TECHNICAL AND ECONOMICAL FEASIBILITY STUDY OF THE ELECTRON BEAM PROCESS FOR SO_2 AND NO_X REMOVAL FROM COMBUSTION FLUE GASES IN BRAZIL

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ABSTRACT

The release of toxic gases into the atmosphere, mainly because of acid rain has been object of many discussions in all the world resulting in international programs of research for the development of efficient flue gas removal techniques, mainly SO_2 and NO_x, and in setting more and more limits of emission. Among the flue gas treatment methods, the process of electron beam irradiation has shown to be promising. Under irradiation, those gases are simultaneously removed from the combustion gases. In the presence of ammonia, the byproduct of the process is ammonium sulfate and ammonium nitrate and after filtration it can be used as a fertilizer. The process has been investigated in Japan, Germany, USA and Poland. Data concerning the present state of the process along with the design and implementation of a laboratory pilot plant for the electron beam flue gases removal process located at IPEN-CNEN/SP are presented.

1. INTRODUCTION

Sulfur oxides are created and exhausted into the air when fossil fuels that contain sulfur (coal, oil and natural gas) are burned. Nitrogen oxides are formed when the nitrogen and oxygen are burned with fossil fuels at high temperature. Latter acids are being formed in the atmosphere and fall to earth as acid rain or snow. In result lakes and forests are being damaged in certain part of Central Europe, China, Northeastern United States and Eastern Canada. Some acid can be transported far away from industrialized zones and cross international borders to ruin the environment in non-urban areas. Trees, crops, and plants may be hurt. The acid rain affects buildings and monuments what can be seen in many european cities. These are the reasons why stricter control of SO_2 and NO_X emissions has become internationally recognized as a global problem and many countries have set limits for the discharge of pollutants. SO_2 and NO_X are listed among them (5).

In the past years, the use of fossil fuels with high sulfur content in Brazilian industrial installations has grown. In addition, estimates indicate such growing will be continuous. Due to environmental regulations enacted, the development of a technique able to remove toxic gases has become essential.

The air pollution in Europe is particularly severe. There exists consequently a strong need for air pollution technology in order to improve such situation. Poland, which produces energy mainly from pit and brown coal, is a big producer of these pollutants. Numbers

regarding the NO_X emission should be multiplied by a factor 2.9 since the nitrogen dioxide form much stronger acid becoming harmful to the environment (2).

1.2 - CONVENTIONAL METHODS FOR SO_2 AND NO_X REMOVAL

Several FGD (Flue Gas Desulphurization) methods have been developed up to now. The methods can be divided into several cathegories: dry, wet and with sulfur recovery system.

Dry Scrabbers
LSD - Lime Spray Dryer
CFB - Lurgi Circulating Fluid Bed
FSI - Furnance Sorbent Injection
EI - Economizer Injection
DSI - Duct Sorbent Injection
DSD - Duct Spray Drying
ADV - Moist Dust Injection
LSFO - Limstone with Forced Oxidation

Wet Scrubbers
LFSO - Limstone with Forced Oxidation
LSWB - Limstone with Wallboard Gypsum
LSINH - Limestone with Inhibited Oxidation
LSDBA - Limestone with Dibase Acids
PURE - Pure Air/Mitsubishi
MGL - Magnesium Enhaced Lime
LDA - Lime Dual Alkali
LSDA - Limestone Dual Alkali

Sulfur Recovery System
WLWN - Wellman Lord
ISPRA - ISPRA - Bromines
MgOx - Magnesium Oxide
LSFO - Limestone with Forced Oxidation

Dry and wet methods can be applied for reduction of NO_x pollutants. SCR selective catalytic reduction, precipitation on solids, catalytic decomposition on solid electrolyte and reduction to N_2 by NH_3 are examples of dry scrubbers. Absorption in liquid with reduction to NH_4, adsorption in liquid with oxidation NO_2, NO_3 are used in the wet method.

The stricter control of NO_X and SO_2 pollutants, which are being forced in many countries, provokes an impact in the development of low cost NO_X/SO_X control technology as alternatives to existing ones: SCR (Selective Catalytic Reduction) for NO_X and FED (Flue gas desulphuration) for SO_2 control. The evaluation of nearly 70 processes has been done under the EPRI project to select the most promising technology (9).

