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Project Summary

SO_2 Abatement for Coal-Fired Boilers in Japan

Jumpei Ando

Strict ambient air quality standards for sulfur dioxide (SO_2) and nitrogen oxides (NO_x) in Japan mandate the use of various air pollution control technologies. This report is a compilation of information on the current status of SO_2 abatement technologies for coal-fired boilers in Japan. It focuses on flue gas desulfurization (FGD) and is based on information gathered from utility company representatives and FGD process developers as well as the author's research in the field. Various technologies including wet lime/limestone and indirect lime/limestone FGD processes and combined FGD/Selective Catalytic Reduction (SCR) systems are described in terms of process descriptions, development and/or commercial application status, and economics. Detailed operation data are given for many of the processes.

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

Introduction

Strict ambient air quality standards for sulfur dioxide (SO_2) and nitrogen oxides (NO_x) in Japan mandate the use of various air pollution control technologies. This report is a compilation of information regarding the current status of SO_2 abatement technologies for coal-fired boilers in that country. Many of the technologies either have been or can be applied in the U.S.

The total capacity of coal-fired utility boilers in Japan is now 4,300 MW (3.7% of total utility power) and is expected to reach 10,000 MW (5.6%) in 1985 and 22,000 MW (10%) in 1990. Several coal-fired industrial boilers also may be constructed in the future. All of these boilers will apply some type of flue gas desulfurization (FGD) to reduce SO_2 emissions.

Overviews

In sequence, the following paragraphs: (1) give an overview of FGD use for all types of boilers in Japan; (2) focus on the wet lime/limestone process, the most common FGD process applied to coal-fired boilers; (3) describe other FGD processes for coal-fired boilers, including several modified wet lime/limestone processes and a number of dry processes; (4) discuss simultaneous removal of both SO_x and NO_x as yet another approach to SO_x abatement presently used in Japan; and (5) describes other coal utilization technologies (e.g., gasification, liquefaction, and fluidized-bed combustion) which have been tested as methods of reducing SO_2.

Flue Gas Desulfurization

FGD has been widely used for SO_2 abatement in Japan and is considered to be an accomplished technology. Over 1,200 commercial FGD units for various gas (40,000 MW equivalent) of flue gas to remove 85-95% of SO_2. The principal byproducts of FGD are gypsum, sodium sulfite, and sulfuric acid. Table 1 summarizes the approximate number and capacities of FGD units now in use in Japan.

Sodium scrubbing, producing sodium sulfite or sulfate byproducts, is the simplest FGD system. The sulfite can be sold to paper mills, and the sulfate can be used for glass production. Over 700 sodium scrubbing units with capacities of 3,000 - 300,000 Nm^3/hr (1-100 MW equivalent) were constructed in Japan between 1968 and 1974.

Table 1. Approximate Number and Capacity of Japanese FGD Units Presently in Operation

Process	Number	Capacity (1,000 Nm^3/hr)
Sodium	1,000	50,000
Wet lime/limestone	110	55,000
Indirect lime/limestone	50	15,000
H_sSO_4, S, $(NH_4)_2SO_4$ byproduct	30	12,000

Most Japanese utility boilers utilize wet lime/limestone FGD scrubbing which produces salable gypsum. This type of FGD is relatively inexpensive and produces gypsum which can be used for cement setting retarder and wallboard.

About 50 indirect lime/limestone FGD process units are in use in Japan. The indirect process is used mainly for medium-sized gas sources with capacities of 50,000 - 500,000 Nm^3/hr since scaling problems for units of this size can be prevented easily. However, it is more expensive than the lime/limestone process. Another 20 FGD units are presently producing a sulfuric acid byproduct. Most of these units use the Wellman-Lord process to recover concentrated SO_2 for use in acid production.

Two large Japanese ammonia scrubbing units use ammonia in coke oven gas to recover SO_2 from boilers for ammonium sulfate production. These recovery processes are not used extensively at power plants because sulfuric acid or sulfur production processes are more costly than limestone-gypsum processes, and ammonium sulfate is currently in oversupply.

