# THE ATMOSPHERIC IMPACT OF EXPANDED COAL UTILISATION

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The combustion of fossil fuels is the main contributor to the increase in the concentration of not only carbon dioxide in the atmosphere but also of oxides of nitrogen and sulphur dioxide. Coal now contributes about 25% to the primary energy generation in the world and its use is projected to increase substantially even as the overall contribution of fossil fuels to energy generation decreases. To mitigate the resulting adverse environmental effects of the increased use of coal, it is imperative that the development and adoption of newer environmentally acceptable technologies for coal conversion is accelerated. These emerging technologies are reviewed and relevant R & D needs identified. These are summarised below:

- Reduction of ash and sulphur in coal to yield cleaner/modified coal.
- Development of better combustion systems such as: pressurised fluidised bed; slagging combustors with sorbents like limestone to fix sulphur; and circulating fluidised bed.
- Conversion of coal into: gaseous fuel for integrated gasification combined cycle power generation; and synthesis gas for methanol production.
- Better techniques for removal of SO<sub>2</sub> and NO<sub>x</sub>.
- Hot Gas Clean-up Technology.
- Retrofit possibilities of some of these technologies in existing power plants.

Reference is made to the need for more refined analytical tools and techniques for H<sub>2</sub>S, COS, CS<sub>2</sub>, SO<sub>2</sub>, SO<sub>3</sub> and NO<sub>x</sub> monitoring in the environment and advanced research on the chemistry of SO<sub>2</sub>/NO<sub>x</sub> and systems modelling but this is not discussed in detail.

### INTRODUCTION

The greenhouse effect is now well understood and it is known that, during this century, CO<sub>2</sub> concentration in the atmosphere has increased from about 280 ppmv to about 350 ppmv. Vegetation loss and fossil fuel combustion are the main contributors to this increase. Fossil fuel combustion further contributes to the increase of oxides of nitrogen and sulphur dioxide; particulate matter in flue gases of coal combustion also releases heavy metal and metal ions in the air. These emissions deleteriously disturb the delicately balanced chemistry of the atmosphere—perhaps even in the thermosphere.

## COAL AS ENERGY SOURCE

On a global basis coal supplied about 30 percent of the primary energy in 1975 and is likely to increase further. There was a time, when coal was the main source of industrial organic chemicals. Then came the "Oil age", during which time coal became the forgotten feed stock of the 1960's; and then came the phenomenal rise in crude oil prices (Table 1). Coal, largely overlooked up to the sixties, became suddenly "affordable"; of course it was always "available". Expansion in coal mining was never behind demand.

Year Cost per barrel (US Dollars)

1950's < 2

December 1978 ~ 13

1979 ~ 36

**TABLE 1: Oil Prices** 

The OECD countries discovered that their energy consumption was outpacing energy production and energy policy decisions were taken towards coal utilisation. Developing countries like India were no better off. In fact, the rising oil prices hit them more severely. Money was neither available to pay for oil nor for converting their economies to coal by increasing coal mining or establishing coal conversion technologies.

The 1979 oil prices shock was much more severe than that of 1973-1974 and the International Energy Agency (IEA) adopted "the Principles for IEA Action on Coal" and established a Coal Industry Advisory Board (CIAB). A landmark report (Wilson, 1980), was published by the World Coal Study Group, generally referred to as the WOCOL report, Wilson (1980). Research and Development in integrated utilisation of coal was very well funded in several countries e.g. Australia, Federal Republic of Germany, German Democratic Republic, Japan, the Netherlands, the United Kingdom, the United States of America and the USSR. Environmental protection aspects were very much kept in mind. As the oil prices declined, funding for environmental protection also decreased, especially in the United States of America

in the Reagan era during the 1980's. Calm in the oil market has always lulled governments and industry into a false sense of security. The proven world reserves of coal are estimated to be around 665 billion tonnes of coal equivalent and at the present rate of mining and consumption will last about 240 years. But the total resource base of coal is much larger; it is about 11,000 billion tonnes. Technological advances in mining and technologies of the future such as underground coal gasification can make these resources recoverable. The single major use of coal is for generation of steam and power by coal combustion. In the industrial fields, iron and steel and cement are the main users; the former is using coking coals. Domestic use is now confined to developing countries by and large.

The global primary energy supply projection estimates are given in Table 2 and the estimated annual rate of growth in coal use is given in Table 3.

