Electron Beam Flue Gas Treatment (EBFGT) Technology for Simultaneous Removal of SO_2 and NO_x from Combustion of Liquid Fuels: Technical and Economic Evaluation

A.A. Basfar*[1], O.I. Fageeha[2], N. Kunnummal[2], A.G. Chmielewski[3], J. Licki[4], A. Pawelec[3], Z. Zimek[3]

[1]King Abdulaziz City for Science and Technology, Riyadh, Saudi Arabia.
[2]Saudi Arabian Oil Company, Dhahran, Saudi Arabia.
[3]Institute of Nuclear Chemistry and Technology, Warsaw, Poland.
[4]Institute of Atomic Energy, Otwock-Świerk, Poland.
*Corresponding author e-mail: abasfar@kacst.edu.sa; tel.: +9661-4813648; fax: +9661-4813640

Abstract

Emission of harmful compounds to the atmosphere caused by combustion of fossil fuels is an environmental concern all over the world. Sulfur and nitrogen oxides are among the most alarming pollutants due to their great global emission and long range transport in the atmosphere. Apart from conventional technologies designed for removal of one pollutant in one process, simultaneous technologies being able to remove two or more pollutants in the same process are being developed. Among such emerging technologies, Electron Beam Flue Gas Treatment (EBFGT) technology is the most promising and was already implemented in few industrial plants in different parts of the world. In order to evaluate the potential of implementing EBFGT technology in the case of flue gas generated during fuel oil combustion, Saudi Arabian Oil Company (Saudi Aramco) initiated a project entitled "Feasibility Study for Electron Beam Flue Gas Treatment (EBFGT) at Oil Fired Boiler" which was conducted by King Abdulaziz City for Science and Technology (KACST) in Saudi Arabia and Institute of Nuclear Chemistry and Technology (INCT) in Poland. From technical evaluation, SO_2 removal is mainly dependent on ammonia stoichiometry, flue gas temperature and humidity and irradiation dose up to 8 kGy. NO_x removal depends primarily on irradiation dose. High removal efficiencies up to 98% for SO_2 and up to 82% for NO_x were obtained under optimum conditions. From economic evaluation, EBFGT is highly competitive to conventional pollution control technologies.

1. INTRODUCTION

The highest percentage of electrical energy and heat (88%) is produced by the combustion of fossil fuels like coal, oil and natural gas [1]. Many pollutants are released in the combustion process. These pollutants affect air quality, human health, environment, economy and contribute to climate change. Liquid fuels are used in some limited applications, but are more prevalent in certain areas of the world such as South America, central and eastern Canada, northeastern states of USA. Heavy fuel oil (HFO) is a mixture of hydrocarbons composed of residual fractions from distillation and processing of crude oil. It is essentially an industrial fuel that is suitable for use in thermal power plants, oil refineries, industrial boilers, pulp and paper industry and metallurgical operations which generally pre-heat the fuel oil. Depending on the source, the sulfur content in the HFO could be as high as 4.0 % wt. EBFGT technology is among the most promising advanced technologies of a new generation. It is a simultaneous dry-scrubbing of SO_2 and NO_x, where no waste, except a useful by-product, is generated. The irradiation of flue gas with fast (300-800 keV) electrons initiates chemical changes that make removal of sulfur and nitrogen oxides easier. After irradiation, fast electrons interact with the main components of flue gas (N_2, O_2, H_2O and CO_2) and generate the same oxidants (OH^*, HO_2^*, O, O_3) which are produced by UV sunlight in the free atmosphere, but at concentrations several orders of magnitude higher. The oxidants convert NO_x and SO_2 to nitric and sulfuric acids which form a solid powder of ammonium nitrate and sulfate in the presence of ammonia which is added to flue gas before its irradiation. The filtered by-product is usable as an agricultural fertilizer. Overviews of the process chemistry and model calculations were given by Tokunaga and Suzuki (1984) [2], Mätzing (1991) [3] and Namba (1993) [4]. High removal efficiencies of SO_2 up to 95% and NO_x up to 85% were simultaneously obtained at the optimum conditions for EBFGT. Three industrial installations have already been built at coal-fired Thermal Power Plants of Chengdu and Hangzhou in China [5] and Pomorzany in Poland [6]. The tests demonstrated the ability of the technology for highly efficient removal of SO_2 and NO_x from flue gas from coal combustion process. New industrial plants are under construction in China and Bulgaria.

