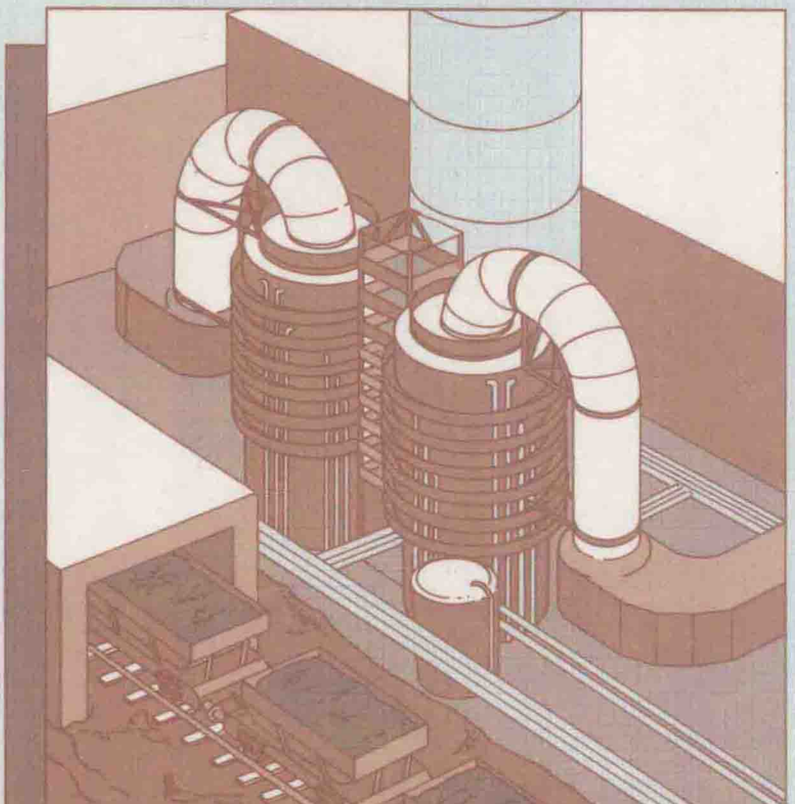
Desulphurisation 2

Technologies and Strategies for Reducing Sulphur Emissions



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Technologies and Strategies for Reducing Sulphur Emissions

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A two-day symposium organised by the Institution of Chemical Engineers in conjunction with the Institute of Energy and held at the University of Sheffield, 20–21 March 1991.

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Errata

Paper 1 (page 1): FGD installations on coal-fired plants (A. Hjalmarsson).
In Table 3 on page 9, the line giving data on the United Kingdom should read:
United Kingdom ..... – ..... 8·0 ..... 8·0
and the line giving totals should read:
Total ..... 40·1 ..... 87·5 ..... 127·6

Paper 7 (page 205): Factors influencing the Drax flue gas desulphurisation process scrubbing system configuration (J. R. Cooper and I. A. Johnston).
Please note that in this paper pages 207 and 208 were inadvertently printed in the wrong order. Page 208 should be read before page 207.

Paper 15 (page 143): Technical and environmental implications of desulphurisation by sea water washing (R. Baty, J. Coughlan and S. K. Reynolds).
(i) Page 143: J. Coughlan's affiliation should read "National Power", not "Powergen".
(ii) Page 150: paragraph 2, line 6, "Figs 5 & 6" should read, "Figs 4 & 5".
(iii) Page 151: final paragraph, line 6, "Fig N" should read, "Fig 6".

Preface

Growing concern about the environment and the role that sulphur dioxide plays in the acidification has led to national and international legislation to limit the emissions of sulphur dioxide. A substantial number of countries have legislation limiting specific emissions from new plant resulting in the use of some form of desulphurisation technology. Legislation has been in place in countries such as the USA, Japan and Germany for a considerable period.

In the last decade the emphasis has been on reducing total emissions including those from existing plant. This has led to international agreements such as the "30% Club" protocol arising from the UN-ECE Convention on Long-Range Transboundary Air Pollution and the EC Large Combustion Plants Directive. Both these require specified percentage "bubble" reductions from individual countries to given timescales. Both the US, through the Clean Air Act, and Canada are requiring reductions in total emissions.