Almost all Japanese FGD units use wet processes. Dry processes were studied in the early stages of FGD development, but most of them have not been used because these early investigations showed them to have low SO_2 removal efficiencies and high cost. Only four dry process units are in operation in Japan: three use carbon adsorption, and one uses copper oxide absorption. A few units use a semi-dry process in which an alkaline slurry is sprayed and a powdery sodium sulfite is recovered. A semi-dry process being developed in the U.S. involves spraying lime slurry to obtain a powdery calcium sulfite. This method probably will not be used in Japan because of difficulties with byproduct disposal and with attaining a high SO_2 removal efficiency.

FGD processes available in Japan for use with coal-fired units are listed in Table 2. Most of them are wet processes which use lime or limestone reagents; one process uses red mud. In addition, three dry processes use activated carbon or electron beams and have been tested for coal-fired applications. The wet lime/limestone process is described below; the other wet and dry FGD processes are discussed later.

Wet Lime/Limestone FGD Processes for Coal-fired Plants

Almost all Japanese coal-fired boilers with capacities greater than 250 MW use the wet limestone FGD process to remove 90-95% of SO_2. The outlet SO_2 concentration for these systems is typically 20-150 ppm.

Table 3 lists all commercial FGD units in use or planned for coal-fired boilers in Japan. All units utilize wet lime/limestone scrubbing and, except for Mitsui Aluminum's two units for industrial boilers, all are applied to utility boilers. Figure 1 illustrates the four types of lime/limestone processes used in the FGD units.

Type I is the throwaway sludge system commonly found in the U.S. Only one Japanese unit is of this type. Type II processes lower the pH to about 5 by using three scrubbers and a catalytic material for oxidation. The Type II system is used in three units. Type III systems have two-stage scrubbers and lower the pH of the slurry from about 5 to about 4 by adding a small amount of sulfuric acid. This type requires a lower investment cost than does a Type II process, and may be preferable where sulfuric acid is available for a reasonable cost. The Electric Power Development Corporation (EPDC) has four System III units.

The Type IV FGD process has a prescrubber and uses sulfuric acid for pH control. The Type IV process was developed and has been used by Mitsubishi Heavy Industries (MHI) since the 1960s. Nearly all of the limestone scrubbing units

Table 2. FGD Processes for Coal-Fired Units

FGD Process Developer	Reagent	Byproduct
Mitsui Miike Machinery Co. (MMMC)[a]	$CaO/CaCO_3$	Sludge/gypsum
Ishikawajima-Harima Heavy Industries (IHI)[a]	$CaCO_3$	Gypsum
Babcock Hitachi K. K. (BHK)[a]	$CaCO_3$	Gypsum
Mitsubishi Heavy Industries (MHI)[a]	$CaCO_3$	Gypsum
Chiyoda Chemical Engineering and Construction (Chiyoda)[b]	$CaCO_3$	Gypsum
Kawasaki Heavy Industries (KHI)[a]	MgO and CaO	Gypsum
Dowa Mining (Dowa)[b]	Aluminum sulfate and $CaCO_3$	Gypsum
Kobe Steel[b]	$CaCl_2$ and CaO	Gypsum
Kureha Chemical Industries (Kureha)[b]	CH_3COONa and CaO	Gypsum
Sumitomo Aluminum[b]	Red Mud	Sludge
Federation of Electric Power Co.[c]	Activated Carbon	Sulfur
Electric Power Development Co. (EPDC) and Sumitomo Heavy Industries (SHI)[d]	Activated Carbon, NH_3	Sulfur
Mitsui Mining[d]	Activated Carbon, NH_3	Sulfuric Acid
Ebara Corp.[d]	NH_3 (Electron Beam)	Ammonium Nitrate sulfate

[a]Commerical.
[b]Tested with coal. Commercial operation with other gases.
[c]Tested for coal.
[d]Tested with coal for simultaneous SO_x and NO_x removal.

