ECOLOGICAL TECHNOLOGY AND MANAGEMENT
KALMAR, SWEDEN, September 22-24, 1999

23

ENVIRONMENTALLY FRIENDLY TREATMENT OF AIRPOLLUTANTS – Evaluation of Cumulative Process for Flue Gas Treatment-

Ryunosuke Kikuchi
University of Helsinki, Finland

Abstract

This paper proposes the combination of two different domains; the environment and agriculture. Emission of sulfur oxides and nitrogen oxides is associated with global acidification (e.g. acid rain), and it has become a serious environmental problem. Meanwhile, the amount of fertilizer consumed globally in 1996 was over 6 times that consumed in 1960. Electron beam flue gas treatment (a dry process) can simultaneously convert sulfur dioxide, sulfur trioxide and nitrogen oxides into nitrogen fertilizer. This process has been considered costly. An electron beam industrial plant has been built and is currently operating. The data delivered from this industrial plant proves that economic performance of the electron beam process is about the same as that of the conventional wet limestone process.

Introduction

Agriculture is a factor of particular significance, being basic to human survival and also important in sustaining global environmental stability. Reducing air pollution and increasing agricultural productivity are considered desirable. There are some large producers of fertilizer such as Kemira and Norsk Hydro in Scandinavia. That is, the basis of the fertilizer industry has already been formed. Therefore, the transformation of air pollutants such as sulfur oxides and nitrogen oxides into N-fertilizer might be advantageous for both polluters and fertilizer producers. The historical trend of fertilizer consumed globally is shown in figure 1 [IFDC 1998].

https://doi.org/10.15626/Eco-Tech.1999.023

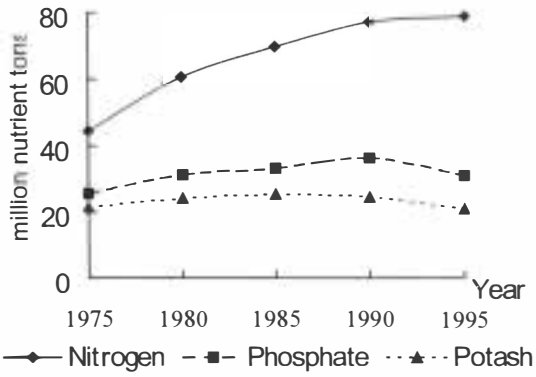

Fig.1. Global consumption of fertilizer by nutrient type

During the above-mentioned period, fertilizer consumption temporarily decreased in Eastern Europe due to economic confusion, and it has not yet completely recovered. On the other hand, the amount of consumed fertilizer has rapidly increased in Asia: the amount in 1990 was around 20 times that in 1960. Therefore, there has been a shortage of fertilizer in Asia. From the global viewpoint, consumption oftN-fertilizer in particular has greatly increased.

Wet limestone-gypsum flue gas desulfurization (FGD) has commonly been used for removing sulfur dioxide from flue gas not only in Scandinavia but throughout the world. Wet limestone-gypsum process and other processes. FGD has spread rapidly throughout the world since the 1970s. It is said that there are over 40 different types of FGD, but the wet limestone process accounts for over 90% of FGD units used globally [Klingspor et al, 1998]. It is reported that the global installation capacity of wet limestone FGD has increased by around 7,000 MW every year [Klingspor et al, 1998]. The above-mentioned limestone FGD process cannot remove NOx at all and cannot effectively remove very harmful SO_3. The limestone process releases gypsum slurry as a by-product, and furthermore releases wastewater (gypsum slurry) and carbon dioxide (a greenhouse gas). The treatment of sulfur dioxide with limestone is simply expressed as follows:

$CaCO_3$•aq (limestone slurry)t+ $SO_2 + 1/2O_2 \rightarrow CaSO_4$•aq (gypsum slurry)t+ $CO_2\uparrow$

Part of FGD wastes, for example gypsum, should be utilized, but the use of gypsum is limited (total use of 200 thousand tons/year) in Finland. The amount of FGD gypsum will significantly increase in Finland also, and then it will no doubt become a significant environmental problem [Ranta 1990]. The most likely user of this gypsum (by-product of FGD), the construction material industry, is not short of raw materials, and hence will only use a portion of the gypsum produced from wet limestone FGD [Ranta 1990]. This means that the disposal gypsum must be stored in buffers, and this results in a secondary solid waste problem. As gypsum powders coagulate, they are not suitable even to export. On the other hand, N-fertilizer is suitable for domestic use and export.