Year	Energy required (exajoules/year)	Percentage available from coal
 1975	250	28
2000	360	<del></del>
2025	530	50
2050	750	_
2075	930	

TABLE 2: Global Primary Energy Supply Projection (Mintzer, 1987)

TARIE 3.	Fetimated	Annual Rate	of Crowth	in Coal	I lea	(Mintzer	1027)

Year	Percent annual rate of growth
25 Year intervals	
1975 - 2000	1.7
2000 - 2025	3.0
2025 - 2050	2.3
2050 - 2075	1.5
Whole period	
1975 - 2075	2.1

## COAL AND POLLUTION

Coal production and combustion contribute to pollution in several ways. As much as 25 percent of coal dust and fines are produced during mining, transport, size reduction and further utilization. During these operations, not only air but also surface waters get polluted. Power generation plants burning coal emit stack gases. These are aerosols loaded with particulate matter and noxious chemicals. In addition there is primary and secondary pollution of streams with ash. The particulate matter, coal

ash and unburnt coal all have elevated concentrations of heavy metals; the noxious gases in the stack contain the oxides of carbon, nitrogen and sulphur including H<sub>2</sub>S and COS.

The emission regulations in advanced countries are strict and are rigidly enforced. This is not yet true of less developed countries, which are power hungry but do not have sufficient funds to invest in electrostatic precipitators or flue gas desulphurisation. India can be cited as an example of a country which has all the knowledge, knowhow and awareness, but has an acute resources constraint with many competing claims on limited finances. India prescribes standards for sulphur dioxide emission, on the basis of control effected only through prescribed height of the stack depending upon the size of the power plant. Particulate matter standards prescribed in India are given in Table 4.

What then will be the global environmental impact of increased consumption of coal for power generation? Large industrial boilers emit  $5-8 \times A$  kg of particulate matter per tonne of coal combusted, where A is the ash percent in coal.

The sulphur dioxide emission is approximately  $19 \times S$  kg per tonne of coal, where S is the percent sulphur in coal.

Mintzer (1987) has estimated the addition of carbon to the atmosphere by coal up to 2075 as given in Table 5.

Boiler size	Emission (mg/Nm³)		
	Pre 1979	Post 1979	Protected area
Less than 200 MWe	600	350	150
200 MWe and above	-	150	150

TABLE 4: Emission Regulations for Particulate Matter in Power Plants in India

TABLE 5: Addition of Carbon to the Atmosphere by Coal Use

Year	Energy from coal (exajoules)	Carbon added (billion tonnes)
1975	75	-
2000	115	31
2025	243	65
2050	429	115
2075	620	167

## MITIGATING COAL BASED POLLUTION

What then are the methods that can be adopted in the future to mitigate the increased environmental pollution by the projected increase in the utilisation of coal

for energy generation? Electrostatic precipitation and wet flue gas desulphurisation are well established industrial technologies. The challenge is to develop newer cost effective methods of:

- (i) Utilising coal fines;
- (ii) Removing particulate matter and SO, and NO, from gaseous streams;
- (iii) Minimising the formation of NO; and
- (iv) Coal conversion that are, per se, less noxious to environment.

An attempt will be now made to outline the main technological approaches and the state of development. The chemistry involved is simple; much more interesting chemistry is involved in the mechanisms of formation of the gaseous pollutants and their interactions with other reactive chemical entities at various levels and will be referred to later in brief. The chemistry of formation of oxides of nitrogen and sulphur in solid and liquid fuel combustion is reviewed in a special report on Combustion Chemistry (Miller and Fisk, 1987).

## UTILISING COAL FINES

Coming back to mitigating the effects of pollution from coal usage, the first pollutant to be handled is coal fines. The disposal problem is more acute with high ash coal. One promising approach is to agglomerate these with oil, if necessary, after further processing through froth flotation/oleo-flotation. A process developed by the National Research Council of Canada has been demonstrated at a scale of 30-40 tonnes per hour at Pennsylvania Coal Cleaning Facilities. It uses less than 7 percent oil as compared to 10 percent or more in earlier processes and the cost of production of the agglomerates is estimated to be between US \$ 6.50-11.00 per tonne.

A 10 tph pilot plant is about to be commissioned at the Patherdih washery of Bharat Coking Coal Ltd. (India), using the process knowhow of Central Fuel Research Institute, Dhanbad of the Council of Scientific & Industrial Research, New Delhi. The oil consumption is 8-12 percent and laboratory studies on reducing it are in progress.