Table 3. Commercial FGD Units for Coal-Fired Boilers in Japan

Owner	*Plant Site*	*Boiler No.*	*MW*	*N/R*[a]	*FGD Constructor*	*Year of Completion*	*Type See Figure 1*
Mitsui Aluminum	*Omuta*	*1*	*156*	*R*	*MMMC*[b]	*1972*	*I*
Mitsui Aluminum	*Omuta*	*2*	*175*	*N*	*MMMC*	*1975*	*II*
EPDC	*Takasago*	*1*	*250*	*R*	*MMMC*	*1975*	*II*
EPDC	*Takasago*	*2*	*250*	*R*	*MMMC*	*1976*	*II*
EPDC	*Isogo*	*1*	*265*	*R*	*IHI*[c]	*1976*	*III*
EPDC	*Isogo*	*2*	*265*	*R*	*IHI*	*1976*	*III*
EPDC	*Takehara*	*1*	*250*	*R*	*BHK*[d]	*1977*	*III*[g]
EPDC	*Takehara*	*3*	*700*	*N*	*IHI*	*1982*	*III*[g]
EPDC	*Matsushima*	*1*	*500x3/4*	*N*	*IHI*	*1981*	*IV*
EPDC	*Matsushima*	*2*	*500x3/4*	*N*	*BHK*	*1981*	*IV*
Chugoku Electric	*Shimonoseki*	*1*	*175*	*R*	*MHI*[e]	*1979*	*IV*[g]
Chugoku Electric	*Shin-Ube*	*1*	*75*	*R*	*MHI*	*1982*	*IV*[g]
Chugoku Electric	*Shin-Ube*	*2*	*75*	*R*	*MHI*	*1982*	*IV*[g]
Chugoku Electric	*Shin-Ube*	*3*	*156*	*R*	*MHI*	*1982*	*IV*[g]
Chugoku Electric	*Mizushima*	*1*	*125*	*R*	*BHK*	*1983*	*IV*[g]
Chugoku Electric	*Mizushima*	*2*	*156*	*R*	*BHK*	*1983*	*IV*[g]
Hokkaido Electric	*Tomato-Atsuma*	*1*	*350x1/2*	*N*	*BHK*	*1980*	*IV*[g]
Hokkaido Electric	*Tomato-Atsuma*	*2*	*600*	*N*	*MHI*	*1984*	*IV*
Kyushu Electric	*Omura*	*2*	*156*	*R*	*MHI*	*1982*	*IV*
Kyushu Electric	*Minato*	*1*	*156*	*R*	*MMMC*	*1983*	*III*
Joban Joint	*Nakoso*	*8*	*600*	*N*	*MHI*	*1983*	*IV*[g]
Joban Joint	*Nakoso*	*9*	*600*	*N*	*MHI*	*1983*	*IV*[g]
Tohoku Electric	*Sendai*	*2*	*175*	*R*	*BHK*	*1983*	*IV*
Tohoku Electric	*Sendai*	*3*	*175*	*R*	*BHK*	*1983*	*IV*
Tokyo Electric	*Yokosuka*	*1*	*265*	*R*	*MHI*	*1984*	*IV*[g]
Tokyo Electric	*Yokosuka*	*2*	*265*	*R*	*MHI*	*1984*	*IV*[g]
Shikoku Electric	*Saijo*	*1*	*175*	*R*	*KHI*[f]	*1983*	*IV*
Shikoku Electric	*Saijo*	*2*	*250*	*R*	*KHI*	*1984*	*IV*

[a] *New or retrofit.*
[b] *Mitsui Miike Machinery Co.*
[c] *Ishikawajima-Harima Heavy Industries.*
[d] *Babcock Hitachi K. K.*
[e] *Mitsubishi Heavy Industries.*
[f] *Kawasaki Heavy Industries.*
[g] *Selective catalytic reduction of NO_x is also applied.*

constructed since 1979 use the Type IV process.

Table 4 shows the capacities, types of scrubbers, and operational parameters of the wet lime/limestone FGD units for coal-fired utility boilers in use in Japan.

