

REDUCTION OF SO_x AND NO_x EMISSIONS BY ELECTRON BEAM FLUE GAS TREATMENT

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ABSTRACT

Sulphur and nitrogen oxides (SO_x and NO_x) emitted in flue gases by large industrial facilities contribute both to acid rain, low-level smog and indirectly to global warming. Society is concerned about public health being jeopardized by air pollution and the increase in health care costs as a result of the environmental and social impact. Consequently, regulatory authorities throughout the world are increasing the pressure on industry by reducing emission permits with respect to these substances. Pilot and industrial electron beam flue gas treatment (EBFGT) demonstration facilities of this emerging technology have demonstrated to simultaneously reduce SO_x up to 99.3% and NO_x up to 85%. Currently research is performed to add removal capability of other pollutants by means of catalytic reactions or other reacting agents. EBFGT for NO_x and SO_x is a dry process, which is catalyst free and produces valuable by-products: ammonium sulfate (NH₄)₂SO₄ and ammonium nitrate NH₄NO₃. These common fertilizer ingredients are formed in the EBFGT reactor and collected inside an ESP. Today the so-called clean burning fuels, such as natural gas, are increasingly expensive due to demand and supply restraints. In addition, natural gas must be considered to be a resource for higher value products such as fertilizer, hydrogen and others essential resources. EBFGT allows the use of "dirty fuels" such as coal or petcoke and reduces the pollution associated with these fuels to acceptable levels.

EBFGT has a twofold effect on indirectly reducing CO₂ emissions. Firstly, due to the fact that EBFGT produces fertilizer, emission trading offsets can be used, by calculating the reduction in CO₂ emissions from traditional fertilizer productions. Secondly, considering EBFGT as a retrofit solution, the high removal rates will allow users to place CO₂ capture technologies downstream of the EBFGT facility. Therefore, these facilities with EBFGT will be CO₂ capture ready.

INTRODUCTION

Since the 1970s, laboratories and pilot demonstrations by the Japan Atomic Energy Research Institute (JAERI), the University of Tokyo, have known the treatment of industrial flue gases by exposure to electron beams (EBFGT). Moreover, a number of installations have been built world wide since then, supported by the IAEA (International Atomic Energy Agency). This resulted in 2000 in the most recent facility – the treatment of up to 270 000 Nm³/h of flue gas - at the Pomorzany power station in Poland. This installation has proved operational feasibility that allows the integration of EBFGT facilities world-wide.

The EBFGT reactions are initiated inside the chemical reactor by the transfer of energy from accelerated electrons into the flue gas. Figure 4 outlines the sequence and time scales of the chemical reactions taking place. The energy absorbed by each component of the flue gas is proportional to its mass fraction. Experiments in dry air have shown that more than 90% of the beam energy results in dissociation and ionization of molecules, of which the majority are nitrogen. Oxidizing radicals OH and HO₂ together with excited species O(³P) are the most important products formed [8].

EBFGT can be applied to new and retrofit projects. It requires 5 key elements; a flue gas conditioning system, which adds water and ammonia to the flue gas. The conditioning system will require a storage holding anhydrous or aqueous ammonia delivering the ammonia to the gas flue gas treatment system. After conditioning the flue gas, the gas stream splits into multiples and is transferred into a chemical reactor. Attached to the chemical reactor an electron beam accelerator is mounted. The electron beam is scanning across the gas stream and irradiates the flue gas continuously inside the reactor. After triggering the chemical reaction, the flue gas is delivered to an electro static precipitator (ESP). The ESP is designed to remove and collect the by-product from the flue gas stream.

Dynamically changing legislation, such as green house gas legislations, will require the implementation of CO₂ capture options downstream of EBFGT installations. CO₂ capture must consider “electron beam multi pollution control processes” as being capable to treat flue gases in preparation for any downstream CO₂ treatment. Thus, it might be required to focus on specific pollutants, such as SO_x, in order to allow alternative downstream procedures to take place. It is known that CO₂ sequestration or catalytic reactions will require the removal of SO_x to very low concentrations. In addition, sequestration technologies (such as CO₂ compression) will require a very low temperature upstream stream of the flue gas compressor.