It would be better to convert SO_2, SO_3 and NOx simultaneously into N-fertilizer than convert only SO_2 into gypsum from the viewpoints of anti-pollution control and marketability of by-products.

Principle mechanism of electron beam process and industrial plants of this type

Electron beam processing simultaneously removes SO_2, SO_3 and NOx, and transforms them into N-fertilizer without generating wastewater. The schematic process flow of electron beam processing for flue gas treatment is illustrated in figure 2. The flue gas emitted from an incineration plant which burns fossil fuel is cooled in a spray cooler, and the fine mists of water supplied from the cooler's spray nozzles are totally evaporated by the heat of the flue gas. Prior to input to a process vessel, the flue gas is injected with gaseous ammonia.

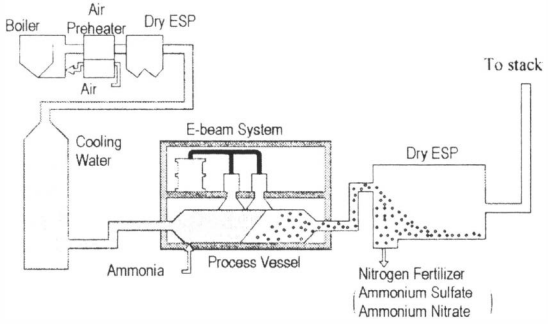

Fig.2. Schematic process flow of electron beam processing

In the vessel, the flue gas is irradiated by electron beams, causing free radicals to be generated. These radicals readily oxidize SOx and NOx to form intermediate substances which react with the ammonia to form ammonium sulfate and ammonium nitrate particulate. These reactions are summarized as follows [Ebara 1991]t

- Oxidation: $SOx + radicals \rightarrow H_2SO_4$,
  $NOx + tradicalst \rightarrow HNO_3$
- Formation oftN-fertilizer: $H_2SO_4 + 2NH_3 \rightarrow (NH_4)_2SO_4$,
  $HNO_3 + NH_3 \rightarrow NH_4NO_3$

The formed by-products are typical N-fertilizers. Before the flue gas is discharged from a stack to the atmosphere, it is fed to a by-product collector (e.g. electrostatic precipitator) where the particulate fertilizers are removed from the flue gas.

An electron beam plant for flue gas treatment with a 300,000-Nm^3/h capacity has been operating on the site of Chengdu coal-fired thermal power station in the city of Szechwan (China) since September 1997. The construction project was carried out jointly by the National Planning Committee of China, the Electric Power Department of China, Szechwan Power Industry Bureau, and Ebara Corporation. The gas conditions in the E-beam plant inlet are as follows: 150℃ temperature, 8% H_2O, 10% CO_2, 12% O_2, 1800 ppm SOx, 400 ppm NOx, and 800 mg/m^3 dust. The electron beam plant hourly consumes 1800 kWh power, 625 kg ammonia, 2 tons steam (as a heat source), and 22 tons industrial water. Using the above-mentioned utilities under the inlet conditions, the following output is obtained: 80% DeSOx efficiency – the inlet concentration of SOx varies widely (from 800 ppm to 1,700 ppm), but the measured DeSOx efficiency is almost constant and is as high as 82-88%, which meets the project target of 80%; 10% DeNOx efficiency – high efficiency is not required for this project;

200 mg/m^3 dust; and 2,450 kg/h by-product consisting of 92% ammonium sulfate, 1% ammonium nitrate and 7% fly ash, and a chemical analysis of the by-product shows that the nitrogen content is 19.7%. Parallel to this long period of operation, a vegetable (*Brassica campestris)* growth pot test using the obtained by-product was conducted to verify its fertilizing effect on the vegetable, and the results demonstrate that the vegetable grows better in fields fertilized with the by-product than in other fields treated with commercial ammonium sulfate and urea and in non-fertilized fields. Taking by-product sales (US$60/ton) into account, the annual operation cost is estimated to be US$74,200 at 6,570 hours of operation per year. The initial cost was around US$8 million. In contrast, US$12 to 13 million is the average initial cost and US$474,500 is the average annual cost of operation (6,570 hr/year) of a wet limestone-gypsum FGD unit with a 300,000-Nm^3/h capacity in China [Vowden et al, 1997].