The first International Desulphurisation Conference was held in Sheffield in 1989 against the backdrop of the passing of the EC Directive on the Limitation of Emissions from Large Combustion Plants. Since then, the groundswell of public interest in environmental matters has continued to grow and work on emission limitation has continued apace. The second conference, in 1991, whose proceedings are reported in this publication, was held soon after the passing of the Environmental Protection Act in the UK, amendments to the Clean Air Act in the US and the publication of the UK National Plan for reducing sulphur dioxide and nitrogen oxide emissions. Both conferences have therefore been most timely.

The aim of the Conference is well summarised in its subtitle — "Technologies and Strategies for Reducing Sulphur Emissions". Reflecting this the programme covers:

- Strategies and technologies for reducing SO_2 emissions;
- Waste disposal;
- Future developments.

More than half the papers were presented by non-UK authors from seven countries with attendees coming from 15 different countries, making this a truly international conference.

W. S. Kyte
(Chairman)

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FGD INSTALLATIONS ON COAL-FIRED PLANTS

Anna-Karin Hjalmarsson*

As regulations for sulphur dioxide emissions have become progressively more stringent during the 1980s, an increasing number of coal-fired plants have been equipped with flue gas desulphurisation (FGD) installations to comply with the regulations. Coal-fired units with a total capacity of about 147 GWe have already been equipped with FGD worldwide. The capacity is likely to increase significantly as emission control regulations are implemented, probably reaching 260-280 GWe by the end of the century.

INTRODUCTION

The number of coal-fired plants that are equipped with flue gas desulphurisation (FGD) is increasing as the requirements to limit sulphur emissions are being introduced in more and more countries.

Limitation of SO_2 emissions are imposed through international and national regulations. There are currently national emission standards in about 20 countries with Poland and the former GDR being the most recent countries to introduce regulations on emissions of sulphur from coal combustion. The stringency of emission standards in combination with the uncontrolled concentration of SO_2 in the flue gases determines whether any measures are needed to control the SO_2 emissions. Several options are available and they vary depending on combustion technology, fuel properties, whether a plant is already there or newly built and the emission standards applicable for SO_2.

The main alternatives to control sulphur emissions are:

- decreasing the sulphur content in the fuel by fuel switching or coal cleaning;
- the use of processes that decrease emissions from the combustion system;
- the use of processes that decrease emissions from the flue gases.

There are several technologies available for each alternative, resulting in different emissions of SO_2 depending on the conditions. This paper outlines briefly regulations on sulphur emissions and presents the worldwide use of FGD based on the IEA Coal Research in-house data base on installations of FGD and NO_x control on conventional coal-fired plants. Combustion technologies also exist where the control of emission is an integral part of the process, but these are not included in the data base. Fluidised bed combustion is an example of this combined approach.

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REGULATIONS LIMITING SULPHUR EMISSIONS

Recently there have been significant developments in sulphur control legislation, both nationally and internationally.

There are various types of regulations on sulphur emissions. They can be based on the emission of sulphur, the sulphur removal rate, sulphur content in the coal or the requirement for control technology. They can be used separately or in combination. National regulations based on sulphur emissions are the most common type. The unit used for emission standards varies between countries so it is hard to draw comparisons. The main unit used is mg SO_2/m^3 at specified temperature, pressure and oxygen content (generally O°C, 1.1013 bar on a dry gas basis and with a specified oxygen content, normally 6% for coal). In some countries the emission is based on the energy input, such as g S/MJ, g SO_2/MJ and lb SO_2/MBtu. These units can, however, be based on either the efficient (net) heat value, as in Scandinavian countries, or the calorimetric (gross) heat value, as in the USA. Standards are also set on different time bases, that is averaged over a year, a month, or an hour. The categorisation of standards applicable for a plant can be based on unit size, whether it is a new or an existing plant, combustion technology, total amount of sulphur emission and type of fuel.

International agreements

The United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution was signed in Geneva in 1979 by 33 countries. In 1985, 21 countries signed a protocol to the convention which called for a 30% reduction in total national emissions of SO_2 by 1993, compared to 1980 levels. This protocol has become known as the "30% club".

The EC Directive on the limitation of emissions of pollutants into the atmosphere from large combustion plants was agreed in 1988, and applies to twelve European states. The directive specifies country by country target reductions in aggregate emissions from existing combustion plants compared with 1980 emission levels. It covers combustion plants having a thermal input of more than 50 MWt. Existing combustion plants are defined as those in place before July 1987.