The recommended methods were selected under screening technology condition based on:

1. Development status (empirical experience, on-going development, commercial use);
2. Technical feasibility (probability that commercially viable process can be developed);
3. Retrofitability (land requirements for process and waste disposal, use of existing equipment and required point of access to the flue gas stream);
4. Environmental risk (high volume waste, low volume waste, secondary gaseous emissions, potencial risk due to process upset);

5. Process reliability (chemical and mechanical complexity, sensitivity to process runnings, corrosive environment);

6. Energy and resource requirements (quantity, reagent consumption rate, catalyst/sorbent consumption)

In addition to EB technology three other processes have been selected:

- NO_XSO (solid phase adsorbent with fluidized bed reactor)
- SNRB (SO_X - NO_X - ROX - BOX)
- WSA-SNO_X (wet scrubbing iron-chelate process).

a) THE NO_XSO PROCESS

The NO_XSO process is based on the use of solid phase adsorbents to remove SO_X and NO_X in a fluidized bed reactor. The adsorbent is removed from the reactor by means of several steps processing. In first stage NO_X is removed under controled temperature treatment. The concentrated NO_X stream from this stage is directed to the boiler inhibiting the formation of additional NO_X under thermodynamic equilibrium. In a second stage, a reducting gas is applied (methane, carbon monoxide) to produce gas consisting of SO_X, H_2S and elemental sulphur what can be later processed to produce marketable byproduct of sulphur. The adsorbent is returned to the reactor after a stream-treatment and cooling operation.

The advantages of the NO_XSO method is the low temperature (120^0C) process which corresponds to the ESP outlet. It also means retrofit applications because of the dowstream of the ESP location. The use of fluidized-bed reactor for improving efficiency cost makes a high pressure drop of the flue gas. The demonstration program includes the NO_XSO Corporation, the DOE, the Ohio Edison Company, the EPRI and other organizations.

b) THE SNRB PROCESS

In this process a lime or sodium reagent is injected into the flue gas duct wherein ammonia is also injected. The alkaline reagent reacts with SO_X in a duct and on hot filter bags. A SCR catalyst is located on or within the bags to reduce NO_X with the ammonia presence and form elemental nitrogen. The development of the filter bags with SCR catalyst suitable for high temperature operation will demonstrate the capability of this heat recovery process. Babcocle & Wilcox, DOE, EPRI and others are engaged in this dry injection technique development program.

c) THE WSA-SNO_X PROCESS

The wet scrubbing Iron-Chelate Process can be easly adopted to retrofit the plants wherein a FED System has been already implanted. The iron-chelate additives react with NO_X in wet scrubbing process to form compounds that include sulphur-nitrogen species.

In comparison with FED process a longer gas/liquid contact or a higher flue gas measure drops may be required for appropiate NO_X removal. The Iron-Chelate oxidation and the stream of waste which should be additionally treated before disposal may create a technical problem and a significantly increase of the cost of the process.

2. PRINCIPLE OF THE EB PROCESS

The research on flue gas treatment by radiation was initiated by the Ebara Corp. in 1970. Fundamental work and pilot scale experiments have been performed in Japan, USA, Germany, Poland and other countries since then. It was founded in a basic and experimental way that EB technology for flue gas treatment has the following advantages (5):

- Simultaneous removal of SO_2 and NO_x
- Dry process without wastewater
- Byproduct can be used as fertilizer
- No need of a catalyst
- Low capital and operating costs compared with conventional methods.

The process is based on three stages. In the first one the flue gas is irradiated leading to radical formation such as OH, O, HO_2. In the second stage SO_2 and NO_x are being oxidized to H_2SO_4 and HNO_3 in presence of water through a concurrent number of chemical reactions. In the third stage the intermediate product reacts with the ammonia presence to form ammonium sulfate and ammonium nitrate. Ammonia in near stechiometric quantity is injected into the vessel prior to the flue gas entrance into the process vessel. These dry powdery ammonium salts are collected by the filtering units (ESP or bag filters) and can be used as agricultural fertilizers (4).

The process can be used for treatment of the gases from coal and oil fixed power stations, industrial boilers, furnaces and municipal solid waste incinerators.

Also retrofitting of existing facilities to reduce SO_2 and NO_x concentrations is possible regarding to the low space requirement and location between the ESP and the stack where a move space is available.

In practical instalations 95% of SO_2 and 85% of NO_x removal efficiency can be obtained. The main components of the facility are the spray cooler, the process vessel accelerator and byproduct collector which can be fully automated what makes the process easier to be operated.