### PARTICULATES REMOVAL

The quantity of particulate matter in stack gases depends both on the ash content of the coal and on the type of boiler. The average emission levels are as given in Table 6

Ash in coal (%)	Particulates in coal (kg/tonne)	
10	50-80	
20	100-160	
30	150-240	
40	200-320	

TABLE 6: Particulate Emissions from Coal Fired Industrial Boilers

Electrostatic precipitators do remove more than 99 percent of these but at atmospheric pressure. With many pressurised fluidised bed combustion and coal gasification plants likely to be built, knocking down the particulates under pressure has become imperative. Several technologies are under various stages of development. Ideally such a technology should aim at:

- (i) Working at emission gas temperature, pressure and velocity;
- (ii) High particle loading;
- (iii) High efficiencies, both overall and for the removal of submicron size particles;
- (iv) Low pressure drops; and
- (v) Higher cycle times.

Some of the technologies being developed are outlined below:

## Granular shallow bed (GSB)

This is meant to be used with pressurised fluidised bed combustion (PFBC) and uses 620 µM sand as the bed medium. A six element bed was used and gave a collecting efficiency of 99.2 percent at a flowrate of 500 cfm and at 11 atmospheres and 1600°F (~870°C) (Westinghouse Electric Corporation with Ducon Inc., and Burns and Roe, Inc.).

## High temperature moving bed granular bed filter

This has been evaluated with an atmospheric fluidised bed combustion and operated at a gas velocity of 1600 cfm at 1500-1600°F (~820-870°C). The overall efficiency was above 99 percent with a submicron collecting efficiency of more than 96 percent.

## Electrostatic granular bed and electrocyclones

These are being developed and tested by General Electric Co.

# High pressure electrostatic precipitators

These are also under development. Tests at the PFBC test facility at Curtis-Wright, New Jersey, showed a particle collecting efficiency in the range of 98-99.5 percent at 1400-1500°F and 84 psi.

### Ceramic filters

These are receiving the most attention and are of several types.

### POROUS CERAMIC CROSSFLOW FILTERS

These permit combustion at high gas velocities (3.35 metres/min.) and operate at 800°C and 162 psi, with a modest pressure drop of only about 0.3 psi. Particle loading up to 5000 ppm can be tolerated; the cycle time is 25 min.

## HIGH TEMPERATURE HIGH PRESSURE (HTHP) CERAMIC TUBE FILTERS

These are considered the most promising for commercial application. EPRI have tested a ceramic element filter module at the PFBC pilot facility in Grimethorpe, UK. The module contains 130 cylindrical elements made of porous ceramic, each 4.5 ft. long and with an i.d. of 1.7 in. and with an inner support made of silicon carbide granules and a thin outer layer of fine alumina fibres and silicon carbide grains. Ceramic seals and a counterweight hold the elements in place in a refractory lined vessel; cleaning is by pulse jet air. Though the process leads in scale up, commercially available hardware may not be available for a few years to come.

#### SULPHUR COMPOUNDS, FORMATION AND RELEASE

The formation and release of S compounds (SO<sub>2</sub>, H<sub>2</sub>S, COS, CS<sub>2</sub>) during coal combustion depends upon several factors including the following:

- (i) Coal type—Bituminous coal produces more SO<sub>2</sub> and less H<sub>2</sub>S, COS and CS<sub>2</sub> levels compared to sub-bituminous coal;
- (ii) Stoichiometric ratio—Increase of air increases S conversion and SO<sub>2</sub> levels and decreases S, COS and CS<sub>2</sub> levels;
- (iii) Swirl of the secondary air also affects the distribution of the sulphur species, if the stoichiometric ratio is kept constant, Zang (1989);
- (iv) Sulphur composition of the coal (pyritic, sulphate and organic S)—The organic sulphur is released early during coal devolatilisation and the pyritic sulphur is released only later during char combustion. X-Ray studies have shown that the reaction products of pyritic combustion are FeS<sub>(1+x)</sub>, FeS and Fe<sub>2</sub>O<sub>3</sub>;

- (v) Combustion temperature and combustion rate also affect the sulphur release pattern; and
- (vi) The nature of the coal bed, fixed or fluidised, also plays a part in this (Schouter et al., 1987).

The main research and development emphasis on removal of sulphur compounds from flue gases is on the development of cost effective hot gas desulphurisation methods as an alternative to wet flue gas desulphurisation. Hot gas cleaning assumes special importance in integrated gasification combined cycle (IGCC) mode of power generation, because the sensible heat of raw gas is retained.

