. The capacity is divided about 50:50 between lime and limestone but the more recent installations have mostly used limestone. Currently there is little activity in wet scrubbing, mainly because low-sulphur oil is considered a better approach. It is expected, however, that there will be a trend to coal as the boiler fuel and that the relatively high sulphur content will make more scrubbing necessary.

In West Germany, activity began only a few years ago with passage of a federal regulation similar to the U.S. Clean Air Act. Two systems are operating and five are under design or construction. All use lime since the cost is not much higher than for limestone. The high cost and low quality of the coal makes the future of scrubbing in Germany uncertain. There is little activity elsewhere in Europe.

In the U.S., there was considerable small-scale research and pilot-plant development beginning in the early 1940s. In 1968, full-scale lime systems (with limestone injected into the boiler to get "free" calcination to lime) were installed by Union Electric (Missouri) and by Kansas City Power and Light. Both the limestone injection and the scrubber type were found to be unsatisfactory and were abandoned.

In the 1970-75 period, several scrubber systems were installed by power companies such as Commonwealth Edison (Illinois), Arizona Public Service, Nevada Power, Kansas City Power and Light, Louisville Gas and Electric, Duquesne Light (Pennsylvania), Kentucky Utilities, and Montana Power. The total capacity in 1975 was about 3 000 MW. Since then the capacity has mushroomed, with over 19 000 MW now operational (56 units), 17 000 MW under construction (42 units), and 36 000 MW planned (63 units). (This is for total wet scrubbing capacity, of which about 90% is lime-limestone. Thus for lime-limestone the levels are 17 000 MW, 15 000 MW, and 32 000 MW respectively.)

One of the reasons for the rapid growth was the Clean Air Act Amendments of 1977, which in effect require SO₂ control on all new boilers.

Based on recent estimates of coal-fired capacity in the U.S. over the next 20 years, and assuming that all new capacity will be equipped with SO₂ controls (90% of which is lime-limestone scrubbing), it is estimated that installed capacity for lime-limestone will be 93 000 MW by 1985 and 158 000 MW by 1990. This does not include the retrofitting of existing units that may be required over the next 10 years.

2.7.1.1 Choice of absorbent. Limestone is usually chosen over lime because of the lower cost. Even a small cost differential per ton becomes significant when the absorbent cost is extrapolated over a 30-year system life, the usual practice by power companies in evaluating bids for FGD systems on new boilers. Limestone usually costs \$5-10 per ton delivered to the site whereas typical prices for lime are \$45-60 per ton. (Two tons of limestone are roughly equivalent to one ton of lime.)

Lime has some advantages in both design and operation. The pumping load can be smaller, scaling problems are usually less severe, and system reliability is probably a little higher. These have not been enough to offset the higher cost in most of the recent process selections.

2.7.1.2 Absorption efficiency. The main problem in choosing limestone as the absorbent is its relatively low reactivity and the resulting difficulty in getting the high removal efficiency required by the regulations. This has been complicated by the new regulations in the U.S., which require 90% SO₂ removal on a 30-day rolling average basis (for each day the average for the preceding 30 days (including the day in question) must not be below 90%). There is no experience with this regulation yet but it is considered that the limestone system should have a capability of more than 90% so as to produce some "good days" to offset the bad ones that are likely to be encountered because of

equipment breakdown, non-delivery of limestone, and the many other problems that can decrease the average below 90%.

Limestone scrubbing capability can be increased in several ways, including higher L/G ratios (gallons of slurry recirculated per 1 000 cu ft of gas), use of elements in the scrubber to improve mass transfer, and addition to the scrubber slurry of mass transfer promoters such as magnesium sulphate or organic acids. The last of these is especially promising. In tests at the EPA-TVA test facility (two 10-MW scrubbers at TVA's Shawnee station), a small amount of adipic acid (about 1 500 parts per million) improved removal with limestone to a level over 95%. Magnesium sulphate was also quite effective.

Suppliers are now specifying high L/G ratios (70 to 90 level) in their bids and some have also adopted magnesium sulphate addition as a way to get high efficiency with limestone. None has used adipic acid, however, which may be the best approach. Further testing is needed to evaluate the material better.

One of the problems is that there are no data on the full-scale use of high L/G ratios or promoters. Systems now under construction must come on line before it will be known whether the new design parameters will promote enough limestone efficiency to meet the new regulations.

This mainly applies to high-sulphur coal. Low-sulphur coal is less of a problem; some of the systems now operating achieve more than 90% SO₂ removal with no difficulty. The main concern, for both high- and low-sulphur coal, is that continuous compliance with a 90% rolling average has never been attempted before. Under the 1970 Amendments, it was merely necessary to "qualify" the scrubber system by a short test; continuous compliance thereafter was seldom if ever attained and was not enforced by EPA. The rule under the 1977 Amendments is more stringent, requiring a report to EPA each day of the average for the preceding 30 days.

The SO₂ removal efficiency can also be increased by raising the slurry pH, at the expense of using more limestone. Limestone utilization is better at low pH but SO₂ removal drops off. Thus the utilization attainable depends on the mass transfer capability of the scrubbers, which in turn depends on several factors such as L/G ratio, gas velocity, size of slurry droplets, and scrubber internals. Suppliers are currently guaranteeing 1.05-1.15 moles of limestone per mole of SO₂ removed, or a utilization of 95-87%.

One of the lesser known areas in limestone scrubbing technology is the effect of limestone type on removal efficiency. Small-scale tests have indicated a fourfold

range in reactivity, a serious consideration when a scrubbing system is bought before the limestone supply is finalized -- as is often the case in bidding situations. The main research agencies in the U.S. (TVA, EPA, EPRI, DOE) are attempting to get more data on the problem.

2.7.1.3 System reliability. The most perplexing question in FGD is that of reliability. From the power company standpoint, it would be highly desirable for the scrubber system to be 100% reliable so that no interference with power production would occur. This is unlikely, however, because the boiler train proper has an average reliability of only 70-80%. An EPA study concluded that an FGD scrubbing system, without spare scrubbers, would have a reliability considerably lower than this.

Spare scrubbers have not been installed in many of the currently operating systems, mainly because the regulatory structure did not require a high degree of reliability. With the new regulations, however, any unreliability (defined as failure to comply with the regulations) presumably will result either in a fine or in a forced reduction in power production. As a result, almost all the recent specifications for FGD systems have included a provision that a spare scrubber train be provided -- thereby increasing reliability by redundancy.

It is not clear whether a spare scrubber will eliminate interference with power production. There is some indication it will because at Northern States' Sherburne County station -- one of the few FGD systems with a full spare scrubber -- 100% "operability" has been reported several times (reporting not consistent or complete).

Several definitions of scrubber performance have been advanced, including availability, operability, reliability, and utilization. All are confusing because none gives a measure of interference with power production, avoidance of which is the main objective of reliable operation. For Sherburne County as an example, the 100% "operability" (defined as hours the FGD system was operated divided by boiler operating hours) indicates only that the FGD system operated all the time the boiler operated. It does not indicate whether there was adequate operation of the FGD system, whether there was interference with power production, or whether there was compliance with the regulatory standard.