Previous evaluation of electron beam processing

At the SO_2 control symposium [Cichanowicz et al, 1991], it was pointed out that electron beam processing would be an expensive method for flue gas treatment. When this was reported, no industrial plants for electron beam processing yet existed in the world. Real economic data could not be obtained till an electron beam industrial plant with a 300.000-Nm3/h capacity was put into commercial operation in China in September 1997. Furthermore, the economic data used at this symposium was based on the index dated January 1990, and no industrial SNOx plants had yet been operated at that time. The world's first SNOx industrial plant (900,000-Nm^3/h capacity) was put into commercial operation [Nordjyllandsværket 1992] in Denmark on 1 November 1991.
The initial costs of 3 different electron beam plants in China, Poland [Chmielewski et al, 1996] and Ukraine have been divulged and are summarized in table 1.

Table 1. Initial cost of electron beam plant

Location (country)	Capacity (Nm^3/h)	Initial Cost (US$ mil.)	Levelization (US$/kW)	Situation
Chegdu (China)	300,000	8.0	85.3	under operation
Pomorzany (Poland)	270,000	18.6	220.4	under construction
Slavyanskaya (Ukraine)	100,000	4.3	137.6	under project

Adding updated data, the figure showing capital cost which was presented at the above-mentioned symposium can be rewritten as shown in figure 3. The costs for "real electron beam" and "real SNOx" in figure 3 represent the data delivered from actual industrial plants and the costs for "electron beam" and "SNOx" represent the data reported at the SO_2 control symposium.

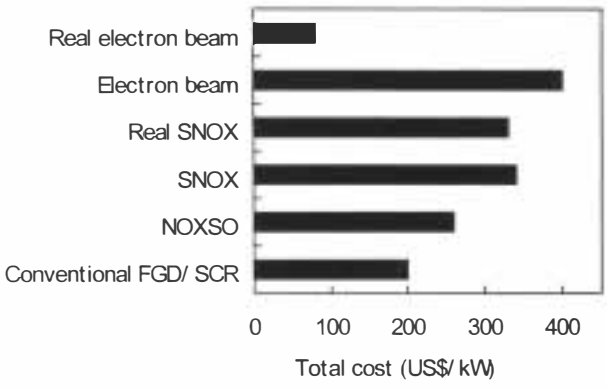

Fig.3. Cumulative process parameter on total capital cost

It follows from figure 3 that the electron beam process was the most expensive at the symposium in 1991 but this process is the cheapest on the basis of actual data. Commonly, 3.2 Nm^3/h of coal-fired flue gas is emitted in the process of obtaining 1 kW of energy by burning coal with 6,200 kcal/kg of heat value at 40% thermal efficiency, and thus the value of 3.2 Nm^3/h is applied to convert the unit from US$/$Nm^3/h$ to US$/kW in figure 3 and table 1. Each levelized initial cost of the electron beam plants listed in table 1 is less than or about equal to that of combined FGD/SCR shown in figure 3: conventional flue gas desulfurization (FGD) for SO_2 removal and selective catalytic reduction (SCR) for NOx removal.

Considerations and conclusion

An economic comparison of electron beam processing with cumulative processes is described above. FGD is used to remove only SO_2, and the electron beam process is used to remove SO_2, SO_3 and NOx. Nevertheless, economic comparison between electron beam processing and normal FGD (wet limestone type) has been a frequent topic of discussion. An industrial-scale project for building an electron beam plant is currently under way at the Maritsa East power station in Bulgaria. An economic comparison with FGD is considered based on this case [Aoki and Kikuchi 1997]. Figure 4 shows the initial cost of the electron beam plant for unit 8 (215 MW) of the Maritsa East power station plotted on a graph of recent FGD cost data presented at the 1995 SO_2 control symposium [Keeth 1995].