Most of the new FGD units for coal-fired boilers either built since 1978 or under construction have a prescrubber with a separate liquor loop to remove impurities in flue gas such as fly ash, chloride, and fluoride. They also employ low pressure drop scrubbers and a rotating gas-gas heater (heat exchanger) for gas reheating and for water conservation.

All FGD units in Table 3 use limestone ground so that over 95% passes through 325 mesh at a stoichiometry of 1.00-1.05 to remove 94-97% of inlet SO_2. Gas reheating adds a small additional amount of SO_2 so that the overall SO_2 removal efficiencies are 90 - 95%. The dust removal efficiencies of the units are 70-90%.

The most significant operational problem associated with the lime/limestone FGD process has been the formation of gypsum scale in the scrubber and mist eliminator systems. In Japan these scaling problems have largely been solved, resulting in a reliability rate of more than 99% for lime/limestone FGD units.

Next to scaling, corrosion is the most serious problem associated with FGD systems. This problem has been solved by using suitable materials and good lining fabrications. With these measures FGD operations have become so reliable that not a single FGD unit in Japan has a stand-by scrubber, although most units have stand-by pumps and centrifuges.

A limestone FGD unit for a 500 MW coal-fired boiler requires 40-60 t/hr of water. Water is fed into the Type II and III process (Figure 1) scrubbers and into both the prescrubber and scrubber of Type IV. To prevent the accumulation of impurities such as chloride and fluoride derived from coal, 5-30 t/hr of wastewater must be purged. In a typical FGD wastewater treatment system, the wastewater from the prescrubber and scrubber is treated with lime to precipitate heavy metals and fluoride prior to filtration. The filtrate is treated to remove dithionate $S_2O_6^{=}$ which can cause a chemical oxygen demand (COD).

Treated FGD wastewater typically has a pH of 6.5-8.5 and contains less than 10-15 mg/l each of suspended solids and COD, less than 15 ppm fluoride, and less than 1 ppm oily material.

The estimated FGD cost for a 500 MW coal-fired boiler with a limestone-gypsum process unit and a gas-gas heater is shown in Table 5. The annualized FGD cost in 1981 was about 1.9 ¥/kWhr.* About 70% of the cost consists of the fixed cost with a 7 year depreciation schedule. When the annualized cost is based on a 15 to 20 year depreciation schedule, as is commonly done in the U.S., the annualized cost is considerably lower.

Table 6 shows a rough estimated cost for power generation at a 1000 MW plant with FGD, Selective Catalytic Reduction (SCR), and an electrostatic precipitator (ESP) for flue gas treatment. The estimated annualized cost of power generation for

(*) $1 = ¥250.