2.1 - PROCESS MECHANISM

When high energy electrons are applied for flue gas irradiation, radicals and free atoms are generated. The interaction of these electrons and flue gas molecules results in ionizing and dissociation. The fraction of energy absorbed by each gas component is proportional to its partial pressure. Principal reactions in primary processes can be schematicaly represented by (4):

N_2 ---> N_2^+ (2.27); e- (2.68); N^+ (0.69); N (3.05); N_2^* (0.29)

O_2 ---> O_2^+ (2.07); e- (3.30); O^+ (1.23); O (1.41); O_2^* (1.90)

H_2O --> H_2O^+ (2.56), e- (3.23); H (4.07); OH (4.17); O (0.45)

CO_2 --> CO_2^+ (2.24); e- (2.96); CO^+ (0.51); O^+ (0.21); O (0.38)

Where the number in parentheses represent the G values of the species and the G is the number of molecules produced per 100eV of energy absorbed in the system. This is the first stage of the process.

During the second stage radicals and atoms containing the oxigen react with SO_2 and NO_X to form, in the presence of water, sulphuric and nitric acids. There is also the ion-molecule reaction mechanism for the decay of the primary species. Low concentration components have to compete with the primary radical decay processes. Above 760 reactions were listed in Agate Code to describe the undergone processes. Some reactions from the secondary stage, wherein SO_2 and NO_x are involved, are listed below (4):

$$SO_2 + HO_2 \text{ ---------> } SO_3 + OH$$

$$SO_2 + OH \text{ ---------> } HSO_3$$

$$SO_2 + O \text{ ---------> } SO_3$$

$$SO_3 + H_2O \text{ ---------> } H_2SO_4$$

$$NO + OH \text{ ---------> } HNO_2$$

$$NO_2 + O_3 \text{ ---------> } NHO_3 + O_2$$

$$NO_2 + HO_2 \text{ ---------> } HNO_2 + O_2$$

$$NO_2 + OH \text{ ---------> } HNO_3$$

Most than 20% of the NO is converted into free N_2 being released in the EB process in the presence of ammonia according to JAERI and KFK's tests.The last stage is the product formation. Finally, the gas conversion process is initiated by the reaction of sulphuric and nitric acids in the presence of water and stoichiometric amount of ammonia. These acids are converted into ammonium sulphate and ammonium nitrate and are collected by a filtering system (4).

The efficiency of the EB process was determined in many experimental facilities to optimize process conditions. Last data show that 95% of SO_2 removal efficiency can be obtained at a 5kGy dose being the water content and the thermal reaction condition properly optimized. The multistage irradiation can significantly improve the NO_x removal. The 7kGy dose for the two stages and the 6kGy dose for a three stage irradiation is required for a 80 % efficient removal (5).

2.2. ELECTRON BEAM FACILITY FOR FLUE GAS TREATMENT

The first experimental facility for EB process applied to flue gas treatment was built by the Ebara Corp. in Japan. The batch tests where carried out in the 1970-71 period. The experiments proved that SO_2 and NO_x can be removed from irradiated flue gas in results of radiation chemical reactions. Subsequent development of the process has been continued by Ebara, JAERI, University of Tokio, NKK in Japan, Ebara, Research Cortrell, Department of Energy, Electric Power Research Institute, University of Karlsruhe, KFK, Badenwerk in Germany, Institute of Nuclear Chemistry and Technology, Warsaw Power Station in Poland (5).

The EB process is being used now to remove other kinds of gas pollutants. The results obtained from experimental works already underwent proved the capability of the process gas, traffic tunnel ventilation gas and various VOC pollutants in the gas phase (3, 7).

In order to demonstrate the capability of the EB process, four pilot plant demonstration facilities are being now used both in Poland and Japan. They are based on the Ebara process where ammonia is injected before the process vessel wherein the flue gas is irradiated (5).

The Table1 shows the parameters of the pilot plants for the flue gas treatment which have been installed since 1991 and are being used now to demonstrate the capability of the EB technology for commercial use (5).

In 1991, a 3-year 14.3 million USD project was initiated in Japan by the Ebara Corp. together with the Japan Atomic Energy Research Institute (JAERI, Takasaki) and the Chubu Electric Power Company (Nagoya). The main objectives of the research carried out at this pilot plant are as follows:

- To recognize the quantitative characteristics of the process;
- To test multistage irradiation;
- To optimize collecting (ESP/bag house) and byproduct handling systems;
- Study and evaluation of the commercial characteristics of the process;
- To evaluate the reliability of the process during a long period operation;
- To improve necessary areas of the facility.

TABLE 1. The major parameters of the pilot/demonstration plants for the flue gas treatment which have been installed since 1991.