Technical solutions for two options of electron beam facility for treatment of flue gas from heavy oil burning boilers were elaborated. The first option was assumed for flow rate of 65 000 Nm^3/h, while the second one for 130 000 Nm^3/h. On this basis, economic evaluation of investment and operation cost of the installation were estimated. Factors affecting costs of the installation are presented in this paper. The primary objective of this paper is to demonstrate the feasibility of the electron beam flue gas treatment process for simultaneous removal of SO_2 and NO_x from flue gas from combustion of heavy liquid fuels. In addition to technical evaluation, this paper presents economic feasibility of EBFGT technology based on selected parameters and assumptions.

2. EXPERIMENTAL

2.1. Technical Viability

The experiments were performed using a laboratory-scale EBFGT installation at Institute of Nuclear Chemistry and Technology (INCT) in Warsaw, Poland. For this study, the installation was equipped with fuel oil combustion unit. The laboratory test facility consists of the following technological units: fuel oil combustion unit, flue gas conditioning system, electron beam irradiation of flue gas, filtration unit, monitoring system and exhaust of purified flue gas. The details of operation of these units are presented elsewhere [7].

Three grades of Arabian liquid fuels were combusted: Arabian Medium Crude Oil (AM), Arabian Heavy Crude Oil (AH) and Arabian Heavy Fuel Oil (HFO). A portable flue gas analyzer type Lancom II, manufactured by Land Combustion (U.K.), measures the concentrations of CO, CO_2, O_2, SO_2, NO, NO_2, NO_x, and hydrocarbons in the flue gas was used in this study. Such measurements were performed in the flue gas at outlet of oil burner and occasionally at process inlet and outlet to check readings of both CEM systems. The temperature of flue gas at six essential points of experimental set-up was measured and controlled using K thermocouples. The humidity of flue gas was determined by manual analytical method based on EPA method 4 using u-tubes filled with granular silica gel.

Electron beam energy absorbed in the flue gas, defined as an absorbed dose in units of gray (Gy=J/kg), was estimated using a cellulose triacetate (CTA) tape of 8 (w) x 0.125 (t) mm (Fuji photo film, FTR-125, Japan) [8].

2.2. Economic Feasibility

Due to differences of flue gas parameters compared to flue gas from coal combustion, some new solutions were elaborated. The proposed EBFGT installation is to be composed of components such as:

- Gas-gas heat exchanger
- Flue gas conditioning unit integrated with dust removal system
- Ammonia storage and dosing system
- Reaction unit including reaction vessel, accelerators with supporting structure and biological shielding of radiation
- By-product handling and storage unit with electrostatic precipitator
- Flue gas ducts and auxiliary fan
- Control and monitoring system.

The conceptual scheme of proposed EBFGT installation, the details of these units and selected parameters are presented elsewhere [9].

3. RESULTS AND DISCUSSION

3.1. Technical Viability

Measurement of the emission of SO_2 and NO_x from the burning of these fuels was the first task of this work. The measurements of flue gas composition were performed at the process inlet for each grade of fuel. Table 1 summarizes averaged volumetric emission concentrations for all tested fuels. A 10% of light oil (LO) was added to HFO to facilitate combustion process. Observations from these measurements are as follows:

- Very high level of SO_2 emission. All SO_2 concentrations were above 1200 ppmv while the sulfur content in the burned oils was above 2.7% wt.
- NO_x emissions ranged from 150 to 170 ppmv.
- Oxygen content in the analysed flue gases ranged from 3.4 to 4.4% vol.
- Natural humidity of flue gas was higher than 8.48% vol.

To evaluate SO_2 and NO_x emissions under comparable conditions, the measurements were normalized to 3% vol. oxygen and dry gas basis conditions. The data indicate that SO_2 emissions from combustion of these high-sulfur fuel oils were very high, about 1220 ng/J, which if released untreated, will adversely impact the ambient air quality standards. Emissions of NO_x were also high, in the range of 110 to 120 ng/J. In order to meet air quality standards, these flue gases require post-treatment with high SO_2 and NO_x removal efficiencies. The parameters which influence removal efficiency of SO_2 and NO_x were evaluated for flue gas obtained from combustion of the three Arabian fuels.

Effect of electron beam dose on SO_2 and NO_x removal

Figure 1 presents the dose dependence of SO_2 and NO_x removal efficiency. The absorbed dose is the primary factor influencing NO_x removal efficiency. The reaction starts at zero efficiency for zero dose leading to a saturation at high dose. This demonstrates that NO_x removal is a radiation-induced process. Higher NO_x removal was achieved with higher absorbed doses because the amount of NO_x molecules removed corresponds to the amount of active species formed by electron beam irradiation of flue gas.