## **DESULPHURISATION TECHNOLOGIES**

The different methods of removal of sulphur compounds from flue gases can be roughly grouped as follows:

- (i) Addition of sorbents like limestone during combustion. The sulphur dioxide is converted into calcium sulphite and oxidized to calcium sulphate which will be distributed in the ash and flue gas particulates;
- (ii) Injecting lime based dust in flue gas duct;
- (iii) Conversion of sulphur dioxide to sulphites; the subsequent release of sulphur dioxide, its concentration and oxidation yields sulphuric acid; and
- (iv) Use of metal oxides or mixed metal oxides such as Fe<sub>2</sub>O<sub>3</sub>; CuO; ZnO; CuO/ZnO; V<sub>2</sub>O<sub>5</sub>/V<sub>2</sub>O<sub>3</sub>; zinc ferrite (ZnFe<sub>2</sub>O<sub>4</sub>); zinc vanadium oxide/V<sub>2</sub>O<sub>5</sub>.

These methods are briefly described below:

As stated, one method of fixing sulphur in coal is the addition of sorbents like limestone. In fluidized bed combustion, the sorbent can be fed directly into the combustor. The particle size of the sorbent and its structure—crystalline or amorphous, hard or soft—and composition are important. Dolomitic limestones with over 20 percent magnesium carbonate enhance sulphur dioxide absorption at lower Ca/S molar ratios, reduce carryover and can slightly lower NO, in the flue gas.

Addition of sorbents during combustion changes the nature of the ash and discharge mechanisms have to be carefully designed, e.g. the ash contains reactive pozzolanas of small particle size; unreacted limestone, grit from limestone and calcium sulphate from the sulphur absorption reactions are the other constituents; possible uses are soil stabilisation, structural fill and cement additives.

Sorbents also change the nature of the solids in the flue gas. For example, calcium sulphate becomes cementatious in the presence of moisture and filter bags can be blinded. ESP designs are also affected because sorbents in fluidised bed operations increase the dust loading compared to fly ash without sorbent addition. If sorbent

addition equipment is to be retrofitted to an existing power plant with an electrostatic precipitator, the change in the quality and quantity of the solid emissions is a matter of some concern. Different boilers have different requirements for sorbents but, generally the Ca/S ratio varies between 2/1 and 3/1 for removal of 80-90 percent SO<sub>2</sub>. Some Circulating Fluidised Beds (CFBs) have recorded Ca/S between 1.4/1 and 2.2/1 for 90 percent SO<sub>2</sub> removal from coals containing 5 percent S (Makanse, 1987).

Amongst the several Clean Coal projects under the US Department of Energy Clean Coal Technology Program, a TRW Proposal of Advanced Slagging Combustor Utility Demonstration Project is typical of the magnitude of work, the time scale and the funding involved.

Research and development started in 1975 with coal injection experiments and flow parameters; coal fired combustion tests that varied air injection techniques and pressures followed. A full sized plastic coldflow model was constructed to optimise combustor geometry through detailed aerodynamic flow visualisation tests. A computer model was developed in parallel to analyse thermochemistry, aerodynamics, combustion, heat transfer and slag flow. Feasibility test units with 34, 70 and 170 MM Btu/hr (6 atm) were then successively constructed and used for power generation on an MHD test unit.

Parallel development work on similar atmospheric pressure combustors was carried out to construct test units of 10 MM Btu/hr with sorbent injection and then of a 40 MM Btu/hr unit. In all, well over a thousand test runs on different units were carried out before the construction of the 40 MM Btu/hr unit. In this unit, 94 percent of the slag and 90 percent SO<sub>2</sub> could be removed and NO<sub>x</sub> controlled at 230-450 ppm with over 99.5 percent carbon burnout. Then followed a 40 MM Btu/hr combustor built to commercial standards and installed on an industrial scale boiler. Till October 1988, over 6000 hours of operating time had been accumulated.

It has now been proposed to retrofit a 69 MWe boiler with four 160 MM Btu/hr slagging combustor operating at a high enough temperature to keep the ash molten and in an oxygen deficient mode to minimise NO<sub>x</sub> formation which would otherwise be increased due to the high operating temperature. Combustion is completed in a secondary burner; the sorbent is injected into the combustion products as they enter the host boiler. The sorbent is removed from the flue gas along with residual ash by bag house filtration. Essentially it is a staged combustion system to prevent excessive NO<sub>x</sub> formation, the SO<sub>x</sub> being removed by sorbent injection.