The current rule for 30-day averaging has put reliability in a different light because it allows some days of operation below the emission standard as long as the 30-day average is in compliance. In contrast, in the past a day of below-standard

performance was a violation. The longer averaging time should smooth out the performance curve and make 100% reliability (i.e., continuing compliance with the 30-day average) easier to attain. This will not be known until a system has operated under the current regulations.

There are many reasons for unreliability, including corrosion, erosion, gas maldistribution, wet/dry interface deposition, scaling, nozzle plugging, mist eliminator blockage, poor process control, failure of continuous monitors, failure of linings, mechanical breakdowns, damper maloperation, and non-delivery of lime or limestone. As a result of these, design has become complicated. Progress is being made but it is not yet clear whether a high degree of reliability can be attained even with redundancy, particularly with high-sulphur coal.

2.7.1.4 Sludge disposal. Lime-limestone scrubbing produces waste solids (mainly calcium sulphite) with undesirable properties -- difficult to dewater and incapable of supporting much weight when placed in the waste disposal area. Moreover, potential leaching of constituents is regarded by environmental agencies as a serious problem.

The dewatering and strength difficulties can be remedied to a considerable extent by forced oxidation -- bubbling air through the scrubber slurry to oxidize calcium sulphite to calcium sulphate (gypsum), a material that precipitates as large crystals that are easier to dewater and stronger when placed in a waste pond or landfill. There is a current trend to specify forced oxidation when purchasing scrubber systems.

The leaching problem is vague and ill-defined. Calcium sulphite and sulphate are relatively innocuous but regulatory authorities express concern about the leaching of certain elements (selenium, arsenic, mercury, and others) from the residual fly ash collected in the scrubber and present in the sludge. Thus the problem is more connected with ash disposal than with SO₂ control.

Regulations in the U.S. for solid waste disposal under the Resource Conservation and Recovery Act (RCRA) are to be finalized soon. There has been some concern that scrubber sludge will be classified as a hazardous waste under these regulations, a classification that would probably increase sludge disposal cost and perhaps make lime-limestone scrubbing so expensive that other processes would be favoured. This seems unlikely but is a possibility.

2.7.1.5 Water balance. Regulatory considerations require that scrubbing systems be operated in a closed loop fashion, that is, with no discharge of liquid effluent to the

environment. Thus all materials entering the system, both soluble and insoluble, must end up in the sludge. Depending on the degree of dewatering, there may not be much liquid phase left with the sludge to hold the soluble impurities in solution.

This has an adverse effect on the capability of the scrubber system to accept waste water from other power plant operations as makeup water, a helpful aspect of scrubber operation in solving the overall waste water problems of the power system. Since the scrubber evaporates about half a ton of water per ton of coal burned, using waste water as makeup can be a significant contribution.

The usual waste water sent to the scrubbers is cooling tower blowdown, which has a fairly high concentration of sulphate and chloride. Evaporation in the scrubber concentrates these further, to the extent that when the amount of purged liquor is small, the concentration can reach several thousand parts per million. As a result, the sulphate can cause scaling and the chloride corrosion.

It is not clear how much of a problem this will be. Previously lime-limestone scrubbing was the only FGD process capable of using waste water from other sources and thus could be assigned a credit. The spray drier approach now offers the same advantage but without the problems of scaling and corrosion, which could make spray drying the favoured process in some instances.

2.7.1.6 Cost. The costs associated with lime-limestone scrubbing have been covered partially by previous comparisons in this report. Since it is the basic process, evaluations of other approaches almost always include an estimate of lime-limestone as the baseline for comparison.

The best data on costs come from current bidding situations because estimating uncertainties is eliminated. The main difficulty is that bids cover only the equipment supplied by the vendor (usually on an erected basis), whereas the total cost includes various owner costs such as overheads, erection distributables, foundations, large motors, power and water supply, sludge disposal, interest during construction, working capital, and startup costs. Some of these are often omitted in cost estimating and are difficult to pull out of accounting statements even in actual construction situations. As a result it is difficult to determine what a scrubber system really costs.

Another difficulty is that bids vary widely for no apparent reason. In recent purchasing situations, the bids ranged from \$30/kW to \$45/kW (erected, 400-800-MW range, high-sulphur coal). Assuming that owner costs increase this two to threefold, the

resulting total cost is \$60-135/kW. This is about the best that can be done in specifying costs. Bid variations and site-specific considerations prevent narrowing the range very much.

Of the published estimates, those prepared by TVA are probably the best. In a recent estimate the total capital cost for limestone scrubbing was \$98/kW, about the midpoint of the range given above. The operating cost was estimated at 4.11 mills/kWh, or \$9.58 per ton of coal burned.

Some estimators increase the cost by including items for power demand penalty and extra costs for purchased power due to low reliability of the FGD system, a procedure that can produce a very high estimate.

2.7.1.7 Status. Limestone scrubbing is likely to remain the basic FGD process for a long time. It is relatively simple, energy efficient, the limestone is cheaper than other absorbents, and the capital cost seems to be as low as, or lower than, competing processes.

There is nothing on the horizon likely to remove these advantages, except perhaps increasing stringency of the rules on waste solids disposal and the resulting increase in cost. The effect of this will not be known until the rules are promulgated.

2.7.2 Double Alkali. The main advantage of double alkali processes is that a clear solution is used in the scrubber and the problems in scrubbing with a slurry are thereby avoided. Development work on the process has been mainly in Japan with a limited amount in the U.S.

Although double alkali (or dual alkali) is the term normally used to identify this method, a more descriptive one is indirect lime-limestone, because some processes using a clear scrubber solution followed by regeneration with lime or limestone do not use an alkali in the scrubber. In Japan, for example, the Chiyoda process uses weak sulphuric acid and the Dowa process uses a solution of basic aluminum sulphate.

Japan has over 50 commercial installations based on indirect lime-limestone, of which eight are on utility boilers with a total capacity of about 2 400 MW. The rest of the installations are mainly on industrial boilers and sintering plants with a gas capacity totalling about 2 700 MW. Eight different processes are represented, most of them based on use of limestone as the regenerant.

In the U.S., there is only limited commercial use. Two installations have been made by utilities and a third has been contracted, totalling about 1 700 MW. In addition a

275-MW test unit has been installed at the Cane Run station of Louisville Gas and Electric, partly funded by EPA.

All the U.S. installations use sodium sulphite as the absorbent and lime as the regenerant. Although limestone would be cheaper, it reacts so slowly with the scrubber solution that lime is considered preferable. Development work is underway to improve limestone reactivity. Limestone is widely used as the regenerant in Japan but it has not been given much consideration in the U.S.

The capital cost for indirect lime processes appears to be about the same as for direct lime-limestone scrubbing. Although two steps are involved, the regeneration vessel takes the place of the reaction tank in direct scrubbing and the lower scrubber cost tends to offset the cost of soda ash supply facilities. In recent bidding situations, the indirect process (double alkali) has been somewhere near the midpoint of the bidding range.