TABLE 1. COMPOSITION OF FLUE GAS FROM COMBUSTION OF THREE ARABIAN FUELS MEASURED AT PROCESS INLET

Parameter	Unit	AM	AH	HFO+10%LO
SO_2	ppmv	1215	1270	1250/1355*
NO_x	ppmv	153	168	169
O_2	% vol.	4.40	3.42	3.78
H_2O	% vol.	8.48	9.06	9.58
CO	ppmv	0	0	36
CO_2	% vol.	12.4	15.5	13.6
C_xH_y	% vol.	0.18	0.16	0.17
Sulfur content in oil	% wt.	2.81	2.90	2.7/3.1

* SO_2 concentration recalculated for combustion of HFO only.

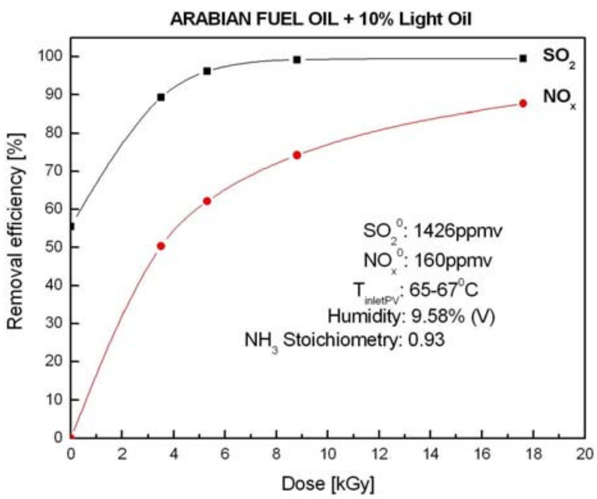

FIG.1. SO_2 and NO_x removal efficiency as a function of dose from combustion of Arabian Heavy Fuel Oil +10 % Light Oil.

Effect of ammonia stoichiometry

Figure 2 presents the SO_2 and NO_x removal efficiency obtained in two experiments with combustion of Arabian Medium Oil. First experiment was performed with electron beam treatment of the flue gas without any additives (without NH_3). Low SO_2 and NO_x removal efficiencies were obtained. In the second experiment, gaseous ammonia was added to flue gas before its inlet to reaction vessel. The NH_3 stoichiometry was equal to 0.90. In this case higher removal efficiencies of SO_2 and NO_x were obtained. Ammonia addition significantly influences SO_2 removal.

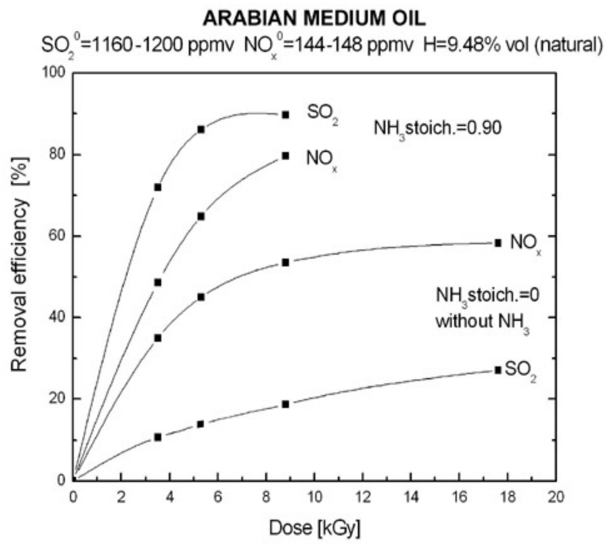

FIG. 2. SO_2 and NO_x removal efficiencies as a function of dose with and without additional ammonia to flue gas for the combustion of Arabian Medium Oil.

Effect of flue gas humidity

Two experiments for EBFGT at different humidity levels were preformed. Flue gas with natural humidity (9.62%vol.) was irradiated in the first experiment. Additional water vapor was injected to the flue gas in the second experiment to increase its humidity to 11.57 % vol. Figure 3 presents the SO_2 and NO_x removal efficiencies obtained in both experiments. The increase of flue gas humidity does not affect NO_x removal. The SO_2 removal efficiency increases markedly with the increase in humidity of flue gas. This increase is due to the thermal reaction of SO_2 with ammonia in the absence of electron beam irradiation.