MAIN DISCUSSION

History

Since 1970, operational experience of a number of industrial electron beam treatment plants in China and Poland has demonstrated the advantages of using this EBFGT technology for simultaneous removal of SO_x and NO_x from industrial flue gas. The following table shows a short summary of the historical development of EBFGT:

Name	Location	Max. Flow Rate (m ³ /h)	Pollutant Concentrations (ppmv)	Years of Operation
JAERI	Takasaki, Japan	1		Since 1970
Ebara	Japan	10 000	200 SO ₂ /180 NO _x	1977 - 1978
Karlsruhe Research Centre (FZK)	Karlsruhe, Germany	300 1200	300 SO ₂ /300 NO _x 3000 SO ₂ /500 NO _x	1985 - 1995
Institute of Thermal Turbomachines (ITS)	Karlsruhe, Germany	1000	50 SO ₂ /500 NO _x	1985 - 1991
FZK	Karlsruhe, Germany	1000	VOC/dioxins	Since 1994
JAERI /NKK	Matsudo, Japan	1000	100 SO ₂ /100 NO _x /1000 HCl	1992
Ebara Corp.	Indianapolis, USA	24 000	1000 SO ₂ /400 NO _x	1984 - 1988
Badenwerk	Karlsruhe, Germany	20 000	50 SO ₂ /500 NO _x 300 SO ₂ /500 NO _x	1985 - 1989
Kaweczyn Power Station/Institute of Nuclear Chemistry and Technology (INCT)	Warsaw, Poland	20 000	250 SO ₂ /200 NO _x	Since 1990
Ebara Corp./EPA	Tokyo, Japan	50 000	0 - 5 NO _x	1992
Ebara/JAERI	Chubu, Japan	12 000	800 - 1000 SO ₂ /150 - 300 NO _x	1992
Ebara Corp.	Nishi-Nagoya, Japan	620 000		Since 1999
Ebara Corp.	Chengdu, China	300 000	2000 SO ₂ /400 NO _x	Since 1997
International Atomic Energy Agency (IAEA)/Pomorzany Power Station/INCT	Pomorzany, Poland	270 000	450 SO ₂ /300 NO _x	Since 2000

TABLE 1: EXTRACT OF EBFGT INSTALLATIONS WORLDWIDE [3]

The results of these installations proved the efficiency and viability of the electron beam treatment process and its application to multi pollution reduction capabilities. The electron beam technology was applied to coal power plants, steel mills and other industry sectors.

Equipment and Technology

Attractive capital costs for EBFGT, as well as a simplified process encourage retrofitting of boiler facilities with EBFGT. Regarding future facilities,

more integrated designs can be chosen with a number of options with respect to flue gas conditioning and reaction chambers. A full size EBFGT facility, as shown in Figure 1, consists of the following main components:

1. Conditioning tower
2. Ammonia storage
3. Electron beam accelerator
4. Chemical reaction chamber
5. ESP for by-product removal

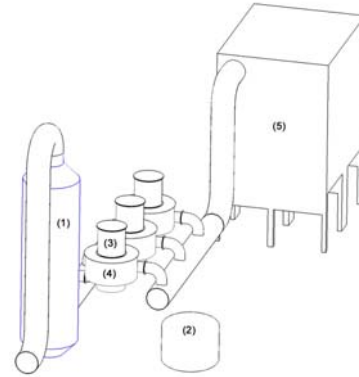


FIGURE 1: EBFGT CONCEPT

The major advantage of splitting the main gas stream into multiple sub-treatment streams is the ability to shut down one stream for maintenance, without disrupting the treatment process. Each reactor (4) has an in-flow guiding the gas stream and exposing the gas to the electron beam for a prolonged period. The electron beam (6), Figure 2, will scan across the gas stream and is pulsed in order to allow optimized energy control. By this process, synchronized with the beam scan, the electrons will transit through a segmented beam window from a vacuum pressure into the flue gas stream. A full scale EBFGT facility will allow each accelerator to be designed with its own power supply and chemical reaction chamber. Subsequently, the flue gas stream (7), Figure 2, will be divided into sub streams ranging from 100,000 Am³/h to 300,000 Am³/h.