The situation is not as favourable for operating costs. Lime is normally considerably more expensive than limestone and there is some net loss of sodium, making the raw material cost quite high compared with limestone scrubbing. Proponents claim cost advantages for lower maintenance, higher SO₂ removal capability, and better reliability. Although such claims are probably justified, at least to some extent, they have not been demonstrated in full-scale installations and have not been enough to offset high raw material costs in process selection situations.

Another drawback is that forced oxidation to improve sludge properties is not feasible in double alkali operation of the U.S. type.

The future of indirect lime-limestone processes is not clear. If the current test work shows that limestone is usable, the position of the method will be improved. Moreover, if direct limestone scrubbing proves to be incapable of meeting the current regulations, either because of difficulty in attaining the removal efficiency required or because of low reliability, then double alkali could move in to fill the gap as the only qualifying process available. The situation will not be known until limestone systems that must comply with the current regulations are operated, which will be at least three years.

2.8 Flue Gas Desulphurization -- Recovery Processes

Recovery of the SO₂ in power plant flue gases as a useful material has been a research goal for several decades. Numerous companies have seen this as a promising business venture and have expended large sums on development. Various agencies and

institutes have also taken part, including TVA, EPA, DOE, and EPRI in the U.S., Bergbau Forschung in Germany, and several groups in Japan.

The results of this work have not been promising. In Japan, there are only four installations on utility boilers, totalling just over 500 MW. There are also about 25 units on industrial boilers and other industrial operations with a total gas flow of about 2 500 MW. In the U.S., two utilities have installed recovery processes on a commercial basis; the total capacity is about 2 500 MW.

2.8.1 Process Description. There are dozens of recovery processes, in various stages of development. The more significant ones are as follows.

- Wellman-Lord. The gas is scrubbed with sodium sulphite solution and the resulting sodium sulphite-bisulphite is heated to evolve a rich stream of SO_2 , convertible to either sulphuric acid or elemental sulphur. The process is used by New Mexico Public Service in the U.S. and by Chubu Electric in Japan.
- Magnesia scrubbing (Chemico, United Engineers). The gas is scrubbed with MgSO_3 slurry to form $\text{Mg}(\text{HSO}_3)_2$, which is then treated with MgO to precipitate MgSO_3 . The sulphite is dried, calcined to evolve a rich stream of SO_2 , and the SO_2 is converted to sulphuric acid. Philadelphia Electric is installing the process at two stations and TVA plans an installation at the Johnsonville station.
- Carbon adsorption (Bergbau Forschung, Sumitomo). The SO_2 is adsorbed in activated carbon at relatively low temperature. The sulphuric acid formed in the pores can either be washed out or the carbon heated to reduce the acid and evolve a rich stream of SO_2 , convertible to either sulphur or strong sulphuric acid (compared with the weak acid obtained by washing the carbon).
- Rockwell. Sodium sulphite produced in a spray drying process is reduced to sodium sulphide in a furnace and the resulting melt is treated with water and carbon dioxide to evolve a rich stream of H_2S , convertible to sulphur by the Claus process. The method has the advantage that coal can be used as the reducing agent whereas the other methods require either natural gas or expensive activated carbon. The process is being tested in a 100-MW facility at Niagara Mohawk's Huntley station.
- Citrate process (U.S. Bureau of Mines). The SO_2 is absorbed in a sodium citrate solution and then treated with H_2S to precipitate elemental sulphur. Two-thirds of the sulphur must be reduced to H_2S for recycling (using natural gas as the

reductant). The process is being tested on a 60-MW industrial boiler by St. Joe Minerals.

- Ammonia scrubbing (NKK). The SO_2 is absorbed in ammonia solution, oxidized to sulphate, and the solution is evaporated to give fertilizer-grade ammonium sulphate. Nippon Kokan Kaisha in Japan has installed the process on a sintering furnace.

2.8.2 Cost. A major drawback to recovery processes is relatively high cost. Complicated process flowsheets, absorbent losses, and high energy requirements contribute to a high cost level.

Bid data are not available because U.S. utilities seldom if ever consider recovery processes in purchasing an FGD system. The Tennessee Valley Authority has published estimates on the leading methods; a recent one presented the following costs (limestone scrubbing is included for comparison).

<u>Process</u>	<u>Capital cost, \$/kW</u>	<u>Operating cost, mills/kWh</u>
Wellman-Lord	143	5.45
Magnesia scrubbing	140	5.24
Rockwell	118	4.82
Limestone scrubbing	98	4.11

One of the items contributing to high cost is the energy requirement. The following levels have been reported:

<u>Process</u>	<u>Energy requirement, % of boiler energy input with no control</u>
Wellman-Lord (sulphur as product)	12-25
Magnesia scrubbing	5-10
Limestone scrubbing	1.5-3

2.8.3 Status. In addition to the drawbacks of high cost and high energy consumption, recovery processes have the disadvantage that most utilities would prefer not to be in the business of selling a chemical product. The main consideration is that the boiler might

have to be shut down if the market for the product failed. This would be especially likely if the product were sulphuric acid or ammonium sulphate because these materials are expensive to store. Elemental sulphur would be a better product in this respect but is the most expensive to produce.

There is currently very little interest by utilities in recovery methods, either in the U.S., Japan, or Germany. In the future, the trends that may favour recovery are higher price levels for sulphur and increases in limestone scrubbing costs because of problems in sludge disposal.

2.9 Conclusions

Control of SO₂ emissions has become a complex technology, with several options available and many factors involved in making the choice between them. One of the main problems is that some of the factors are intangible and therefore difficult to quantify.

In rating the alternatives for SO₂ control, the major consideration is the degree of control required. Some processes are capable of very high removal efficiency but are expensive; others cost much less but are limited to a relatively low level of efficiency. If the required efficiency were the same in all situations the problem would be simple, but regulations vary with plant status (new vs. existing), political entity (federal vs. state vs. local), location (proximity to parks and other public areas), and air quality status. In the U.S., for example, the maze of regulations can result in a different emission regulation for each plant in a given area.

Another consideration is that some of the processes claimed to be more cost effective for a given degree of control have not been proven to be so and thus must be downgraded by a power company faced with the immediate need for control. Others are not developed far enough to make them a reasonable selection even though they are offered by suppliers.

An effort is made below to divide the U.S. regulations into categories and rate the alternatives for use within each category.

1. Higher than 90% removal based on 30-day rolling average. Although not required by the federal regulations, state governments are beginning to set such high standards in various local situations.
2. 90% removal, 30-day rolling average, high-sulphur coal. This is the current federal regulation.

3. 50-90% removal. The current federal regulation for coal with low- to medium-sulphur content is 70-90% removal. This category also covers some of the state regulations for existing plants.
4. Less than 50% removal. This category applies to existing plants where there are no site-specific complications.

The ratings have been developed subjectively, taking into account removal efficiency, estimated cost (including an estimate of maintenance difficulty), projected reliability, and degree of development. Although a more quantitative basis would be desirable, this is not possible in the present state-of-the-art.