Further optimisation to improve process economics will be simultaneously studied at TRW's test site in Cleveland, Ohio where the 40 MM Btu/hr industrial boiler is already fitted with a slagging combustor system (SCS). It will be modified to demonstrate waste sorbent recycle to minimise fresh sorbent requirements and thus minimise Ca/S ratios. Flue gas humidification will also be studied as an approach to reduce sorbent consumption and maximise SO<sub>2</sub> capture. The project is estimated to cost US \$ 49,000,000 and is to be completed in three years.

Another method being developed is the direct injection of sulphur absorbing lime based chemicals in dry or slurry form into the existing duct work through which flue gases leave the coal fired boiler. Careful control of the moisture content in the

flue gases leads to the production of a dry particulate solid waste instead of a sludge. This solid waste can then be easily removed along with the flyash in existing particulate removal system. Removal of sulphur dioxide is, however, claimed to be only 50-70 percent at a total cost which is only about half of the conventional wet scrubbing (Anon., 1989).

### SULPHUR DIOXIDE ABSORPTION METHODS

Other methods centre around the absorption of sulphur dioxide in alkalies or sodium sulphite in aqueous solution or slurry and can be described by the following chemical reactions:

CaCO <sub>3</sub>	<del></del>	CaSO <sub>3</sub>
MgO	<del></del>	MgSO <sub>3</sub>
Na <sub>2</sub> SO <sub>3</sub>		NaHSO <sub>3</sub>

If limestone has been used, the resulting calcium sulphite is air oxidised to calcium sulphate and disposed off. The magnesium sulphite resulting from absorption in magnesium oxide suspension is roasted in a rotary kiln to regenerate magnesium oxide and the sulphur dioxide released is used for sulphuric acid manufacture. From the sodium bisulphite solution resulting from absorption in aqueous sodium sulphite (Wellmann-Lord Process), the sodium sulphite is regenerated by heat and concentrated in a forced circulation evaporator; the sulphur dioxide is used for sulphuric acid manufacture through the following chemical reactions:

CaSO <sub>3</sub>	<del></del>	CaSO₄
MgSO <sub>3</sub>	<del></del>	$MgO + SO_2$
2NaHSO <sub>3</sub>	<del></del>	$Na_2SO_3 + H_2O + SO_2$

The DESOX process is based on the oxidation of sulphur dioxide to sulphuric acid by bromine and electrolysis of the resulting hydrobromic acid to hydrogen and bromine. Hydrogen and sulphuric acid are sold and bromine is recycled:

$$SO_2 + Br_2 + 2H_2O$$
  $\longrightarrow$   $H_2SO_4 + 2HBr$ 

$$2HBr \longrightarrow H_2 + Br_2$$

The process is being currently tested in a power plant in Sardinia which produces 32000 m<sup>3</sup>/h flue gas with 5-10g/m<sup>3</sup> SO<sub>2</sub>. These are contacted with an aqueous

solution containing less than 1 percent bromine and about 10-20 percent sulphuric acid. The method is claimed to be cheaper by 20 percent than other sulphuric acid generating processes for SO<sub>2</sub> removal. Capital costs are estimated to be of the same order as other conventional wet scrubbing processes and the operating costs lower (Anon., 1989).

The SHELL Copper Oxide process uses sorbent supported copper oxide. The bed is regenerated by heating to about 400°C with hydrogen and steam to release sulphur dioxide in a concentration of 4-7 percent; this is subsequently increased to about 90 percent by stripping steam. The process is operating in a 40 MWe boiler in Japan.

Other processes use a variety of sorbents such as sodium carbonate on alumina or copper impregnated alumina spheres. Fluidised bed pilot reactors are also being tried with sorbents.

### **ELECTRON BEAM RADIATION**

Radiation with high energy electrons is quite a departure from the approaches so far outlined. Flue gas is bombarded with short and frequent pulses of electron beams in a pulsed energised electron reactor. Electrons convert sulphur dioxide to sulphur trioxide which is collected as sulphuric acid or reacted with a sorbent. These short electron beams consume less power than in the traditional electron beam approach.

## NITROGEN OXIDES FORMATION

 $NO_x$  formation, during coal combustion, can be derived from nitrogen of the air (thermal  $NO_x$ ) or the nitrogen present in the coal matrix (fuel  $NO_x$ ). As would be expected, the aliphatic compounds of coal nitrogen are rapidly expelled on heating and the aromatic nitrogen compounds are released only after fragmentation of high boiling tars and heterocycles. In pulverised coal flame, the residence time is not sufficient for complete devolatilisation and a substantial portion of the fuel nitrogen remains in the char. However, laboratory studies have shown, if sufficient time is given, all the nitrogen initially present in the coal can be removed from the char matrix, because, unlike carbon, nitrogen does not form stable structures at combustion temperature and the process is kinetically controlled. There is no apparent direct relationship between coal nitrogen content and the  $NO_x$  formed but coals with high volatile nitrogen content, when burnt in fuel lean conditions are more likely to produce high  $NO_x$  levels.