The targets for existing plants will give a 23% reduction in EC total emissions by 1993, a 42% reduction by 1998 and a 57% reduction by 2003 compared to 1980 baseline figures. To achieve this total reduction for the EC, different targets are set in each country. Some countries such as Greece, Ireland and Portugal are allowed an increase in emissions. The most stringent reduction targets are set at 70% for Belgium, the FRG, France and the Netherlands. For new plants smaller than 500 MWt, there is a sliding scale of 400-2000 mg SO_2/m^3 and for plants over 500 MWt the standard is 400 mg SO_2/m^3, Commission of the European Communities, 1988 (1).

National regulations

There are currently national emission standards in about 20 countries with Poland and the former GDR being the most recent countries to introduce regulations on emissions of sulphur.

National emission standards apply to new and existing coal-fired plants; these are summarised in Table 1. Increasingly these standards are being applied retrospectively to existing plants as well; this is the case in Austria, the FRG, Finland, Italy, Sweden and Japan. For smaller plants, controls over SO_2 usually take the form of less stringent emission standards, or a limit on the sulphur content in the fuel they burn. Further extensions of legislation can be expected in the near future as countries strive to meet their international obligations and further reduce total SO_2 emissions.

Other coal-using countries have not yet adopted targets for reducing SO_2 emission. Instead some have strategies based on meeting air quality standards or preventing deterioration of air quality. Taxes and charges have also been introduced on sulphur emissions in some countries, see Vernon (2).

FGD PROCESSES FOR COAL-FIRED PLANTS

The choice of FGD systems depends on several conditions which can vary between plants but also between countries resulting in different processes being used. Some of the conditions that affect the choice of FGD systems are:

- coal properties, mainly sulphur content;
- regulations on SO_2 emissions, disposal of residuals and waste water;
- plant operation, for example load factor;
- plant size;
- new or retrofit installation;
- site specific conditions;
- economic assumptions and cost accounting procedures.

The main principles for SO_2 removal processes are shown in Figure 1, see Klingspor and Cope (3).

Sorbent injection processes involve gas/solid reactions. A dry calcium or sodium based sorbent is injected into the boiler or, the flue gas duct, or a combination of these methods. These processes generally produce a dry sodium or calcium sulphite/sulphate waste mixed with the fly ash. This approach is interesting mainly because of its low capital costs relative to other FGD processes. However, the efficiency of sulphur capture is considerably lower than other systems. Large sorbent to sulphur ratios are needed to achieve SO_2 removal efficiencies above 50%. The efficiency can be increased by the use of specially prepared sorbents, or a hydration step within the ductwork or in a separate reaction vessel. An example of this is the Lifac process where limestone is injected into the boiler and water is later injected into the flue gases to reactivate the unreacted sorbent and a reaction vessel is used. Another process type sorted under sorbent injection in the data base is a process using a circulating fluidised bed with lime or hydrated lime as the sorbent. These latter processes can reach reduction rates up to 70-90%.

Spray dry scrubbers have been developed as an alternative to wet scrubbers especially for smaller sized installations and where the sulphur content in the coal is not particularly high. The sorbent slurry or solution is sprayed in fine droplets into the reactor vessel, drying as it reacts with the SO_2 in the flue gases. An end collector cleans the flue gases from the produced dry particles. Lime slurry is commonly used as a sorbent, resulting in a dry product of mixed calcium sulphite/sulphate. Sodium carbonate solution can also be used as a sorbent resulting in sodium sulphite/sulphate as the by-product. The SO_2 removal is commonly around 80-90%.

The most widely used type of FGD installations are **wet scrubbers**. The processes include a gas/liquid reaction and result in a wet product. The majority of wet scrubbers use a calcium-based slurry as a sorbent, generally of lime or limestone. Sodium based sorbent can also be used. The sorbent is injected into the flue gases, generally in an open tower. The residue of such processes is a wet mixture of calcium sulphite and sulphate. An oxidation step can be incorporated into the scrubber to produce gypsum, which can be dried. The first generation wet scrubbers were designed to use lime as a sorbent, but limestone has become more common in recent years. Processes producing gypsum are increasingly being favoured. Wet scrubbers are used with a wide variety of coal types on installations up to the largest sizes for coal-fired units. Sulphur removal efficiencies of 90% and over are normally achieved.

Dual-alkali wet scrubbers are used less widely. Here a sodium-based solution absorbs SO_2 from the flue gases and the reaction products are regenerated in a second step using lime or limestone for the final SO_2 capture.

Other types of wet scrubber in use on a small number of coal-fired plants employ either aqueous ammonia as a sorbent, giving a fertiliser by-product, or carbide sludge. Another process needs a coastal location to use the natural alkalinity of seawater as a sorbent. In this process large volumes of seawater are needed.