The ratings for alternatives are as follows, in the order of preference:

<u>Category</u>	<u>Process Ranking</u>
Higher than 90%	<ol style="list-style-type: none"> 1. Double alkali 2. Limestone scrubbing with promoters 3. Coal gasification-combined cycle (low Btu gas)^a 4. Recovery processes
90%	<ol style="list-style-type: none"> 1. Limestone scrubbing with promoters 2. Limestone scrubbing 3. Double alkali
50-90% (high-sulphur coal)	<ol style="list-style-type: none"> 1. Limestone scrubbing with physical coal cleaning (only part of gas scrubbed) 2. Fluidized bed combustion^a 3. Chemical coal cleaning (including solvent-refined coal)^a
50-90% (low-sulphur coal)	<ol style="list-style-type: none"> 1. Dry process (spray drier)^b 2. Limestone scrubbing (only part of gas scrubbed)
Below 50%	<ol style="list-style-type: none"> 1. Physical coal cleaning 2. Low-sulphur coal substitution

^a When and if developed.

^b If bid lower than limestone scrubbing.

These ratings are approximate and in a given situation, site-specific considerations will be controlling.

3 NITROGEN OXIDES

The pressure for controlling nitrogen oxides has been generally less than for sulphur oxides in all countries. In the U.S., for example, the emission limit under the current federal standard can be attained by altering combustion conditions; flue gas treatment is not required. In Japan, the main pressure has been on SO₂ control, but this situation is changing because ambient SO₂ concentrations have been reduced enough in most areas to meet the standard (mainly by use of low-sulphur fuel). The emphasis has shifted to NO_x, for which the stringent ambient standards are not being met. In congested areas, combustion modification is not considered to be an adequate control; as a result flue gas treatment methods have been developed and are being installed.

The only area in the U.S. where flue gas treatment is required is California. The California Air Resources Board is requiring utilities in the Los Angeles area and oil producers in the Kern County area (where steam boilers are used to enhance oil recovery) to install such equipment.

Several approaches are used in Japan for NO_x control. One is low-nitrogen fuel but it is not as effective as low-sulphur fuel is for SO₂ control because part of the NO_x is formed from the air used in combustion rather than the fuel. Combustion modification, the most cost-effective method, is used to an extreme degree. If flue gas treatment is then required, injection of ammonia to reduce NO_x to nitrogen is favoured; use of a catalyst promotes the reaction and is preferred over non-catalytic operation.

Various wet scrubbing methods have been developed but none seems very promising.

3.1 Combustion Modification

Since NO_x is formed during combustion both by oxidation of nitrogen compounds in the fuel and by reaction of nitrogen and oxygen in the combustion air at high temperature (fuel NO_x and thermal NO_x respectively), two different routes can be taken to reduce NO_x formation.

Thermal NO_x can be reduced by lowering the flame temperature, by reducing the residence time of the combustion gases in the flame zone, and by decreasing the oxygen concentration in the flame. These measures are most effective for gas firing, less so for oil, and relatively ineffective for coal. Oil and coal burn at a relatively slow rate

and as a result the peak flame temperature does not approach that of gas. Thus, means to reduce flame temperature are not as effective.

Moreover, since natural gas contains little or no nitrogen in the form of nitrogen compounds, thermal NO_x constitutes almost 100% of the total, making the effect of reducing thermal NO_x quite significant. Oil and coal, in contrast, produce fuel NO_x that is not reduced significantly by lowering the flame temperature.

Fuel NO_x formation is more complex and not as well understood. The main approach to control is staged combustion, operating the first stage in a reducing (fuel-rich) atmosphere to reduce the nitrogen compounds to elemental nitrogen and then completing the combustion in the second stage.

Combustion modifications usually rank as follows in increasing order of cost: low excess air; off-stoichiometric (two-stage) combustion; flame temperature reduction (usually by gas recirculation); and changes in boiler design. Low excess air is not very significant because boiler operators normally operate with as low excess air as feasible, in the interest of energy efficiency. Moreover, reduction in excess air, if carried very far, can cause problems such as high outlet CO and an increase in particulate emissions. For coal-fired boilers there can be an increase in boiler slagging and corrosion. In Japan, for example, most boilers appear to be operated at an excess air level, giving an optimum balance between the advantages and drawbacks.

3.1.1 Staged Combustion. Staged combustion (off-stoichiometric firing) is one of the more cost-effective methods for NO_x reduction, at least within its limits for control. The basic principle is to reduce the amount of air fed with the fuel so that the initial combustion takes place in a reducing (fuel-rich) environment. The remaining oxygen is introduced downstream from the burner, at a point where the flame has cooled somewhat. The secondary air can be introduced either through ports over the furnace burner array or by taking some of the burners out of service and introducing the air through them. Ando (6) rates "burners out of service" as the more cost effective of the two as far as investment is concerned (only about \$0.40/kW for oil firing). Overfire air ports are more expensive (about \$4.50/kW according to Ando, \$2/kW by others) but give more NO_x reduction (about 27% vs. 18% for burners out of service).

The main problems with staged combustion, no matter how it is accomplished, are boiler slagging and tube corrosion plus some degree of incomplete combustion and loss of boiler efficiency. The increased cost resulting from these is difficult to estimate.

3.1.2 Gas Recirculation. The main drawback to gas recirculation is the high cost of the fans and ducts required, ranging from \$3/kW to \$11/kW. It is effective in gas and oil firing, however, reducing emissions by up to 50%. The method is normally used in conjunction with other modifications.

The status of gas recirculation for coal firing is not clear. It is usually not considered very effective for coal (since flame temperature is not as important as for other fuels). Japanese test work, however, indicates that if very low emissions are required, say 100 ppm NO_x, gas recirculation is essential to decrease the emissions after other modifications have done all they can.

3.1.3 Boiler Design Changes. There has been a general trend in recent years, in both the U.S. and Japan, to reduce NO_x emissions by using special features such as tangential firing (which develops a "cool" flame) and by building the boilers larger to reduce heat release per unit of boiler volume. This has helped but other modifications such as staged combustion are usually required to meet the regulations.

The costs for such boiler design features are difficult to identify.

3.1.4 Low-NO_x Burners. There have been major efforts in Japan and the U.S. to develop burners that incorporate staged combustion and gas recirculation into the design. Numerous design approaches are used; their main objective is to get the advantages of staged combustion and lower flame temperature without the problems mentioned earlier.

Such burners are used widely in Japan, usually in conjunction with standard staged combustion (normally overfire air) and gas recirculation to decrease the NO_x emissions further. By a combination of all methods, emissions have been attained as low as 100 ppm from coal-fired boilers and 50 ppm from oil firing. This was in a test, however, and is not yet commercial practice.

Low-NO_x burners alone are reported by Ando to give about 27% NO_x reduction and to cost about \$2/kW (presumably the excess in cost over standard burners). This is the most cost-effective method other than the staged combustion obtained by taking burners out of service, which involves very little capital cost.