The mechanism of formation of oxides of nitrogen is not yet fully understood, but the nitrogen species are perhaps first converted into hydrogen cyanide and ammonia. Two pathways are then available: (i) reaction with oxygen to generate nitric oxide; or (ii) combining with nitric oxide itself to produce nitrogen. Char catalysed reduction of nitric oxide by carbon monoxide also takes place (Burdett and Pye, 1987) giving the following chemical reaction:

Coal 
$$N_2 \xrightarrow{\text{combustion}} HCN \qquad NH_3$$
processes  $NO$ 

The kinetics of this reduction are of interest. These have been measured in the temperature range 400-750°C, the effective diffusion coefficient was determined by fitting effectiveness factors in different particle sizes. Using these data the NO conversion by coal char under the conditions of fluidised bed combustor was estimated (Schuler et al., 1987). Another study has shown that in the combustion of the residual coke, the degradation of coke and the formation of NO<sub>x</sub> are parallel. 80-90 percent of the fuel nitrogen is converted to NO<sub>x</sub>. A major portion of the NO<sub>x</sub> formed during pyrolysis is, however, reduced to elemental nitrogen. Investigations of the kinetics of the reduction of nitric oxide on char, in the temperature range 300-500°C, showed that both the reaction rate and reaction order are dependent upon particle size (van Heek and Muehlen, 1985). This assumes importance in fluidised bed reactors.

Flame structure has a strong effect on NO emission; measurements of gaseous species formation in a coal combustion test furnace have shown that with proper control of secondary air mixing wlith primary fuel/air jets, NO<sub>x</sub> exhaust gas levels could be reduced to 100 ppm; NO is produced in the oxidising flame zone which envelops the central reducing flame and is destroyed in the mixing interface between the two zones (Arashi et al., 1985).

In  $NO_x$  formation, the reverse reaction is the thermodynamically favoured one, but is practically prevented by the rapid cooling of the products, as follows:

$$N_2 + xO_2$$
  $\longrightarrow$   $2NO_x$ 

Staged combustion is thus a method of minimising the formation of oxides of nitrogen. A reference to staged combustion was made earlier when dealing with TRW's slagging combuster system. In laboratory test furnaces, it has been possible to achieve nitric oxide concentrations as low as 100 ppm or even less. Oxides of nitrogen are potent sinks for odd oxygen species  $O_3$ ,  $O(^3P)$  and  $O^1D$ ), via the following chemical reactions:

This catalytic cycle (30-45 km altitude) destroys two odd oxygen species each time it occurs. Of course, nitric oxide also helps the production of ozone via the following reactions:

$$HO_2 + NO$$
  $\longrightarrow$   $NO_2 + OH$   
 $(2x) NO_2 + hv$   $\longrightarrow$   $NO + O(^3P)$   
 $(2x) O(^3P) + O_2 + M$   $\longrightarrow$   $O_3 + M$ 

(where M is another molecule of oxygen or nitrogen that is unchanged.)

## Nitrogen oxides removal

The NOXOSO process (Fig.1) is a promising one for the simultaneous removal of SO<sub>x</sub> and NO<sub>x</sub> from fuel gas. The sorbent is an alumina substrate loaded with sodium aluminate. The process works at about 250°F i.e. at normal power plant flue gas temperature. An advantage is that particulate removal upstream of the absorption is not required and a cyclone suffices to knock down the large particulate flyash to prevent its entrainment later in the fluidised bed. The spent sorbent is heated with combustion gas in a multistage bed to about 1000°F. This decomposes and releases the products of NO<sub>x</sub> absorption as nitrogen, oxygen and nitric oxide; these automatically mix with the combustion air and are fed to the boiler. The regeneration of the sorbent is then effected by feeding the hot sorbent to a countercurrent fluid bed generator where a reducing gas like methane reduces the sulphur dioxide to a 1:2 molar mixture of sulphur dioxide and hydrogen sulphide, which is then fed to a Claus unit to make elemental sulphur. The process is completely dry and produces no sludge.