A range of **regenerable** FGD systems are in use, although the total number of installations on coal-fired plants is small. Regenerable processes are processes where the sorbent for SO_2 can be re-used following a regeneration step which releases concentrated sulphur dioxide. The sulphur dioxide can then be further processed to either liquid SO_2, sulphuric acid or elemental sulphur. Regenerable systems tend to be more complex than non-regenerable processes, and have higher capital costs. Their overall economics are highly dependent upon the price obtained for the end product. The most widely used regenerable FGD system is the **Wellman-Lord** process, which uses a gas/liquid reaction with a sodium sulphite solution as the sorbent. The reaction product is regenerated by heat treatment to produce an SO_2-rich gas that can be further processed to give sulphuric acid or elemental sulphur. Another regenerable process uses **magnesium oxide** slurry as a sorbent. As with the Wellman-Lord process, the sorbent is thermally regenerated producing SO_2-rich gas.

There are also **combined SO_2/NO_x** processes where the flue gas desulphurisation is combined with a denitrification step. An example is the activated carbon process where SO_2 is adsorbed on activated carbon. Ammonia is injected into the flue gases in a mixing chamber before the flue gases enter the second stage. In the second stage NO_x reacts catalytically with ammonia to form N_2 and H_2O. Desorption of the sulphur-laden carbon is performed thermally. Another combined SO_x/NO_x method is catalytic absorption, two processes exist; the German DESONO$_x$ process and the Danish SNOX process. The first step is a deNO$_x$ step with selective catalytic reduction and a catalyst is used to convert SO_2 to SO_3. The flue gases are cooled and the SO_3 is hydrated to sulphuric acid by water vapour contained in the flue gases, see Hjalmarsson (4).

FGD INSTALLATIONS WORLDWIDE

The first FGD installations on coal-fired plants were completed in the early 1970s in the USA and Japan. A major expansion of FGD use has taken place since then, with installations currently operating in fifteen countries and under construction or planned in further countries. By the end of 1990 there were about 540 boilers equipped with FGD plants, equal to a total installed capacity of about 147 GWe. 137 GWe of these are in Japan, the FRG and the USA, see IEA Coal Research (5), Vernon and Soud (6). Figure 2 shows the growth in number of boilers that have been equipped with FGD worldwide.

Figure 3 gives the growth in installed capacities of different FGD processes over time. It can be seen in the figure that the interest in producing usable residues has been increasing rapidly and the limestone/gypsum processes are now installed at approximately the same capacity as other wet systems. The capacity of wet scrubbers producing non-usable residues are levelling off after being the most common system in the 1970s and the beginning of the 1980s. Most of the recent growth in installed capacity of wet limestone scrubbers producing gypsum is in the FRG where the new emission standards have had to be met. There has been growth in the total capacity of installed spray dry scrubbers since they were introduced in the early 1980s - they are, though, in general used on smaller units so the total capacity is still not large. Sorbent injection is not used that widely and mostly also on smaller units. The use of regenerable processes has grown very slowly.

Table 2 shows the installed capacities of FGD systems for separate countries. The largest installed capacity, about 82 GWe, is in the USA, where the most common method is wet scrubbers producing a calcium sulphate/sulphite sludge. The FRG has the second largest capacity, about 42 GWe, mostly limestone wet scrubbers producing gypsum. The third largest capacity is in Japan with about 13 GWe. The wet limestone gypsum process is totally dominant in Japan. The rest of the countries have an installed capacity of less than 2 GWe each. In comparing these three countries it can be seen that the conditions in the countries favour different FGD systems. Wet scrubbers used in the USA produce a sludge that is disposed of as there are fewer regulations on residues and more space is available for disposal sites. On the other hand strict controls over waste disposal in the FRG and Japan have favoured processes producing a saleable gypsum residue.

A small percentage (about 2%) of the total install FGD capacity is sorbent injection. It is in operation in eight countries, mainly where the required reduction rate is limited. Several plants using this process are burning brown coal or lignite and most installations have been retrofitted to smaller boilers. Hybrid versions of sorbent injection are also included, like the Lifac process and circulating fluidised beds with lime or hydrated lime. The former process is operating in Canada, Finland and the USSR and the latter in the FRG.

Spray dry scrubbers are used in about 9% of the total installed FGD capacity and are operating in six countries. Spray dry scrubbers are installed on both new and existing plants, with equal capacities. The boiler sizes are larger than for sorbent injection and high reduction rates are achieved.