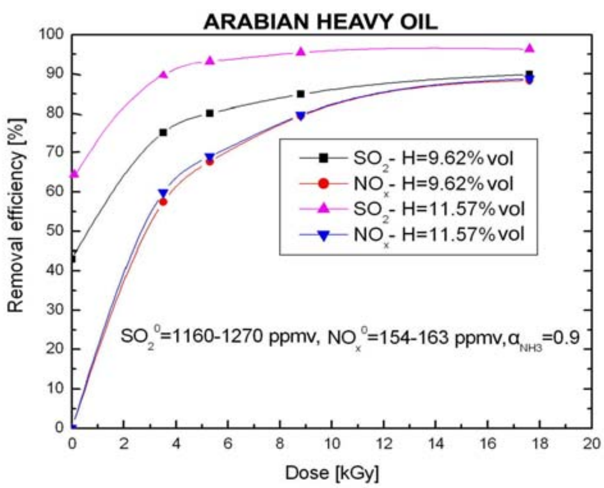

FIG. 3. Effect of humidity on the SO_2 and NO_x removal efficiencies for combustion of Arabian Heavy Oil.

By-product analysis

In each experiment, a by-product was collected in the bag filter. The by-product (white solid particles) was analyzed using ion chromatography 2000i/SP (Dionex, USA). The major constituents of by-products were sulfate, nitrate, and ammonium ions. The concentrations of these constituents varied depending on the type of fuel (i.e. AM, AH, or HFO). Ion chromatography analyses indicate that the by-product consists primarily of ammonium sulfate and ammonium nitrate.

Optimization study

Based on the evaluation of parameters which influence EBFGT process, it was possible to establish that the optimum parameters for highest SO_2 and NO_x removal efficiencies are as follows:

- Dose: 6 kGy for SO_2 removal only and 12 kGy for simultaneous SO_2 and NO_x removal.
- Ammonia stoichiometry: 0.95.
- Gas temperature at inlet to process vessel: 65^{o}C.
- Gas humidity: 11.5 % vol.

The chemical composition of the by-product suggests that it is suitable as an agricultural fertilizer.

3.2. Economic Feasibility

Total investment cost of EBFGT installation planned at a selected oil refinery was evaluated for 13 800 000 USD in the case of option 1 (flue gas flow rate 65 000 Nm^3/h) and 19 200 000 USD in the case of option 2 (flue gas flow rate 130 000 Nm^3/h). The elements of investment cost are presented in Table 2.

The investment cost evaluated in this paper was elaborated according to best available data including offers from equipment manufacturers (ESP, accelerators) and it meets the cost level of such investment. The design criteria determined minimum availability of electron beam flue gas treatment installation in a selected oil refinery for 8000 hours/year. Calculations of annual operation cost of the installation were performed assuming 95% availability which gives 8320 hours/year for continuous operation of oil refinery.

The expenses will be partly compensated by the sale of obtained by-product for agricultural purposes. Usually income from sale of by-product shall cover expenses of raw materials. By-product yield was calculated to be 460 kg/hour and 920 kg/hour depending on the option of installation. The income depends on the price negotiated with the by-product buyer. Assuming mean market price of ammonium sulfate 135 USD/ton, the income will reach 516 000 USD/year for option 1 and 1033 USD/year for option 2.

Total annual operation cost of proposed EBFGT installation will reach 558000 USD/year for option 1 and 681000 USD/year for option 2. The costs shown above are direct annual expenditures connected with operation of EBFGT installation and don't cover any non cash expenditures as amortization or bank credit costs.

Comparison with Conventional Technologies

The removal of SO_2 and NO_x from flue gas is normally realized by combination of de-SO_2 and de-NO_x methods. The removal efficiency of both systems (i.e. electron beam and combined wet FGD and SCR) is comparable. The desulfurization efficiency is at least 95% (achieving 98%) in the case of EBFGT technology, while it is reported at the level of 95 –

99% in the case of wet FGD technique. Similarly, NO_x removal efficiency based on EBFGT technology is at the level of 70% (in pilot plant it was reported up to 80%) while SCR efficiency is usually 70 – 80%. The efficiency of SNCR process is much lower.

TABLE 2. INVESTMENT COST OF EBFGT INSTALLATION AT A SELECTED OIL REFINERY

Item	Description	Cost [1000 USD]	
		Option 1 (65 000 Nm^3/h)	Option 2 (130 000 Nm^3/h)
1	Site engineering and design	1 000	1 450
2	Heat exchanger	530	800
3	Gas conditioning unit	80	110
4	Ammonia storage and dosing	510	770
5	Accelerators	1 600	2 000
6	Reactor with radiation shielding	800	1 100
7	By-product ESP	2 600	4 000
8	By-product handling and storage	610	830
9	Auxiliary fan	300	400
10	Ducts and piping	480	720
11	Electric power supply system	400	600
12	Control and monitoring system	1 100	1 600
13	Structural elements	450	680
14	Land development	590	890
15	Supervision, training, start-up	1 500	1 500
16	Reserve (10%), spare parts	1250	1 750
TOTAL		13 800	19 200

In the case of SCR methods, the operation costs are reported to be in the range of 3800 – 4600 USD/MWe (from 2 667 000 USD for 700 MWe unit to 580 000 USD for 125 MWe unit) [10]. Similar to capital cost, SNCR operation costs are about 1/3 lower than SCR costs.
In the case of flue gas treatment from oil fired boiler without modification of boiler itself, the most useful and popular solution is a combination of wet SO_2 scrubbing and removal of nitrogen oxides by SCR or SNCR method. The investment and operation costs of such combination are presented in Table 3.