INSTITUTION	YEAR OF INSTAL.	VOLUME FLOW RATE (NM^3/h)	SO_2/NO_x (ppm)	TEMP (°C)	ACCELER.
INCT/KAWENCZYN POLAND	1991	20.000	200/600 to 250	60 to 80	500 - 700 KeV 2 X 50 kW
EBARA / JAERI JAPAN	1992	12.000	800 to 1000 or 150/300	65	800 KeV 3 x 36 KW
EBARA / TOKYO JAPAN	1992	50.000	---- 0 - 5	20	500 KeV 2 x 12.5KW
NKK / JAERI MATSUDO-JAPAN	1992	1000	100 100 HCl = 1000	150	400 - 350 KeV 15 KW

To confirm capability of the EB method in low NO_x content gas, a Tokyo plant was built by the Ebara Corp. and the Tokyo Metropolitan Government to treat ventilation exhausted gases from a highway at the Tokyo Bay Tunnel. The facility was finished in June 1992. The main parameters of the pilot plant are shown in Table.1. 50.000Nm^3/h of gas from the ventilation exhauster is introduced into the irradiation vessel for EB treatment with the ammonia presence. As a result NO_x is converted into powdery ammonia nitrate products. The activated carbon is used to remove the ozone formed by the irradiation. A 80% targed removal efficiency is being obtained at 3ppm level of NO_x in inlet parts.

To evaluate the EB process applied to the flue gas from municipal waste incinerators a pilot plant was built by NKK, JAERI and Matsudo City Government Clean Center. The plant was completed in June 1992. The main parameters of the plant are shown in Table 1. Targets of the removal efficiences are as follows:

NO_x: 100ppm --------> < 50ppm
SO_2: 100ppm --------> < 10ppm

Hcl : 1000ppm -------> < 10ppm

The irradiation is being done where the slurry of calcium hydroxide is sprayed at a temperature higher than 150°C. The bag filter is used to collect powdery products (mixture of calcium nitrate, sulfate and chloride) formed by the irradiation. During the process HCl and SO_2 are removed by spraying the slurry of $Ca(OH)_2$. NO_x is effectively removed by EB irradiation (6).

The Polish Pilot Plant, with a 20000Nm^3/h capacity, has been built at EPS Kawenczyn in Warsaw. The installation was constructed on the by pass of the main stream of the flue gas with total flow net 260000Nm^3/h from the WP-120 boiler (nominal heat output 120Gcal/h, efficiency 84%, coal consumption 26-32 t/h). The black coal used contains 1.2% sulphur, 18% ash content and a calorific value of 4700 Kcal/kg.

The Polish Pilot Plant is the first installation in which two stage irradiation by electron beam was applied resulting in a significant decrease of energy consumption. The other novelties of this construction are connected with the process vessel where irradiation zones are located along the flue gas system flow and a double window construction was applied with perpendicular streams of air for cooling the output windows at the accelerators and the inlet windows of the process vessel.

The main objectives of the research carried out at the pilot plant are (2):

- Testing of all parts of the installation under industrial conditions;
- Optimizing of the process parameters leading to the reduction of energy consumption with high efficiency of SO_2 and NO_X removal;
- Selecting and testing filter devices and filtration process;
- Developing of the monitoring and control systems at industrial plant for flue gas cleaning;
- Preparation of the design for an industrial scale facility.

2.3. PRESENT STATUS OF ELECTRON BEAM PROCESS

The EB process applied to the flue gases treatment is suitable for full scale commercial application. It was determined by basic experiments and operation of pilot plant facilities. This is a dry process with a usable byproduct which can offset the operating and investment costs. The EB technology was recognized as flexible and adaptable with excellent turndown ratios. The process can be easy controlled for different removal efficiencies and adjusted for the utilization of different fuels. Major conclusion regarding the EB process for flue gas treatment are as follows:

- More than 95% of SO_2 and 85% of NO_x can be simultaneously removed from the flue gas under optimal operating conditions;
- Ammonia should be injected into the process in near stoichiometric amount, upstream injection was found to be more efficient;
- SO_2 removal efficiency depends on the temperature injection, the filter condition and the EB dose;
- The quantity of SO_2 removed by EB is relatively independent from the inlet SO_2 concentration;
- NO_x removal occurs almost entirely under EB application and depends strongly on the dose, gas temperature and ammonia stoichiometry are the second order effects;
- NO_x removal efficiency is increased as the inlet SO_2 concentration increases. This occurs as a result of the formation of nitrosulphuric compounds;
- 5kGy is required for 95% of SO_2 removal efficiency and 7kGy is required for 80% of NO_x removal efficiency in a two stage irradiation facility in optimal conditions;