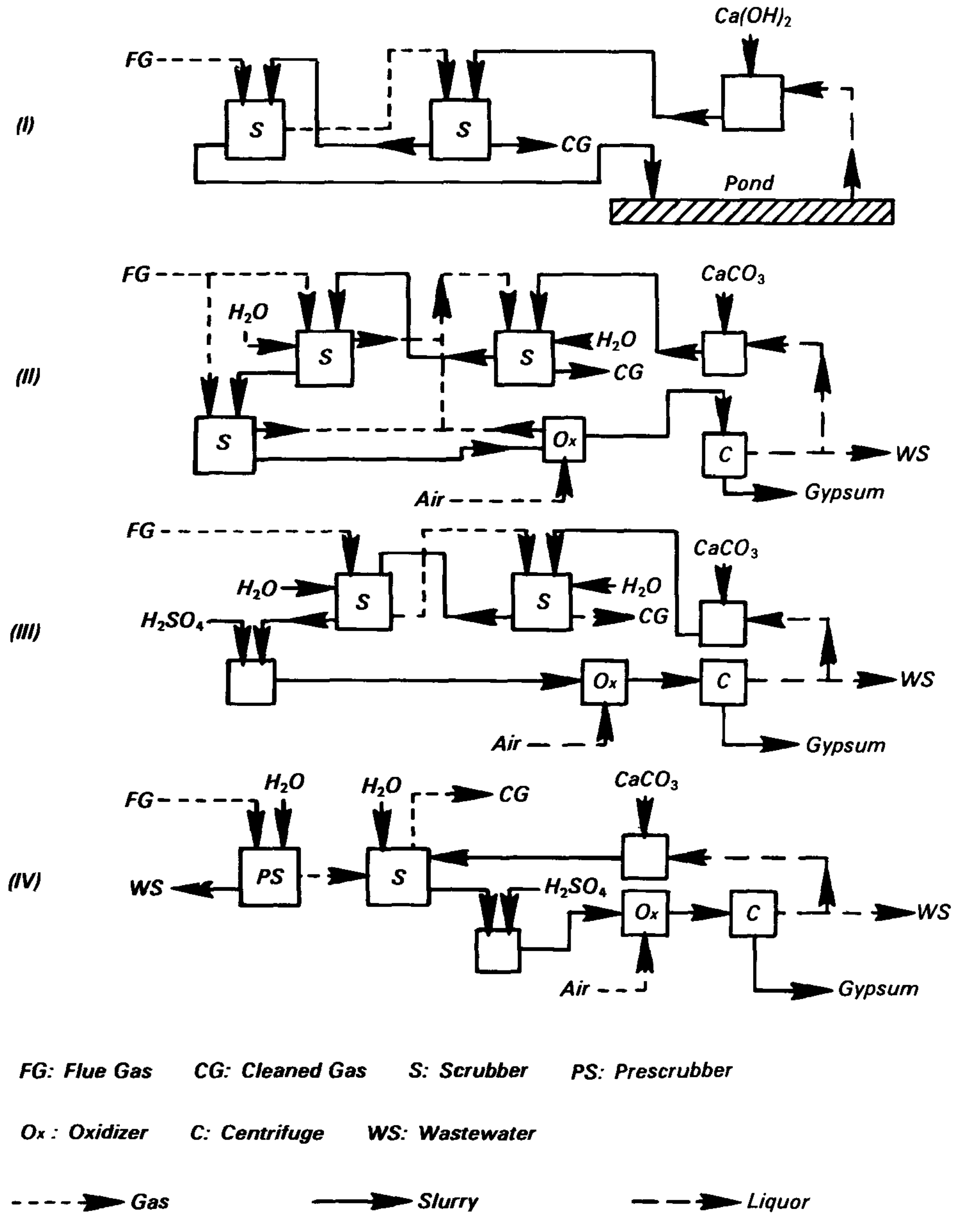

Figure 1. *Types of commercial wet lime/limestone processes for coal.*

this coal-fired power station is 18 ¥/kWhr, of which nearly 3 ¥/kWhr is for flue gas treatment.

Other FGD Processes for Coal

Although most FGD units for coal-fired boilers use a standard limestone-gypsum process, several modified lime/limestone processes have also been developed. Many of these processes (Table 7) offer advantages over the standard limestone-gypsum process. Kawasaki Heavy Industries' (KHI) magnesium-gypsum process, Dowa's aluminum sulfate-limestone process, Kureha Chemical Industries' sodium acetate-lime-gypsum process, and Kobe Steel's calcium chloride lime-gypsum process all reduce the possibility of scaling by using additives; Chiyoda's jet bubbling process lowers the pH to reduce scaling. Many of these processes are also characterized by low power consumption, low investment costs, and small amounts of wastewater. The red mud process is a low cost process, but is probably limited in its potential use.

Several dry FGD processes which use activated carbon have been tested on coal flue gas in an attempt to produce a sulfur byproduct. No information is available on these processes.

Simultaneous SO_x and NO_x Removal

The combination of selective catalytic reduction (SCR) of NO_x with wet process FGD has been used commercially for several coal-fired boilers in Japan since 1980. In a combined system, the SCR reactor is located between the boiler and the air preheater and operates at 350-400°C. The flue gas leaving the SCR reactor contains 1-5 ppm NH_3, which is removed by the wet FGD system. The ammonia causes no adverse effects on lime/limestone FGD processes, except that the wastewater from the FGD system contains a small amount of NH_3. At some of the plants, NH_3 in wastewater is removed by conventional activated sludge or ammonia stripping processes. Where SCR is applied upstream, the concentration of nitrate in wastewater derived from NO_x in flue gas may be reduced.