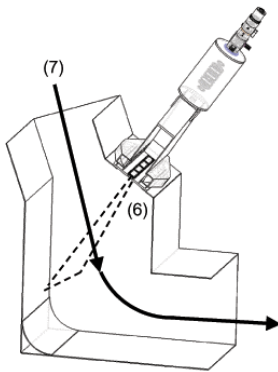


FIGURE 2: PAVAC – EBFGT REACTOR

The reaction chamber will have a square flue gas inflow opening. The size of the inflow will be designed to reduce the flue gas velocity to approximately 7.5 m/s. The accelerator will be interfaced with an angular incident to the gas direction in order to allow full penetration across and along the gas stream. The accelerator interface has segmented beam windows, which allow the electron beam to transit from the beam generation section into the flue gas chamber. After exposure of the flue gas to the electron beam, the flue gas will propagate the chemical reaction to form the NH₄NO₃ and (NH₄)₂SO₄ particulate.

The facility will require additional elements such as site preparation, piping and ducting and structural steel elements. There are a number of configurations available in order to accommodate the facility to site-specific requirements. In

many cases, the size of the facility is very important and, therefore, a key factor in the decision making process. Due to the accelerated reaction from the e-beam, the site has a smaller footprint and less steel components than convectional dry or semi-dry flue gas desulphurization (FGD) and selective catalytic reduction (SCR).

CO₂ Capture Requirements

In light of the worldwide focus on CO₂ as man-made cause of global warming, EBFGT is an important technological solution. Most CO₂ capture procedures require the removal of SO_x (sulfur) in order not to impact down stream CO₂ reactions or sequestration technologies.

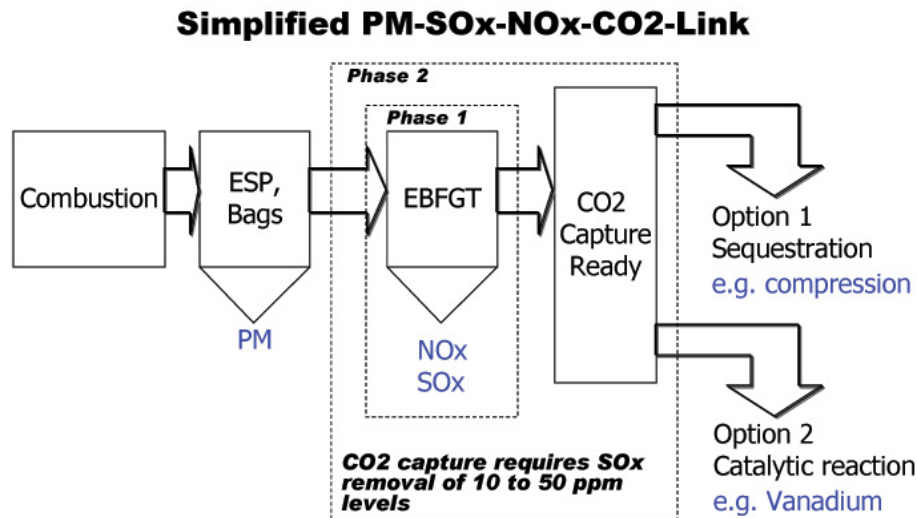


FIGURE 3: CO₂ CORRELATION WITH SO_x AND NO_x

Figure 3 outlines (in simplified version) the steps for an integrated flue gas treatment solution, aiming at a multi pollution removal. EBFGT technology can be staged in 2 phases. In phase 1, the facility is targeting the removal of SO_x and NO_x emissions in order to comply with future emission legislations. In phase 2, the removal rates for SO_x are optimized to allow CO₂ capture technology downstream of EBFGT, such as sequestration or catalytic reactions.