The data are given for 300 MW and larger units. One should remember that for smaller units, the unit cost should be multiplied two – three times. According to another report [11], the cost of combined wet FGD and SCR system is estimated at 270 – 474 USD/kWe for units of 300 – 50 MWe. Annual operation cost varies from 15 000 to over 30 000 USD/MW.

ACKNOWLEDGMENTS

This work was financed by Saudi Arabian Oil Company (SAUDI ARAMCO) - Project No. 6600011081 entitled " Feasibility Study for EBFGT at Oil Fired Boiler".

TABLE 3. INVESTMENT AND OPERATION COSTS OF COMBINATION OF CONVENTIONAL DE-SO_2 AND DE-NO_x METHODS

Flue gas treatment method	Investment cost [USD/kW]	Annual operational cost (variable) [USD/MW]
Wet SO_2 scrubbing:	190 – 240	12 750 to 28 050
Selective Catalytic Reduction	60 – 110	3 800 – 4 600
Selective Non Catalytic Reduction	15 – 30	2 500 – 3 000
Combination of methods:		
Wet de-SO_2 + SCR	250 – 350	16 550 – 32 650
Wet de-SO_2 + SNCR	205 – 270	15 250 – 31 050

REFERENCES

[1] Baukal, Ch. E., *Industrial Combustion Pollution and Control*; Marcel Dekker, Inc: New York (2004).

[2] Tokunaga, O., Suzuki, N., Radiation chemical reactions in NO_x and SO_2 removals from flue gas. *Radiat. Phys. Chem*. 24 (1984) 145-165.

[3] Mätzing, H., Chemical kinetics of flue gas cleaning by irradiation with electron. *Advances in Chem. Phys*. 80 (1991) 315-402.

[4] Namba, H., EB processing of flue gas-chemistry. In *Regional Training Course on Radiation Technology for Environmental Conservation*; UNDP/IAEA/RCA, Takasaki, Japan (1993) 99-104.

[5] Mao, B.J., Process of flue gas desulfurization with electron beam irradiation in China. In Radiation Treatment of Gaseous and Liquid Effluents for Contaminant Removal, IAEA-TECDOC-1473, Vienna (2005) 45-51.

[6] Chmielewski, A.G., Licki, J., Pawelec, A., Tymiński, B., Zimek, Z., Operational experience of the industrial plant for electron beam flue gas treatment. *Radiat. Phys. Chem*. 71 (2004) 439-442.

[7] Basfar A.A., Fageeha O.I., Kunnummal N., Al-Ghamdi S., Chmielewski A.G., Licki J., Pawelec A., Tyminski B., Zimek Z., Electron Beam Flue Gas Treatment (EBFGT) Technology for Simultaneous Removal of SO2 and NOx from Combustion of Liquid Fuels, Fuel, 87 (2008) 1446 – 1452.

[8] American Society for Testing and Material. *Annual Book of ASTM Standards, Vol. 12.02 Nuclear, Solar and Geothermal Energy*; ISO/ASTM 51650; American Society for Testing and Material, Philadelphia, (2002).

[9] Basfar A.A., Fageeha O.I., Kunnummal N., Al-Ghamdi S., Pawelec A., Chmielewski A.G., Licki J., Tyminski B., Zimek Z., Economic Feasbility of EBFGT Technology for Removal of Pollutants from Combustion of Liquid Fuels, IAEA Technical Meeting on " *Prospects and Chllenges in Application of Radiation for Treating Exhaust Gases*" (2007) (in Warsaw, Poland).

[10] Demonstration of selective catalytic reduction technology for the control of NO_x emissions from high-sulfur, coal-fired boilers, Department of Energy Project Fact Sheet (2003), http://www.netl.doe.gov/cctc/factsheets/scr/selcatreddemo.html

[11] Integrated Dry NO_x/SO_2 Emissions Control System, Department of Energy Project Fact Sheet (2003), http://www.netl.doe.gov/cctc/factsheets/pscol/intryemdemo.html