Two SCR systems are available for flue gas treatment from coal-fired boilers. One, the "high-dust" system, treats flue gas with full dust loads; the other, the "low-dust" system, has a hot-side ESP upstream of the SCR reactor to remove fly ash. The hot-side ESP is expensive but is suitable for low-sulfur coals for which cold-side ESP is not highly efficient.

Tests on dry simultaneous removal of SO_x and NO_x also have been conducted. The electron beam process for simultaneous removal has been tested by the Ebara Manufacturing Co. mainly for flue gases from oil-fired boilers and iron-ore sintering machines. In the U.S., AVCO, the licensee of the Ebara process, has tested the effects of coal fly ash on the electron beam process.

The activated carbon process for the simultaneous removal of SO_x and NO_x was tested by EPDC jointly with Sumitomo Heavy Industries. A demonstration plant will be completed by 1984. Activated carbon processes also have been tested by Mitsui Mining Co.

Wet simultaneous SO_x/NO_x removal processes have been tested and applied to several small commercial units. The wet processes, however, may not be suitable for large scale applications, particularly coal-fired boilers. Large scale systems need wastewater treatment to remove a considerable amount of impurities including nitrate and nitrite.

Other Coal-Utilization Technologies

Other coal-utilization technologies such as gasification, liquefaction, and fluidized-bed combustion (FBC), have been tested but may not be practical in Japan for

Table 4. *Operation Parameters of FGD Units for Coal-Fired Utility Boilers (Limestone-Gypsum Process)*

Station	EPDC Isogo	EPDC Takasago	EPDC Takehara	EPDC Matsushima		Chugoku Shimonoseki	Hokkaido Tomato-Atsuma
Boiler No.	1	1	1	1	2	1	1
Capacity (MW)	256	250	250	500x3/4	500x3/4	175	350x1/2
Gas treated (1,000 Nm^3/hr)	821	792	793	1,300	1,300	610	610
FGD constructor	IHI [a]	MM [b]	BHK [c]	IHI	BHK	MHI [d]	BHK
FGD start-up year	May '76	Feb. '75	Feb. '77	Jan. '81	July '81	July '79	Oct. '80
Inlet SO_2 (ppm)	450	1,500	1,730	1,000	1,000	770	230
Inlet dust (mg/Nm^3)	1,500	100	100	300	300	1000	25
1st scrubber (prescrubber)	Venturi	Venturi	Venturi	Spray	Spray	Spray	Venturi
L/G (liter/Nm^3)	7	6	2.5	3	3	3	2.2
2nd Scrubber (scrubber)	Venturi	Venturi	pp [e]	Spray	Spray	Packed	pp [e]
L/G (liter/Nm^3)	7	6	7	15	15	14	5
Outlet SO^2 (ppm)	20	100	100	60	60	30	10
Outlet dust (mg/Nm^3)	50	30	50	30	30	25	5
SO^2 removal efficiency (%)	95.5 [f]	93.3 [f]	94.2 [f]	94.0 [g]	94.0 [g]	96.1 [g]	95.7 [g]
Dust removal efficiency (%)	96.6 [f]	70.0	87.5	90.0	90.0	97.5	80.0
Pressure drop (mmH_2O)	360 [h]	325 [h]	615 [h]	145 [i]	133 [i]	240 [h]	450 [j]
Wastewater (t/hr)	10	7.5	12	15		9	2.2
Power requirement (%) [k]	2.2	2.6	2.6	2.2		2.2	1.7
Gas reheating facility	OF [l]	OF [l]	OF [l]	GGH [m]	GGH [m]	GGH [m]	GGH [m]

[a] Ishikawajima-Harima Heavy Industries.
[b] Mitsui Miike Machinery Co.
[c] Babcock-Hitachi K. K.
[d] Mitsubishi Heavy Industries.
[e] Perforated plate.
[f] 1-3% lower after reheating by oil-firing.
[g] 2-3% lower after gas reheating by gas-gas heater.
[h] By two scrubbers and mist eliminators.
[i] By two scrubbers.
[j] Two scrubbers, mist eliminator, and gas-gas heater.
[k] Percent of power generated.
[l] Oil firing.
[m] Gas-gas heater.