3.2 Fluidized Bed Combustion

Since fluidized bed boilers operate at much lower flame temperatures than standard boilers, 1500-1600°F vs. about 3000°F, thermal NO_x formation should therefore be greatly reduced. Unfortunately, reduction in flame temperature is most

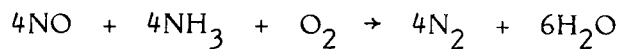
effective for gas and oil whereas coal is the fuel that would normally be used in fluidized bed combustion. For coal, studies have indicated that less than 20% of the NO_x is of the thermal type; fuel NO_x , the main portion, is not reduced very much by lower temperatures.

In tests carried out in England, NO_x emissions in the 1500-1600°F (815-870°C) range were about 325 ppm in coal-fired operation. This is lower than the NO_x concentration of 460 ppm, which is roughly equivalent to the current U.S. standard (0.6 lb/10⁶ Btu), but is higher than the emission reduction suppliers will guarantee for conventional boilers equipped with combustion modification (225-300 ppm, assuming state-of-the-art design).

It is not clear whether combustion modification can be applied to fluidized bed boilers if NO_x emission regulations become more stringent. The air could possibly be injected in stages to give staged combustion but no tests of such a procedure seem to have been carried out.

3.3 Non-catalytic Reduction

Ammonia reacts with NO at high temperatures according to the reaction



The reaction temperature is quite critical. If it is much above a range of 1650-1830°F (900-1000°C), the reaction of ammonia with oxygen to form NO becomes significant, and if much below this range the reaction rate decreases and part of the ammonia remains unreacted. However, if the injection can be made at the correct temperature, if the temperature remains in the proper range, and if rapid mixing is accomplished, a high degree of NO_x reduction can be attained.

Only one utility, Chubu Electric in Japan, has tried this method. In the U.S., Southern California Edison and the Los Angeles Department of Water and Power are installing test units by order of the state environmental agency.

The operation at Chubu Electric (Chita station) has not been satisfactory, the main problem being a change of temperature at the injection point with load variation. The use of a second injector upstream helped but the temperature swings still cause loss of efficiency, as does non-uniformity of gas flow over the cross-section and incomplete mixing in the short residence time available in the proper temperature range. Removal efficiency can be improved by using more ammonia but the resulting ammonia emission is

undesirable. If the emission is limited to 10 ppm, Chubu has found that only about 35% NO_x removal can be attained.

The limited removal efficiency of the method makes its widespread adoption by utilities questionable. If the regulations were such that combustion modification followed by non-catalytic reduction gave compliance, then the low capital cost of the method would make it attractive. Catalytic reduction is so much more efficient, however, that it would probably be regarded as the Best Available Control Technology (BACT).

3.4 Catalytic Reduction

When a catalyst is used to promote the reaction, most of the problems associated with ammonia injection are eliminated. The temperature is not as critical, good mixing can be obtained by injecting farther upstream, and an efficiency of 90% or higher can be reached without excessive ammonia emissions.

The catalytic process is being adopted rapidly in Japan. Starting with the first installation in 1976, there are now 30 utility boilers for which catalytic reduction has either been installed or is planned. The total capacity involved is over 13 000 MW.

The California Air Resources Board has ruled that all utility boilers in the Los Angeles area must be equipped with catalytic reduction equipment. In the first phase of this program, Southern California Edison (SCE) and the Los Angeles Department of Water and Power (LADWP) are each installing a full-scale unit to demonstrate the process. The former has already contracted for the unit and LADWP is in the process of doing so.

3.4.1 Process Evaluation. For catalytic operation, the reactor is located between the boiler economizer and the air heater (see Figure 5), where the temperature is usually in the range of 625-750°F (330-400°C). This is an ideal temperature level for the process; above 750°F (400°C) ammonia decomposition and large reactor volume become problems and below 625°F (330°C), ammonia tends to react with SO_3 in the gas and plug the catalyst by forming solid ammonium bisulphate in the pores.

Catalytic reduction was first used with very clean fuels such as natural gas and naphtha. When the method was applied to "dirty fuels" such as oil and coal, the problem of catalyst plugging with dust became so serious that it was necessary to develop a new type of catalyst resistant to dust. This is known as parallel-flow catalyst, formed in shapes like tubes or plates that allow the dust to pass through while the ammonia and NO_x diffuse to the catalyst walls and react.