Chemical Reaction

The electron beam energy is scanned and absorbed in the flue gas in proportion to its mass fraction. It is then consumed in the ionization, excitation and dissociation of the molecules and results in the formation of active free

radicals $\cdot\text{OH}$, $\text{HO}_2\cdot$, O , N and H . These radicals oxidize SO_2 and NO to SO_3 and NO_2 which in reaction with water vapor, present in the stack gas, form H_2SO_4 and HNO_3 , respectively. The acids react with added ammonia to form ammonium sulfate $((\text{NH}_4)_2\text{SO}_4)$ and ammonium sulfate-nitrate $((\text{NH}_4)_2\text{SO}_4 \cdot 2\text{NH}_4\text{NO}_3)$. These components are recovered as dry powder inside a particle collector (e.g. Electro Static Precipitator).

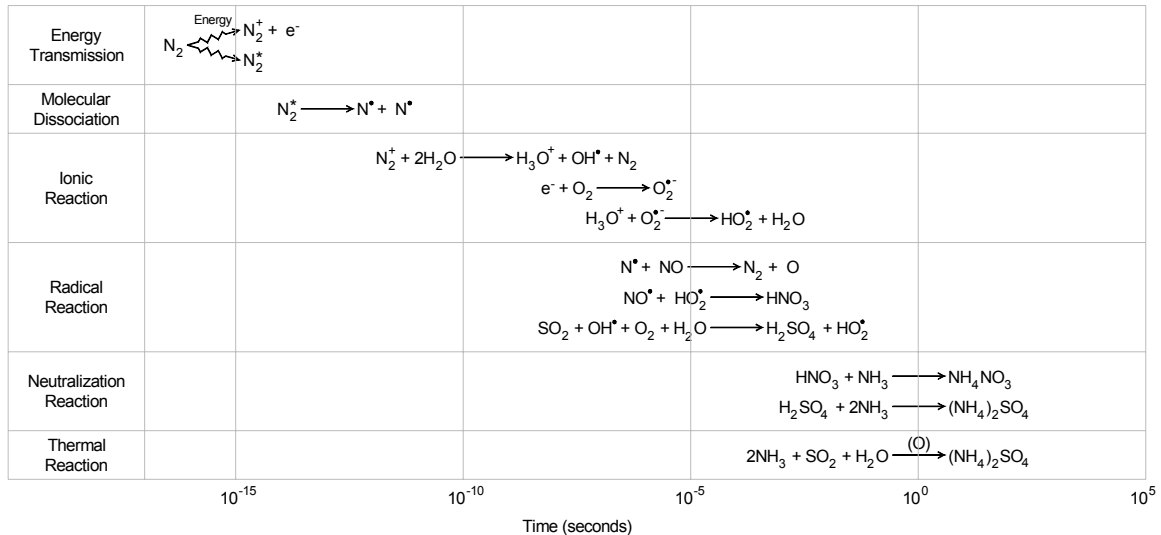


FIGURE 4: REACTION SEQUENCE AS OUTLINED FROM [3]

The stoichiometric ratio of the ammonia conditioning is in relation to the pollution concentration in the flue gas stream. The complexity of the process lays in the combination of pollution concentration to ammonia ratio and temperature of the flue gas. The flue gas is conditioned and cooled to 60° to 80° C measured at the entry point of the chemical reactor.

Economic Sustainability

Concurrently with the technical solutions presented by the EBFGT technology, the value proposition to the end user must be analysed. Electron beam technology can establish significant financial and economic benefits in many ways. Some of the key benefits are the more efficient utilization of plant infrastructures, possible lower overall fuel costs, and the selling of a by-product (fertilizer).