Table 5. *Estimated FGD Cost in 1981 (Assuming a new 500 MW coal-fired boiler, limestone-gypsum process, 70% boiler utilization, 3,066,000 MWhr/year, inlet SO_2 1,000 ppm, 90% SO_2 removal, 7 years depreciation, 10% interest.)*

Investment Cost (including gas-gas heater)	16 billion ¥ [a] (32,000 ¥/kW)
Annual Cost (millions of ¥)	
Fixed Cost (25% of investment)	4,000
Power (2.1% of power generated, 17 ¥/kWhr)	1,100
Labor (13 persons)	100
Limestone (43,000 t) and chemicals	250
Others (maintenance, etc.)	500
Total	5,950
Gypsum (84,000 t, ¥2500/t)	- 210
Total	5,740
Annualized Cost	1,87 ¥/kWh

[a] $1 = ¥250.

several reasons. Liquefaction and gasification are energy consuming and therefore too costly because Japan has to import coal. FBC processes with limestone to remove SO_2 create ash disposal problems. Moreover, SO_2 and NO_x reduction by FBC may not be sufficient to meet the stringent Japanese regulations.

Table 6. *Estimated Power Generation Cost for a New 1000 MW Coal-Fired Boiler (1981)*

Coal-fired Boiler	
Investment cost (¥/kW)[a]	
Power station excluding pollution control facilities	*210,000*
Flue gas desulfurization (90% efficiency)	*30,000*
Selective catalytic reduction (80% efficiency)	*7,000*
Electrostatic precipitator (99.5% efficiency)	*4,000*
Other pollution control facilities including ash disposal	*17,000*
Total	*268,000*
Annualized cost (¥/kWhr)	
Flue gas desulfurization	*1.7*
Selective catalytic reduction	*0.7*
Electrostatic precipitator	*0.2*
Other pollution control including ash disposal	*1.7*
Fuel (Coal at 15,000 ¥/ton)	*6.4*
Others	*7.3*
Total	*18.0*
Oil-fired Boiler	
Annualized power cost with low-sulfur oil at 64,000 ¥/kl with electrostatic precipitator (¥/kWhr)[b]	*21.0*

[a]$1 = ¥250.
[b]Including 7 years depreciation for pollution control facilities and 15 years depreciation for power generation facilities.

Table 7. *Other FGD Processes for Coal*

Absorbent	By-Product	Process Developer	Commercial Application	Status of Application for Coal
CaO, MgO	Gypsum	KHI	Oil-fired boiler	Commercial in 1981 in West Germany, in 1983 in Japan
$CaCO_3$	Gypsum	Chiyoda	Oil-fired boiler	23 MW test in U.S.
$Al_2(SO_4)_3$, $CaCO_3$	Gypsum	Dowa	Various sources	10 MW test in U.S.
CaO, CH_3COON_a	Gypsum	Kureha	Oil-fired boiler	1.5 MW test in Japan
CaO, $CaCl_2$	Gypsum	Kobe Steel	Sintering machine	1.5 MW test in Germany
Red Mud	Sludge	Sumitomo Aluminum	Oil-fired boiler	Possible commercial

Jumpei Ando is on the Faculty of Science and Engineering, Chuo University, Tokyo, Japan.

***J. David Mobley** is the EPA Project Officer (see below).*

The complete report, entitled "SO_2 Abatement for Coal-Fired Boilers in Japan," (Order No. PB 83-225 938; Cost: $20.50, subject to change) will be available only from:

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