- Good reliability of the long time operation was demonstrated in pilot plant facilities;
- The byproduct collected during the process consists of ammonium sulfate and ammonium nitrate which can be effectively used as a fertilizer. The small amount contaminants does not affect the quality of the product;
- No waste water in the process is being produced;
- Relatively low capital investment and operating cost of the EB process facility can rate this method as equivalent or preferable to compare with FED/SCR ones;
- Low space requirements produce a significant advantage in the retrofit installations;

To complete present data of the EB process intense experiments are being done in Japan, Poland and Germany. The number of the most interesting subjects are listed below:

- Experimental study of quantitative characteristics of the process at the pilot plant level;
- Design study and evaluation of commercial characteristics of the process;
- Experimental study to apply this method for other kind of gases treated by radiation;
- Wet and dry ESP, baghouse, gravel bag filter experimental study to optimize byproduct collecting system;
- Optimization of the spray cooler construction to obtain dry bottom and reduction of power consumption;
- Optimization of the systems preventing or removing duct clogging byproduct;
- Duct configuration (rectangular, cylindrical) and gas velocity in duct and process vessel are investigated;
- Multistage irradiation (two and three zones);
- Ammonia slip and ammonia injecting (location, quantity);
- Byproduct handling studies (granulation, liquid, storage, fertilizer tests).

The Electron Beam process for flue gas treatment could be used beneficially in the future. Experimental studies describe above improve the technology and promote it for future applications (2).

3. EQUIPMENT SPECIFICATION

BOILER - Oil or coal fired to produce thermal or electrical energy.
ESP - Electrostatic precipitator to reduce the fly ash content downstream to the boiler.
HEAT EXCHANGER - To reduce inlet or increase outlet gas temperature by additional stream of air or water.
SPRAY COOLER - Vertically installed down to the boiler and ESP is used to increase water content in flue gas and describe its temperature by complete evaporation injected water.
AMMONIA INJECTION - To keep stoichometric quantity of NH_3 in flue gas stream.
PROCESS VESSEL - Horizontally mounted with multistage irradiation capability.
ACCELERATOR - To initiate radiation chemical process of flue gas treatment.
ANALYTICAL AND CONTROL SYSTEM - to keep automatic control over the process.
COLLECTOR - as baghouse/ ESP/gravel bed filter to collect byproduct.
BYPRODUCT HANDLING SYSTEM - To prepare powder, granules or wet sort of byproduct.
INDUCED DRAFT FAN - To overcome pressure drop in ducts and byproduct collector.

3.1. GENERAL ARRANGEMENT OF THE TECHNOLOGICAL PROCESS

Flue gas generated by the coal heated boilers enters the EB process after ESP where the ash content is reduced in order to improve the quality of the fertilizer byproduct. No such filter is foreseen after the oil-fired boiler. The initial concentration of SO_2 depends on the sulphur content of the applied fuel. NO_x concentration depends on the combustion process temperature and is different for different burners and boiler construction.

Heat exchanger is usually used to reduce the gas temperature in the initial cooling stage up to 150-250°C level. Then flue gas enters the spray cooler where the temperature is reduced to 65-80°C by atomized water injection. Usually a dry bottom principle is applied to operate the spray cooler facility, to eliminate a residual wastewater stream. Water is totally evaporated by a heat exchange with the hot flue gas once the dew point of the gas is approximately 50°C. Water content in the flue gas should be increased up to 8-12% in this stage.

Ammonia in stoichiometric quantity is injected before the flue gas enters the process vessel where it is irradiated by the electron beam to promote the reaction of the ammonia and flue gas. The beam interacts with nitrogen, oxygen, water and others substances in the flue gas to produce active free radicals such as OH, O, HO_2. In results SO_2 and NO_x are converted to sulphuric and nitric acids and finally forms a byproduct consisting of ammonium sulfate and ammonium nitrate (6).

The ammonium sulfate and the ammonium nitrate are collected by electrostatic precipitator or bag filters and the cleaned flue gas is released through the fan into the stack.

3.2. MAJOR EQUIPMENT

3.2.1. ACCELERATORS

The present estimate of the required dose level for an efficient NO_x removal (80%) shows that the radiation dose should be in the range of 10kGy for low sulphur content coals. Multistage irradiation can reduce this figure up to 7kGy. It is necessary to remember that 95% of SO_2 removal can be obtained with a 5kGy dose. Significant improvement in NO_x removal can be achieved when high sulphur coal is applied. If it is assumed that gas absorbs 85% of the total beam energy then 1MW accelerator facility will be sufficient for a 100MW generator with the dose range described above.