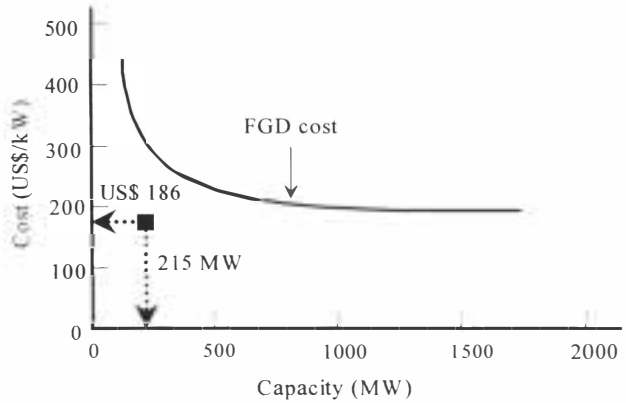

Fig. 4. Recent FGD cost vs. cost of electron beam plant

It follows from figure 4 that the cost of an electron beam flue gas treatment plant is about one-half that of FGD. However, the FGD cost curve presented in figure 4 is based on economic data delivered from projects in the USA, and the cost of an electron beam plant is based on the Bulgarian project. Therefore, the costs should not be directly compared. Taking the economic difference between the USA and Bulgaria into account, it is considered that the cost of an electron beam plant is about the same as that of FGD. Energy consumption is an important factor when an anti-pollution control facility treats flue gas at a power station. Energy consumption and removal efficiency of air pollutants are compared for FGD and the electron beam process in the Marista East project in the case of treatment of 1,500,000 Nm^3/h flue gas containing 5,500 ppm SO_2, 140 ppm SO_3 and 390 ppm NOx, and the results are summarized in table 2. Energy consumption does not depend on removal efficiency in wet limestone FGD, but it depends on removal efficiency in an electron beam unit.

Table 2. Removal efficiency and power consumption

Parameter	Electron beam process	*Wet limestone FGD
Removal efficiency of SO_2 (%)	86	90
Removal efficiency of SO_3 (%)	80	40 - 50
Removal efficiency oftNOx (%)	72	0
Power consumption (kWh/h)	10,310	10,383

[Ministry of the Environment, 1996]

Table 2 shows removal performance where energy consumption of an electron beam unit is lower than that of a wet limestone FGD unit, and it is seen that 3 different air pollutants can be treated effectively by the electron beam process with comparatively lower power consumption. When this power consumption is increased, the removal efficiency of each

pollutant also increases in the electron beam process. The electron beam process is considered advantageous in terms of cost performance and effective air pollution control (removal of SO_2, SO_3 and NOx), but it has not been widely adopted. The reason for this can be considered as follows: the wet limestone process is now a mainstay FGD method, and there is no collaboration between the energy sector (polluter) and the agricultural sector (by-product consumer). As there is mainly only one type of FGD in use, well-balanced use of FGD may be desirable. The fertilizer industries have a central role to play in developing productive agricultural practices, and the utilization of air pollutants in agriculture will lead to global-scale recycling. The linking of these two sectors by electron beam processing will be useful in realizing global recycling.

References

Aoki. S. and Kikuch, R. (1997). Combining Zero emission with N-fertilizer production. Fertilizer International, 360 (9/10), pp. 86-91

Chmielewski, A.tG., et al. (1995). Industrial Demonstration plant for electron beam flue gas treatment, Radiat. Phys. Chem. vol.46, No.4-5, pp. 1063-1066. DOI: https://doi.org/10.1016/0969-806X(95)00321-N

Cichanowicz, J.tE. et al. (1991). Engineering Evaluation of Combined NOx/SOx controls for utility Application, SO2 control Symposium, Dec. 3-6, Washington.

Ebara Corp. (1991). Tech. catalogue. No. CC3402EB, EBA Eng. dept., Tokyo. International Fertilizer Development Center-IFDC-(1998). *Global and Regional Data on fertilizer Production and Consumption*, IFDC, Alabama, USA.

Keeth. R.J., et al. (1995). Utility Response to Phase I and II Acid Rain Legislation - An economic Analysis, EPRI/DOE/EPA SO_2 Symposium, March 20-31, Maiami.

Klingspor,J., Tokerud,A. and Ahman,S. (1998). Low-cost FGD system for emerging market, ABB review, No. **1/98**, pp. 30-40.

Ministry of the Environment of Bulgaria. (1996). Pre-feasibility study report on introduction of flue gas treatment to Maritsa East No.2, Sofia.

Nordjyllandsværket (1992). NV SNOX, I/S Nordjyllandsværket, Vodskov.

Ranta, J. (1990). Amount, Quality and Utilization of FGD Wastes in Finland. In: *Acidification in Finland*, P. Kauppi, et al. (eds), Sringer, Berlin, pp. 1209-1215.

Vowden S, Hamada K, et al. (1997). Electron Beam Flue Treatment, Tech. seminar, European Bank for Reconstruction & Development, April 18, London