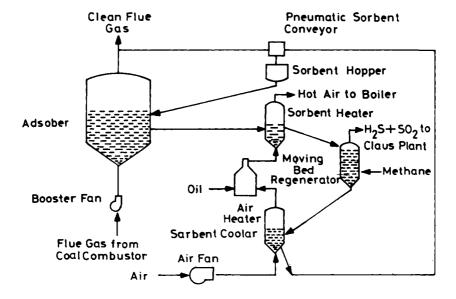


Fig. 1. Schematic diagram of the combined NO<sub>x</sub> and SO<sub>2</sub> reduction (NOXOSO) process.

The process has been tested on a small scale at the Tennessee Valley Authority's Shaunee facility and at the U.S. Department of Energy's Pittsburgh Energy Technology Research Centre (PETC). The test facility at PETC was equivalent to 0.75 MWe and the fluidised bed had a cross section of 1m² with variable expanded heights. The sorbent used was 3-8 percent sodium aluminate coated on a gamma alumina substrate. The sulphur dioxide removal efficiency was about 95 percent and NO<sub>x</sub> removal averaged 80 percent.

The NOXOSO process is being tried at a 5 MW coal fired unit at an Ohio Edison plant in Toronto, Ohio. A consortium of NOXOSO Corporation, M.W. Ferguson, W.R. Grace, Ohio Edison, the State of Ohio and PETC is funding the project. The 1984 estimates placed the cost of NOXOSO units at US \$ 162/kw of installed capacity (Wuebbles *et al.*, 1989).

## Environment friendly coal conversion technologies

There are several environmentally acceptable technologies now available for generating electricity from coal. These are:

- (i) Fluidised bed combustion— atmospheric (AFBC) (bubbling bed), circulating fluidised bed (CFB); and pressurised (PFBC);
- (ii) Gasification—IGCC and underground; and
- (iii) Partially solvent refined coal.

## Atmospheric fluidized and pressurized bed combustion

Atmospheric fluidised bed combustion is now a well established technology for steam/power generation with numerous plants in operation and several international technology vendors to choose from. The year 1987 was regarded as the watershed year for the scale up of fluidised bed boilers in the United States, with boilers as large as 900,000 lb/hr on order. The emphasis has shifted to pressurised from atmospheric fluidised bed combustion and also PFBC based combined cycle operations (Fig. 2).

## Circulating fluidized bed combustion

Circulating fluidised bed (CFB) technology has also penetrated many different industries such as pulp and paper, food processing and cement. CFB is also a fluidised bed combustion technology, but in contrast to the bubbling bed it employs much higher fluidisation velocities. In a bubbling bed, the solids should not be elutriated from the bed into the convective passes, so the fluidisation velocities are kept between 5-12 feet per second. The very purpose of CFB is to promote the elutriation of the solids and, therefore, fluidisation velocities can be as high as 30 feet per second. CFB permits longer residence time, increases combustion efficiency and improves the absorption of sulphur dioxide. All fluidised bed combustions are environmentally

#### GAS CLEANING GAS TURBINE GENERATOR COMBUSTION GAS CLEAN - POWER GAS **EXHAUST** COMBUSTOR HEAT EXCHANGER GAS ASH HOT B.F.WATER STACK AIR COAL **GENERATOR** STEAM TURBINE AIR STEAM **ASH EXHAUST** STEAM B. F. WATER CONDENSOR -

## P.F.B.C. COMBINED CYCLE POWER PLANT

Fig. 2. PFBC Combined cycle power plant.

friendly, the oxides of sulphur being removed by adding limestone to the bed; the formation of oxides of nitrogen is minimised because of the lower temperatures of combustion and 'staged combustion', as already mentioned.

## Integrated gasification combined cycle

In conventional power plants, the high pressure steam from coal combustion—pulverised coal or stoker type—is expanded through steam turbines to produce electricity. In PFBC, the hot combustion gas, after being cleaned, is expanded through a gas turbine and the hot exhaust gases from the gas turbine preheat the boiler feed water/raise steam. Steam for the steam turbine is raised by steam coils placed in the PFB combustor.

In Integrated Gasification Combined Cycle (IGCC), most of the ash is discharged from the gasifier and the gas is much easier to clean by scrubbing. In most processes, the sensible heat is recovered from the raw gas before cleaning and combusting and the chemical heat (CO, H<sub>2</sub>, CH<sub>4</sub>) is utilised through the gas turbine, with the exhaust gases being again used for steam generation. Additional power is thus generated through steam turbines viz. Heat Recovery Steam Generator (HRSG).