The required beam power level is significantly higher than in those accelerators utilized for industrial beam processing but there are technical prospects to build accelerators with a 200-500kW unit power what sharply reduces the number of accelerators in industrial facilites and their cost.

According to accelerator producers the cost of high power 800keV machines is in the range of 5 US$/W at present. The new developments which are under progress in USA (induction linac) give some prospect to reduce the cost level by factor 2.

Many factors should be considered when specifying the location of the accelerator/scanner relative to the process vessel. The most important are: dose uniformity, cost and easy access to maintenance. The best position of the scanner was found to be at the top of the process vessel with the irradiation zones along the gas stream flow. The multistage irradiation is recommended to increase the process efficiency (10).

The process vessel location in horizontal position and at the underground level can reduce shielding costs and allows to have an easy access and change of certain components of scanner/process vessel systems.

The Table 2 shows the basic electron beam parameters which have been applied in laboratory and pilot plant facilities for flue gas treatment.

The Table 3 shows producers and accelerators which are suitable for flue gas treatment in capacity 10.000 - 20.000 Nm^3/h (10).

3.2.2. FILTERS, BYPRODUCT HANDLING

The process of particles formation and filtration has been intensively investigated during the recent years. The mass median aerodynamic diameter of the product aerosol facilities around $1_u m$ depend on the dose and flue gas parameters (4).

A baghouse was initially selected as a byproduct collector. A pre-coating system was used to protect the bag' surface from direct contact with hydroscopic byproduct. To avoid decreasing property of byproduct by neutral precaution material diatomaceous earth can be used.

TABLE 2. The basic parameters of the electron accelerators applied in facilities for flue gas treatment.

TYPE OF FACILITY	ENERGY (MeV)	BEAM POWER (KW)	TYPE OF ACCELERATOR	REMARKS
	12	1.2	linear	Ebara, Japan
LABORATORY	3	15	Cockrft-Walton	JAERI, Japan
	1.2	1.2	Dynamiton	Tokyo Univ.
FACILITY	1.5	30	----	JAERI, Japan
	0.22	22	Transformer	Karlsh.,Germ
< 1000 Nm^3/h	0.3	3.6	"	KFK, Germany
	0.7	5	Resonance	INCT, Poland
	0.75	30	----	Ebara, Japan
PILOT	0.75	2 x 45	----	Ebara, Japan
	0.8	2 x 40	----	Res. Cott.US
DEMONSTRATION	0.8	2 x 80	----	Ebara, USA
	0.3	2 x 90	Electrocurtain	Badenwerk,GE
FACILITY	0.5	15	Cockrft-Walton	KFK, Germany
	0.5	15	"	Ebara, Japan
	0.7	2 x 50	Transformer	INCT, Poland
1000 - 20.000	0.8	3 x 36	Cockrft-Walton	Ebara,Japan
	0.5	2 x 12.5	"	Ebara, Japan
(Nm^3/h)				
INDUSTRIAL	0.8	8 x 150	Transformer	
PLANT	1.0	4 x 400	Induction linear	
300.000 Nm^3/h				

To remove byproduct deposition from the bag filter and reduce baghouse pressure drops several methods can be applied:

- Pulse jet cleaning
- Reverse flow cleaning
- Mechanical shaking

Acryllic and Teflon covered bags are the best in this application.It was found that other methods can be effectively used in the collection process. Wet and dry ESP and gravel bed filters are being used to optimize byproduct collecting system.

ESP and baghouse can be installed in series to increase the efficiency of the byproduct collection, but at a significantly higher cost of installation.

The usable byproduct is one of the major features of EB process for flue gas treatment. The concentration of ammonium sulfate and ammonium nitrate depends on the fuel composition, but its quality was estimated on 75% of the regular product. The sale of this byproduct can be used to offset the cost of the ammonia which is applied in the process. Such sale can significantly decrease operating costs.