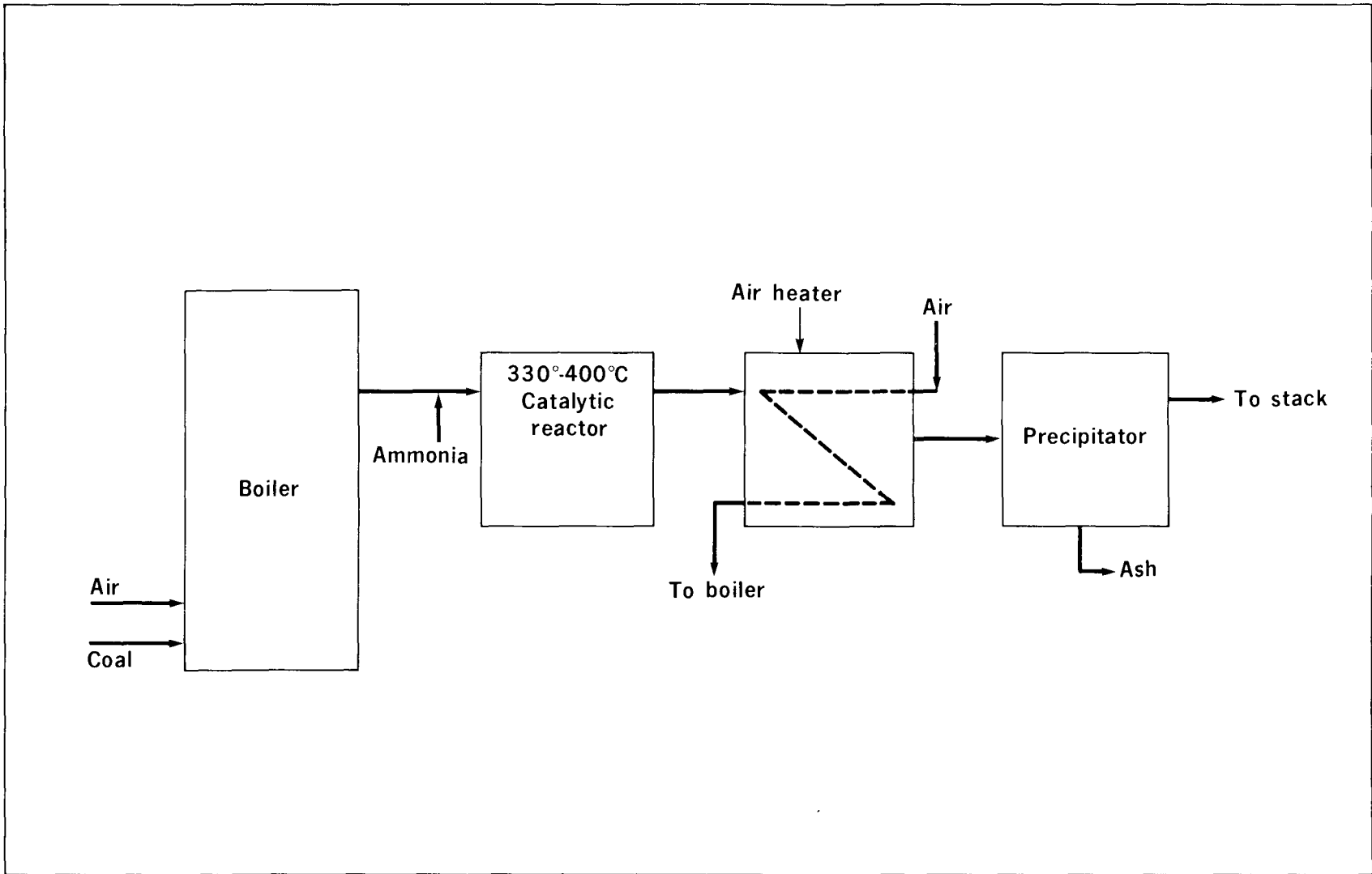


FIGURE 5 CONTROL OF NO_x BY CATALYTIC REDUCTION WITH AMMONIA

The process is still in the development stage with several unanswered questions. The main problems and the status of development are as follows:

- Catalyst composition and preparation. The characteristics of an ideal catalyst (which has not yet been developed) are high NO_x reduction efficiency over the range of temperatures resulting from boiler load swings; low oxidation of SO_2 to SO_3 ; resistance to poisoning by SO_3 and alkalis; resistance to blockage by coal ash; low erosion by ash; and structural integrity (resistance to mechanical stress and thermal shock). The usual catalyst is made up of vanadium and titanium oxides, sometimes deposited on a support. The vanadium pentoxide is quite active but promotes SO_2 oxidation as well as NO_x reduction. Catalysts are being developed that are said to minimize oxidation and to improve performance regarding the other characteristics. Preparation methods include coating on a support (ceramic or metal), impregnation into a support, and molding of a catalyst mixture (no support). It is not clear which is best. Coatings tend to come loose, impregnation is expensive, and molding gives a more bulky catalyst with higher pressure drop.
- Catalyst shape. It is not clear which of the various shapes -- tubes, plates, or various types of "honeycombs" -- is best. The main development objective is to minimize the frontal area of the catalyst to reduce pressure drop.
- Catalyst life. Although improvements have been made on resistance to SO_3 poisoning, there is still some problem in regard to poisoning -- especially by alkali compounds in coal.
- Ammonium bisulphate formation. The main problem in catalytic reduction, especially for coal firing, is plugging of the air heater by deposits of ammonium bisulphate. Deposition in the catalyst is not a problem if the temperature is kept above 330°C or so but since the air heater temperature gets down to about 150°C , deposition cannot be avoided. A remedy is to reduce the amount of ammonia and SO_3 entering the air heater. In Japan the consensus is that the ammonia leaving the catalyst should be no higher than 5 ppm, accomplished by using an adequate amount of catalyst and by not aiming for higher than about 80% NO_x reduction (thus reducing the amount of ammonia fed). The SO_3 is reduced by burning low-sulphur fuel and by using a low oxidation catalyst; such catalysts are more expensive but probably justified.

- Downstream effects include possible ammonium bisulphate emission as a fume and reduction in precipitator efficiency because of less SO₃ in the gas. Unreacted ammonia could be beneficial, however, if caught in a limestone scrubber; the ammonia should have the same beneficial effect on mass transfer as does magnesium.

3.4.2 Cost. Numerous cost data have been reported on catalytic reduction but it is difficult to determine the total owner cost. Most users and suppliers state that the catalyst accounts for about half of the initial battery limits (vendor) cost. Since the catalyst costs about \$9/kW, the supplier cost is thus about \$18/kW. If it is assumed that the total owner cost is twice this, which is sometimes a reasonable assumption and sometimes not, the overall cost is about \$36/kW. This is in general agreement with some of the published estimates but the capital cost obviously varies widely with site-specific considerations (especially design NO_x removal efficiency). The operating cost is even more variable, probably ranging between 1.5 and 5 mills/kWh.

3.4.3 Status. Catalytic reduction is very likely to be the basic NO_x control process in the future but much more development is required, especially for high-sulphur coal. It is not clear, for example, whether the concentrations of SO₃ and unreacted ammonia can be kept low enough with high-sulphur coal to avoid an unacceptable degree of air heater plugging.

3.5 Conclusions

The situation for NO_x is the same as for other pollutants -- the selection of abatement method depends on the degree of control required. For example, combustion modification is adequate for the present U.S. federal regulation (0.6 lb/10⁶ Btu emission limit for coal and 0.3 lb/10⁶ Btu for oil), but flue gas treatment is required in Japan and California, where more stringent regulations are in effect.

Combustion modification can be divided into two categories: the present state-of-the-art, and new technology being developed. If the U.S. federal standard were reduced below the present level, the present combustion modification techniques might be capable of compliance down to about 0.4 lb/10⁶ Btu for coal and 0.15 lb/10⁶ Btu for oil. The main question is the adverse effect of such extreme modifications on boiler operability and efficiency. For the newer technology now under development, Japanese

data indicate a possible reduction to 100 ppm (about 0.15 lb/10⁶ Btu) for coal and 50 ppm (about 0.06 lb/10⁶ Btu) for oil. Again, it is not clear what this does to boiler operability.

Wet methods for NO_x control are much more expensive and energy intensive than either combustion modification or reduction with ammonia. Unless some breakthrough occurs, wet processes are not likely to be a future consideration.

A rough ranking of alternatives is as follows:

<u>Degree of reduction, %</u>	<u>Process alternative</u>
90% or higher	1. Catalytic reduction with more than the normal amount of catalyst (low-oxidation type for high-sulphur coal), preceded by full combustion modification
50-90%	1. As above, with a normal amount of catalyst 2. Combustion modification (all types) ^a followed by non-catalytic reduction 3. Combustion modification alone ^b
Below 50%	1. Low-NO _x burners ^c 2. Overfire air ^c 3. Gas recirculation (except for coal) ^c

^a Applicable mainly in lower part of range.

^b When and if developed for high removal. Applicable mainly in lower part of range.

^c Used in combination with others if necessary to achieve the required reduction level.

4 PARTICULATE MATTER

Requirements for particulate removal vary with the fuel. Gas needs none and in the U.S. only a few oil-fired boilers have equipment for particulate collection. The situation is different in Japan, where the more stringent regulations have required equipping practically all oil-fired boilers with precipitators. For coal, the relatively high ash content has made some sort of particulate collection necessary almost from the beginning of the power industry.

The control method for coal firing has changed over the history of the power industry. Cyclonic collectors were used widely in the beginning but low efficiency and high maintenance have practically eliminated them. Electrostatic precipitators became popular, first in tandem with cyclones, and later as the sole collection device. In the early 1970s, several wet scrubber installations were made, mainly for low-sulphur coal or for combined removal of SO₂ and particulates. More recently, tighter regulations have pushed the industry toward bag filters.