The coal gasification technology is well established commercially and processes are classified as fixed bed, fluidised bed and entrained bed. To date the largest number of gasifiers are the Lurgi fixed bed, producing more than 2000 MM Scf/day medium Btu gas. This process has been modified in collaboration with British Gas so that ash is removed in molten form and not as dry ash—British Gas Lurgi (BGL). A commercial plant is yet to be built.

Fluidised bed gasification technology is at a large PDU scale with only one commercial plant gasifying about 730 tpd of lignite at Berrenrath near Cologne, FRG, by the high temperature Winkler process (HTW). The gas is converted into methanol. At PDU scale, are the IGT Chicago and KRW Pittsburgh plants. The entrained bed processes can be of wet or dry type. The wet process was developed by Texaco and is now operative in Japan (UBE—1350 t/NH<sub>3</sub> per day), Federal Republic of Germany (Oberhausen-Holten) USA (Eastman Kodak 900 t/coal per day) (Cool Water 120 MWe electricity combined cycle).

Dow Chemicals have licensed this process from Texaco and have substantially modified it to gasify about 2000 tpd of lignite to generate about 165 MW power in Plaquemin, Louisiana, USA.

Plants at PDU stage using dry entrained bed are the Shell 200 tpd plant near Houston, USA. and the Prenflo 50 tpd plant at Fuerstenhausen, FRG. A 1000 tpd Prenflo plant was to be built (Krupps-Koppers Technology).

## **Underground Coal Gasification**

This technique was first studied in the USSR at depths of up to about 400 m. Deep underground gasification (1 to 2 kms) is being intensively studied in USA, FRG, France, Denmark and also in India. Two boreholes are drilled and connected underground by horizontal drilling. Air/oxygen and steam are introduced through one borehole; gasification takes place *in situ*, and the gas flows out through the other borehole.

## Partial solvent refining of coal

This technique is being studied in Japan, Peoples Republic of China and India. In the Indian programme (Central Fuel Research Institute), finely powdered coal in a suspension of anthracene oil is hydrogenated at about 400°C and 80 kg/cm²; weak linkages are broken and the coal is solubilised. After filtration from sedimented ash, the solvent is distilled off for reuse and the pasty reconstituted coal can be blended with coal for steel making.

### CONCLUSIONS

To sum up, increased use of coal need not lead to increased pollution. The unanswered question is whether poorer countries having adequate coal reserves and needing more electricity for their development can afford the expense (or luxury?) of antipollution technologies when even a beginning has not been made to monitor the environment, much less to prevent/minimise pollution.

#### REFERENCES

Anon. (1987). Chem. Engg. **94**(16), 21. Anon. (1989). Chem. Engg., **96**(2), 23.

- Arashi, N., et al. (1985). Proceedings of International Conference on Coal Science, Sydney. Pergamon Press, Sydney. p. 445.
- Burdett, N.A. and Pye, J.W. (1987). The effect of coal combustion on No<sub>x</sub>-formation, during combustion under controlled conditions. International Conference on Coal Science, Maastricht, J.A. Moulin (Ed.). Elsevier, Amsterdam, pp. 845-852.
- Makanse, J. (1987). Special Report, Fluidised Bed Boilers, Power, May 1-16.
- Mintzer, I.M. (1987). A Matter of Degrees: The Potential for Controlling the Greenhouse Effect. World Resources Institute, Washington D.C.
- Miller, J.A. and Fisk, A. (1987). Special Report in Combustion Chemistry. C & E News, 65(35), 22.
- Schouter, J.C., et al., (1987). International Conference on Coal Science, Maastricht, J.A. Moulin (Ed.). Elsevier, Amsterdam, pp. 837-841.
- Schuler, J., Baumann, H. and Klein, J. (1987). International Conference on Coal Science, Maastricht, J.A. Moulin (Ed.). Elsevier, Amsterdam, p. 857.
- van Heek, K.H. and Muehlen, J.H. (1985). Brennstoff Waerme Kraft, 37, 20-28.
- Wilson, C.L. (1980). Coal—Bridge to the Future. Report of the World Coal Study, M.I.T. Ballinger Publishing Co., Cambridge, Mass., USA.
- Wuebbles, D.J., Grant, K.E., Cornell, P. S. and Penner, Joyce E. (1989). The role of atmospheric chemistry in climate change, J. Air Pollut. Contr. Assoc., 39(1), 22-28.
- Zang, S.D., et al. (1989). Sulphur pollulant formation during coal-combustion Fuel, 68(3), 346-353.