TABLE 3. The basic parameters of the electron accelerators offered by the different producers for flue gas treatment in the capacity 10000-20000 Nm^3/h

TYPE OF ACCELERATOR	PRODUCER	ELECTRON ENERGY (keV)	BEAM CURRENT (mA)	OUTPUT WINDOW (mm)
600/200/1830, Dynamitron	Radiation Dynamics, USA/Japan	500/600	200	1830
ESI 0.3/90 Electrocurtain	Energy Scien. Corp., USA/Japan	300	300	1400
ELW3A Transformer	Inst. of Nucl. Phys., Russia/Japan	500/700	100	1500
UW-075-2-2-W, Transformer	NIIEFA, Russia	750	2 x 60	2000
EPS-500 Cascade	Nissin High Volt., Japan	500	80	1600
ESH, Transformer	Polimer Physics, Germany	280	220	700

Ammonium nitrate is the basic fertilizer for many plants. Ammonium sulfate is being applied directly on certain sulphur-depleting agricultural crops like corn and cotton. The combination of these two compounds provides a suitable quality material for direct application.

Ammonium sulfate is required by sulphur defficient lands, generally located in the more arid regions of the world. Existing ammonium sulfate sources do not meet market needs. Such lack translates into an excellent opportunity to sell EB process byproduct at an attractive price. Usually ammonium sulfate is a component of the final commercial product of the NPK fertilizer.

An alternative application of the EB process byproduct is under consideration. Enriching various organic compounds like sludge or municipal waste compost with a

byproduct addition may improve the nitrogen content, may adjust the precipitation of the mixture and may be effective and economically replace the chemical fertilizer.

Depending on the coal sulphur content and the level of nitrogen oxides in the flue gas, the nitrogen content of the byproduct mixture will be between 20 - 30%. For those facilities using 2.5% sulphur coal the byproduct production can be estimated on 800 Kg/day/MWe. With a nitrogen content of approximately 25%, the flyash is one of the significant compound of the byproduct. Usually it is efficiently removed by the ESP located before the process vessel. Presently, the flyash is not recognized as a hazardous waste material, but the high flyash content in the byproduct decreases the nitrogen content and increases the distribution and application costs per nitrogen unit (4).

Some trace of heavy metals are present in the flyash. Table 4 shows a record for two different byproduct samples. The byproduct was collected at the installation operated by Badenwork, Karlsruhe, Germany. Product A was a mixture byproduct with filtration, while product B is a pure EB process byproduct having the characteristics of a nitrogenous fertilizer, with properties and fertilizing utility similar to that of the ammonium sulfate. Usually the amounts of trace metal in the byproduct can be controlled at levels equal to or less to those being found in commercial fertilizers. Typically no more than 10%, by weight, of flyash by byproduct is accepted. This level of preremoval can be easily obtained by the use of relatively low efficient collectors.

TABLE 3. Composition and chemical properties of tested products.

	"A" Product	"B" Product
N total	4.45	19.50
$N\text{-}NE_4$	4.16	19.40
$N\text{-}NO_3$	0.75	0.74
P_2O_5	1.12	0.21
K_2O	1.21	0.07
CaO	3.95	0.50
MgO	2.74	0.46
Na_2	0.57	0.04
Cl	2.90	1.40
S total	3.95	25.50
$S\text{-}SO_4$	3.34	24.50
pH	7.35	4.50
% s.m. dry mass	98.20	99.50
including:		
R_2O_3	16.10	0.53
Fe_2O_3	1.27	0.11
SiO_2	43.31	0.89
ash	77.89	2.51
content of heavy metals (ppm)		
Mn	160	60.0
Zn	60	254.0
Cu	38	3.0
Pb	26	26.0
Cd	4	3.0
Cr	10	3.6

3.3. COST ESTIMATE

The costs including capital investment cost, operating and maintenance cost and byproduct credit should be taken into account to evaluate the EB process from the economic

point of view. For 100 MWe power plant 1000kW electron beam power should be applied to achieve 90% of SO_2 removal efficiency and 80% of NO_x removal efficiency at a 7kGy dose. The present status of accelerator development allows to build 500 kW units at a cost rate of 2-5 US$/W of beam power depending on the accelerator construction and its producer. Table 5 shows capital cost estimate depending on the cost of the accelerator. Up to 25% of thecapital cost is applied to buy accelerators what is slightly less than the typical cost of a construction work (buildings, ducts) (4).

According to an Ebara estimate to a 100 MW plant burning 2% sulfur coal and SO_2 removal rate 92% and the NO_x removal rate 60% listed below, performance and economic parameter can be achieved:

- Power consumption 2.6 MW/h
- Ammonia requirements 1500 kg/h
- Inert earth 100 kg/h
- Fertilizer byproduct 600 kg/h
- SO_2 reduction 1400---->112 ppm
- NO_x reduction 400----->160 ppm
- Flue gas - flow rate 300.000 Nm^3/h
- Total capital cost 19.300.000 US$
- Process cost 193 US$/kW
- Operating personnel 3 per 24 h
- Annual maintenance cost 200.000 US$
- Annual operating cost 580.000 US$

It was recognized that the byproduct has 75% of the value of a commercial fertilizer what meant 51 US$/t in1990.