Stabilizing spot market prices of fuels will improve the sustainability of the operation. In the past, fuel prices experienced considerable fluctuations due to seasonal cycles, and escalated on account of unforeseen events such as hurricanes and other natural disasters. EBFGT might allow to reduce costs and

to combust locally available dirty fuels, such as high sulphur fuels, without exceeding emission limits. Emission caps will result in the restriction of facility outputs, in particular of facilities lacking emission control technology. The incurring costs of such shut downs can be expressed as “costs of lost opportunity to deliver the product”. The following chart shows the savings in fuel costs per year (assuming various prices of natural gas versus the costs for petcoke)

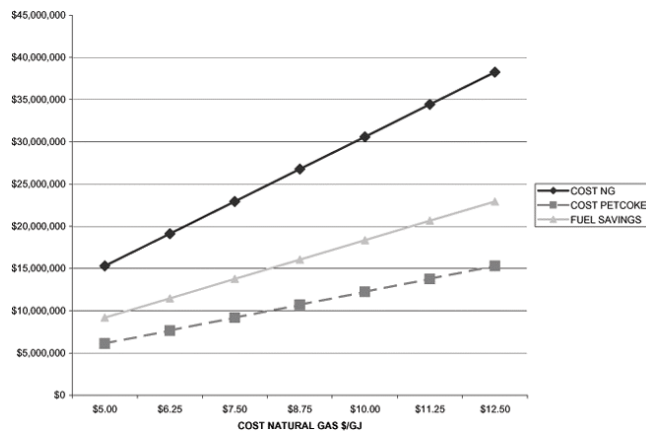


FIGURE 5: SAVINGS FROM FUEL SWITCHING

Figure 5 assumes a 100MW_{th} plant at 8,500 hours annual operation. Costs for natural gas and petcoke are calculated on the bases of 3,060,000GJ consumption per year. The gas prices varies from 5\$/GJ to 12.50\$/GJ. Spot market prices for natural gas showed short-term prices of 15.00\$/GJ. Petcoke prices vary from 2\$/GJ to 5\$/GJ. At a current natural gas price of 10.00\$/GJ switching the fuel might result in \$18,360,000 savings annually.

Not included in this calculation are emission trade revenues from removal of pollution and the profits from trading the fertilizer.

CONCLUSIONS

EBFGT technology is a building block towards a multi pollution control procedure that provides economic viability. The treatment of flue gas and the simultaneous removal of sulfur oxide and nitrogen oxides are key steps towards pollution reduction, and are achievable technological goals with minimized financial impact on current operations. In addition, this technology is designed for existing plants as retrofit application as well as for new facilities.

Sustainability in many industrial sectors is directly connected with the costs of energy. Therefore, all products that are energy intense operations will be looking for cost reduction in fuels. Mostly “lower cost fuels” are considered dirty and will require advanced pollution control that can be flexible and adjust to multiple fuels. EBFGT can be dynamically controlled to allow varying pollution concentrations and changing flue gas volumes. Conventional scrubbing installations have shown to be less efficient under changing conditions.

The fuel flexibility combined with attractive return of investments makes the electron beam flue gas treatment very attractive and guarantees long-term sustainability. EBFGT can accommodate high removal rates for SO_x and NO_x and, therefore, satisfy CO_2 preparation requirements.

FIGURES

FIGURE 1: EBFGT CONCEPT

FIGURE 2: PAVAC – EBFGT REACTOR

FIGURE 3: CO₂ CORRELATION WITH SOX AND NOX

FIGURE 4: REACTION SEQUENCE AS OUTLINED FROM [3]

FIGURE 5: SAVINGS FROM FUEL SWITCHING

TABLES

TABLE 1: EXTRACT OF EBFGT INSTALLATIONS WORLDWIDE [3]

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