Much of the ash can be removed from coal during cleaning operations, either physical or chemical. Physical coal cleaning is used, for example, to remove mining residue from some 50% of the coal mined in the U.S. This is an incomplete removal, however, ranging from 25% to 50% for simple cleaning; even cleaned coal usually contains some ash (typically 8% to 12%).

The methods proposed for removing sulphur by chemical coal cleaning remove more of the ash but not enough to avoid the need for a precipitator or baghouse. Even the most extreme treatment, such as dissolving the coal and separating the ash by filtration as in the solvent-refined coal process, leaves enough ash in the product that the current federal standard in the U.S. (0.03 lb/10⁶ Btu) is exceeded if the solvent-refined coal is burned without particulate collection.

The partial removal of mineral matter in cleaning may reduce precipitator or baghouse cost to some degree.

There is much less latitude in removal requirements for particulates than for SO₂ or NO_x. Whereas 70% removal of SO₂ or 50% reduction in NO_x emission may be acceptable, over 99% removal of particulate is required in most cases. There is more particulate in the first place -- usually over twice as much by weight as for SO₂ -- and the visual impact of the plume has always made particulate emission subject to more stringent regulations, as illustrated by the current U.S. federal standard of 0.03 lb/10⁶ Btu

for particulate, 1.2 lb/10⁶ Btu for SO₂, and 0.6 lb/10⁶ Btu for NO_x (all for coal) for new boilers. Existing units are also subject to regulations that require particulate collection. There is no coal low enough in mineral matter which will avoid the requirement for particulate collection.

4.1 Electrostatic Precipitation

Particulate removal by precipitators has several advantages for large boilers, including low pressure drop, no drop in efficiency when the boiler load is reduced (efficiency actually increases because of the lower gas flow), and relatively low operating cost.

4.1.1 Design Problems. The performance of a precipitator depends mainly on the electrical resistivity of the dust particles; the higher the resistivity the larger the precipitator has to be (longer gas path or lower gas velocity) to give the particles time for sideways movement to the collecting plates. The migration velocity declines rapidly when the resistivity is higher than about 2×10^{10} ohm-cm.

A major factor affecting resistivity is temperature. Unfortunately the usual precipitator operating temperature is the worst that could be selected over the range available in a boiler train. Precipitators are usually placed just after the air heater, at which point the temperature is in the range of 275-300°F (135-150°C) -- as low as possible to get good energy recovery (in the air heater) but high enough to avoid acid condensation from the SO₃ present in the gas.

The sulphur content of the coal is another major consideration, since SO₃ adsorbs on the ash and makes it more conductive (lower resistivity). At sulphur levels of about 2% and higher, there is enough SO₃ to give adequate performance at a reasonable SCA (specific-collection area of the plate electrodes per 1 000 acfm of gas). As the sulphur level declines to the 0.6-0.8% level common for western coal, the required plate area, and the precipitator size and cost, become quite high.

One of the remedies for the high ash resistivity associated with low-sulphur coal is to place the precipitator before the air heater where the temperature is in the range of 625-750°F (330-400°C) and the resistivity is low because of the high temperature. Such "hot" precipitators have been installed on several of the U.S. boilers burning low-sulphur coal. Operation seems to have been generally acceptable but some have not done well, mainly because of factors such as higher resistivity than expected

(resistivity affected by factors other than temperature) and high maintenance due to deposits in the precipitator.

High resistivity can also be countered by adding SO_3 to the gas, with the objective of producing a gas composition about the same as if high-sulphur coal had been burned. When carried out properly, such gas conditioning is effective. Most of the applications so far have been for upgrading existing installations but the method has been used in a few instances for new boilers, the objective being to reduce precipitator size and cost.

Other conditioning agents, either injected or added to the fuel, have also been proposed and some used commercially. They do not seem to be any more cost effective than SO_3 injection.

4.1.2 Cost. The cost of precipitators varies widely with the ash characteristics and the degree of particulate removal required. The lowest cost is for high-sulphur coal and a relatively lenient emission standard such as the $0.1 \text{ lb}/10^6 \text{ Btu}$ allowable under the 1970 Clean Air Act Amendments in the U.S. Conversely, the highest cost is associated with low-sulphur coal and the $0.03 \text{ lb}/10^6 \text{ Btu}$ emission limit under the current U.S. federal standard.

As an example of current precipitator costs, a 1500-MW system was purchased recently for about \$30/kW (not including interest during construction and owner overheads). This was for coal containing 1-1.5% sulphur and an emission standard of $0.1 \text{ lb}/10^6 \text{ Btu}$; the SCA was 460, considerably higher than the 200-300 usually considered adequate for high-sulphur coal but lower than the 500-plus required for extreme conditions of ash resistivity and removal requirement.

Studies have indicated that an SCA of about 700 may be required to meet the $0.03 \text{ lb}/10^6 \text{ Btu}$ standard if the fuel is low-sulphur coal. There is one example of this in the U.S., at the Neal station of Iowa Public Service. The SCA is 880, ash resistivity $5 \times 10^{12} \text{ ohm-cm}$, and boiler size 520 MW. When the SCA was adjusted to 740, the emission was $0.023 \text{ lb}/10^6 \text{ Btu}$ -- complying with the $0.03 \text{ lb}/10^6 \text{ Btu}$ standard. The cost of the precipitator system was \$52 kW (1979 dollars). Since the SCA is 880, a unit with an SCA of 740, as needed to meet the standard, would have cost less.

The operating cost also varies with the emission limit. A rough approximation is 1.5 mills/kWh for $0.1 \text{ lb}/10^6 \text{ Btu}$ and 2 mills/kWh for $0.03 \text{ lb}/10^6 \text{ Btu}$.

4.1.3 Status. Precipitators have been the mainstay of the power industry for particulate collection and will continue in that capacity for some time. However, the growing regulatory pressure to reduce emission of very fine particulates makes precipitators more expensive and therefore not always the clear choice as in the past. Moreover, as the limits on total mass emission (as opposed to emission of fine particulates) are reduced, problems such as rapping losses, re-entrainment, and "sneakage" (bypassing the plates) become more important.

Development work underway may improve the position of precipitators. For example, tests of precharging with a special ionizer have shown promise.

4.2 Fabric Filtration

The use of fabric filters (usually called baghouses in large sizes) is an old technique for dust collection but has not been applied to utility boilers until recently. The cost was too high and there was concern as to operating reliability in view of potential problems such as bag tearing or plugging. Bag filters have the outstanding advantage, however, of very high intrinsic removal capability, so that the cost is no higher for a stringent regulation than for a lenient one. Particulate emission is usually reduced to about 0.007 grain/acf, well below the $0.03 \text{ lb}/10^6 \text{ Btu}$ standard.

Application to utility boilers is so new that only four installations of any significant size are operating. The recent promulgation of the $0.03 \text{ lb}/10^6 \text{ Btu}$ standard, however, has encouraged adoption. Several large systems have been contracted recently and more are likely to be in the future.

4.2.1 Design and Operation. The main consideration in baghouse design and operation is protecting the bags from operating upsets. High temperatures can cause failure because of damage to the fabric, and condensation of acid mist at low temperatures can result in damage and plugging. Various stresses can cause bag breakage or tearing.