TABLE 5. Estimate of the capital cost EB facility for flue gas treatment depending on the cost of the accelerator.

Accelerator Cost (USD/W) beam power	Investment Cost (USD/KWe)	Multistage Irradiation Investment Cost (USD/KWe) 0,75
2	225	169
5	350	262

3.4. LABORATORY INSTALLATION

A batch type laboratory unit with a flow system has been built in Japan, in Germany, in Poland and in some others countries to investigate experimental characteristics of the EB flue gas treatment process (2).

BATCH TYPE facility can be easily adopted to local experimental conditions. This type of laboratory unit was applied in Ebara during the first tests performed in 1970-71 to establish chemical reactions induced by radiation, responsible for SO_2 and NO_x removal from the flue gas.

FLOW SYSTEM incorporates flow gas stream rate lower than 1000 NM^3/h, generated by oil or city gas burners. The gas flow can be arranged by the use of pressure tanks containing NO, SO_2, O_2 and N_2 at a moderate flow rate. An additional amount of water should be incorporated to keep adequate water contents. Flue gas generated by both oil and

gas burners needs the additional injection of SO_2 and NO_x to meet appropriate experimental conditions. The choice between OIL BURNER, CITY GAS BURNER OR A GAS MIXING DEVICE depends mainly on finantial conditions or the possibility of adaptation of the existing facilities. The highest flow rate can be obtained in a system equiped with a boiler.

ANALYTICAL EQUIPMENT should allow to measure number of process parameters:

- Inlet and outlet SO_2, NO_x, O_3, H_2O, NH_3 concentration;
- Dose rate;
- NH_3, SO_2, NOx injection flow rate;
- Flue gas flow rate;
- Temperature in determined points of then facility;
- Aerosol parameters.

ACCELERATOR is used to provide stream of electrons which is applied in the process. Electron beam parameters are not critical in laboratory installations due to experimental requirements. The energy of an electron may range from 0.22 to 12 MeV while the energy beam power from 1.2 - 30 kW in laboratory installations which have been used to investigate the EB process.

PROCESS VESSEL should stand a long time irradiation with appropriate temperature according to the nature of the experimental condition. Stainless steel and other corrosion resistant materials are preferable. Thermal isolation and additional heating system could be used to stabilize experimental conditions.

HEATING EQUIPMENT is required to provide proper temperature conditions to the process vessel and analyze gas paths. Process vessel temperature 60 - 100^{o}C is being used for various experiments. The temperature of the gas paths is recommended by analytical instrument producers and usually is into the 150^{o}C range.

RETENTION CHAMBER located downstream of the process vessel is sometimes used to stimulate the product formation.

PREFILTER is sometimes used after the burner to stop particles coming from the combustion process.

HEAT EXCHANGER is sometimes used before the process vessel to control the temperature of the EB process.

SPRAY COOLER is used for the water injection from an air-assisted manifold of spray nozzles. The quantity of water injected is under control to cover temperature of the flue gas by the evaporation process and increase its relative humidity.

AMMONIA INJECTION is supplied from the pressure tank after conversion from liquid to gas phase. The amount of injected ammonia should be carefully controlled according to experimental requirements. The injection point is usually located before the process vessel.

COLLECTOR of the product is being used to collect the final product. Bag filters and/or ESP may be applied.

FAN located before the stack is necessary to keep proper flow rate of the gas through theprocess vessel and the collector of the product.

STACK and duct line are used to extract the flue gas out of the building. A corrosion effect and the deposition of the byproduct may occur when filter collector units are not applied.

A laboratory pilot plant has been built at IPEN-CNEN/SP, using an electron beam accelerator, from Radiation Dynamics Inc., having the following parameters (8):

- Electron energy ..... 0.5 - 1.5 MeV
- Beam current ..... up to 25 mA
- Scan length ..... 0.6 - 1.2 m
- Scan frequency ..... 100 Hz

The irradiation device allows a four-turn irradiation and was already used for dosimetric studies (1). The gas flow rate will be 25l/min and a synthetic mixture of SO_2 and NO_x will be used in preliminary studies. The carrier gas will be normal cooking gas, that is burned at a proper burner. NH_3 will also be injected and the fertilizer will be collected at a bag filter. Several points will allow the measurement and control of gas rate, temperature and humidity and also the analysis of the gases to calculate the efficiency of their removal.

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