Seven countries use wet limestone scrubbers producing gypsum, covering about 40% of the total FGD capacity. Very high reduction rates are obtained for a variety of boiler sizes and coal types.

Other wet processes consist mainly of processes using limestone as the sorbent and produce a sludge. The majority of installations are in the USA. Two wet scrubbers were supposed to start operating last year in China. Other wet processes include the dual alkali process, with an installed capacity of 2.5 GWe in the USA. Other wet processes are installed on about 40% of the total FGD capacity.

The largest capacity of regenerable processes is in the USA, where the Wellman Lord and the MgO processes are used. In the FRG there is one plant using the Wellman Lord process (in the former GDR) and 3 units with combined SO_2/NO_x reduction using the activated carbon process.

The capacities for both firmly planned plants and estimates for the future are listed in Table 3. The largest number of FGD installations in the near future will be in the USA, where the new Clean Air Act that was introduced at the end of last year has to be met. It is estimated that by the year 2000 a further capacity of about 50 GWe will have been retrofitted according to Dalton (7) and installations on 26 GWe are planned for new boilers.

There are plans in Italy and the UK to install FGD on about 9 GWe in each country. In the eastern republics of the FRG (the former GDR), the new emission standards have to be met by the end of 1996 for existing plants. The capacity that will be retrofitted with FGD depends on how many of the old coal-fired plants will be closed down. An estimate for FGD retrofit capacity is 4 GWe. The first FGD in India will start in 1992 using the sea water scrubbing method. There are plans to install FGD on a capacity of about 20 GWe in Bulgaria, Poland, Hungary, Czechoslovakia and Yugoslavia according to Couch et al (8). However, the major problem will be finance. The estimates show that by the end of the century there will probably be a total global installed capacity of FGD of between 260-280 GWe.

CONCLUSIONS

The application of controls over sulphur emissions from coal fired plants is not new, but the stringency of regulations and the range of plants to which they apply has increased dramatically in the 1980s.

The most commonly used FGD system, wet scrubbing, mainly based on limestone producing either saleable gypsum or a sludge for disposal, is now well developed and used in several countries. The increasing stringency in regulations for residues and wastewater may, however, lead to further development and use of regenerable processes with emphasis on processes for combined reduction of both sulphur and nitrogen oxides.

An important consideration in the choice of emission control systems is to avoid producing new pollutants, either by depositing wastes that have a potential to leach, producing contaminating waste water, or increased CO_2 emissions due to use of limestone and higher energy use in operating the flue gas treatment plant. In the future, total optimisation of the whole power plant will become essential, taking plant efficiency as well as emission control into account, and not just dealing with one pollutant at a time.

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Table 1 Current national emission standards for sulphur emissions (mg SO_2/m^3)(9)

Country	New plants	Existing plants
Austria	200-400	200-400
Belgium+	400-2000(250)*	
Canada#	740	
Denmark+	400-2000	
Finland	400-660	660
France+	400-2000	
FRG+	400-2000	400-2500
Greece+	400-2000	
Ireland+	400-2000	
Italy+	400-2000	400-2000
Japan	$	$
Luxembourg+	400-2000	
Netherlands+	200-700	400-700
Poland	540	2700-4000 (1800-3000)
Portugal+	400-2000	
Spain+	2400-9000	2400-9000
Sweden	290	290-570
Switzerland	400-2000	400-2000
Taiwan	2145-4000	2145-4000
Turkey	400-2000	
UK+	400-2000	
USA	740-1480**	**

# guidelines
$ set on a plant-by-plant basis according to nationally-defined formulae
+ EC countries
* from 1995
** Clean Air Act Amendments, 1990

Table 2 Installed capacity (MWe) of FGD systems in different countries by the end of 1990 (5)

Countries	Sorbent injection	Spray dry scrubbers	Limestone gypsum	Other wet processes	Regenerable processes	Not Known	Total
Austria	315	755	690	100			1860
Canada	300						300
China				700			700
Czechoslovakia					*(200)		*(200)
Denmark		700	500			370	1570
FRG	760	2920	35150	800	1770	470	41870
Finland	280	260					540
France	600						600
Italy					30		30
Japan			13680	200			13880
Netherland			2730				2730
Sweden	530	550		10			1090
Turkey			340				340
USA	550	7600	6510	57970	3010	6090	81730
USSR	45						45
TOTAL	3380	12785	59600	59780	4810	6930	147285

* not in operation