Pressure buildup over an extended period can also be a problem. The two largest operating systems, both in Texas, have had this difficulty. In each case the bags had to be replaced within a year. The cause is not clear; presumably the interstices through which the gas flows become obstructed by very fine particles. One theory is that alkali salts are caught in the fibres and then expand by picking up moisture.

There are many operating problems that may result from the fact that the bag filter is a barrier to gas flow whereas a precipitator is not.

- Boiler startup can result in moisture condensation on cold surfaces, including the baghouse. In addition, oil firing during startup produces hydrocarbon vapours that can either blind the bags or cause a fire or explosion in the baghouse. A gas bypass is usually provided to help prevent this.
- New bags can become plugged rapidly if the ash has a high content of fines. Precoating with a material such as limestone helps prevent this and also minimizes acid attack.
- A preheat system is often used to get the baghouse hot before gas is passed through it.
- Boiler leaks can result in water getting into the bags.
- A boiler "fuel-rich" trip may cause deposition of unburned fuel as far downstream as the baghouse. Air purging at such a time can cause an explosion.
- There is some indication that a high inlet dust level reduces the capability for meeting a fixed emission limit. In tests at the Kramer station of the Nebraska Public Power District (7), for example, the emission at an inlet level of 0.5-0.7 grain/scf was 0.006-0.007 grain/scf, well within the $0.03 \text{ lb}/10^6 \text{ Btu}$ standard (about 0.015 grain/scf). At an inlet loading of 2-3 grain/scf, however, the outlet was 0.018-0.019 grain/scf, which does not meet the standard.

The severity of these problems will not be known until large installations have been on line long enough to produce adequate operating data.

4.2.2 Cost. The consensus among cost estimators seems to be that the cost of a bag filter is about the same as for a precipitator when conditions are such that the latter requires an SCA of about $500 \text{ ft}^2/1000 \text{ acfm}$. This is approximately the level required for a $0.1 \text{ lb}/10^6 \text{ Btu}$ emission standard with medium-sulphur coal. Thus the capital cost would be expected to be \$30-35/kW.

This was not borne out in the actual bidding situation for the 1500-MW system mentioned earlier. The lowest adjusted bid for fabric filters was \$47/kW compared with \$30/kW for precipitators. It is not clear why there should be such a disparity between estimates and actual bids but the latter are more meaningful.

Even though actual bidding indicates that baghouses may cost more than estimates indicate, the deficit disappears rapidly as the emission limit is lowered.

Assuming that baghouses cost the same as for precipitators at the $0.03 \text{ lb}/10^6 \text{ Btu}$ level, which would be a sort of worst case, baghouses would be favoured because they retain their capability better over extended operation than precipitators. The latter require frequent fine-tuning to retain a high removal efficiency.

As to operating cost, fabric filters are at a disadvantage because of bag replacement cost. The cost is variable, depending on the bag life assumed. A two-year life is assumed in most of the estimates but this remains to be proven.

4.2.3 Status. Fabric filtration is in a similar situation to dry scrubbing in the FGD field. Cost estimators have convinced themselves that the approach is more cost effective but there is no full-scale confirmation yet. There is considerable evidence that precipitators are the best choice at the $0.1 \text{ lb}/10^6 \text{ Btu}$ level but at $0.03 \text{ lb}/10^6 \text{ Btu}$ the situation is not clear. Nevertheless, the general pressure to reduce fine particulate emissions is likely to favour fabric filtration in the future.

4.3 Wet Scrubbing

Particulate removal by wet scrubbing is widely used in other industries but was not applied in the U.S. utility industry until the 1970s. The reasoning for using wet scrubbing was that in view of the high resistivity of ash from low-sulphur coal, plus the relatively lenient regulations in effect at that time, scrubbers would be cheaper than precipitators. Venturi scrubbers were used, with a fairly high pressure drop.

Beginning in 1973, systems were installed in which wet scrubbers removed both particulates and SO_2 , using lime or limestone as the SO_2 absorbent. The first of these was at the La Cygne station of Kansas City Power and Light, followed by the very large installation (1 800 MW) at Pennsylvania Power's Mansfield station. Venturi scrubbers were used for particulate removal in each case.

Such combination systems have the outstanding advantage that the scrubbers have a double function and therefore the cost is low in comparison with a precipitator or baghouse for particulate removal followed by scrubbing for SO_2 control. The tightening regulations for particulates, however, have in effect eliminated this very cost-effective combination. Scrubbers are not capable of meeting a $0.03 \text{ lb}/10^6 \text{ Btu}$ emission standard except at a pressure drop so high that the cost advantage of the combination would be lost. With the $0.03 \text{ lb}/10^6 \text{ Btu}$ standard now in effect it is unlikely that any further venturi scrubbers will be constructed. This is unfortunate because these scrubbers have an intrinsic capability for removing about 90% of the incoming particulates with little if

any added cost over that for SO₂ scrubbing. The problem is in removing fine particulate matter, for which scrubbers are not as effective as precipitators or baghouses unless a very high pressure drop is incurred.

There may be some hope for scrubbers in a new combination now being offered by two suppliers. All the dust enters the scrubber, where most of it is removed along with the SO₂, but the gas then passes through a wet precipitator to remove residual particulates plus mist formed in the scrubber (no mist eliminator is needed). There are several advantages to such a combination other than the lower cost. Full-scale testing will be required, however, before any commercial use can be expected.

4.4 Conclusions

The ranking of alternatives for particulate control is simpler than for the other pollutants. For a stringent standard such as 0.03 lb/10⁶ Btu, fabric filtration is very likely the best choice unless development work moves precipitators into a better position. (There does not seem to be any way of improving fabric filter removal efficiency.)

For 0.1 lb/10⁶ Btu and higher emission levels, precipitators are preferable, perhaps with gas conditioning added for low-sulphur coal.

For all emission levels, wet scrubbing followed by a precipitator (coupled with SO₂ removal) could be the most cost effective if development work is successful.

REFERENCES

1. Evaluation of Physical/Chemical Coal Cleaning and Flue Gas Desulfurization, EPA-600/7-79-250 (Nov. 1979).
2. Ando, J., SO₂ Abatement for Stationary Sources in Japan, Report prepared for U.S. EPA (June 1976).
3. Low-Btu Gasification Looks Like a Loser, Electric Power and Light, p. 20 (Feb. 1979).
4. Utility Boiler Design/Cost Comparison: Fluidized Bed Combustion vs. Flue Gas Desulfurization, U.S. EPA-600/7-77-126 (Nov. 1977).
5. Davis, Wayne T. et al., Research on the Removal of SO₂ by Additive Injection Techniques on a Stoker-Fired Boiler, Paper presented at APCA Conference, Houston, Texas (June 1978).
6. Ando, J., NO_x Abatement for Stationary Sources in Japan, U.S. EPA-600/7-79-205 (Aug. 1979).^x
7. Stenby, E.W., et al., The Kramer Baghouse, Combustion (Oct. 1979).