

A review on electron beam flue gas treatment (EBFGT) as a multicomponent air pollution control technology

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Abstract. Electron beam flue gas treatment (EBFGT) technology for coal-fired boilers has been implemented on an industrial scale in two thermal power plants in China and at the Electropower Station (EPS) Pomorzany in Poland. The plants in China have been designed mainly for desulfurization while the plant in Poland for a simultaneous removal of SO₂ and NO_x from flue gases. The successful operation of these plants has demonstrated the advantages of using this technology for removing SO₂ and NO_x from flue gas under varying conditions. At present, the plant in Poland is the only operational installation at an international level. Recent tests performed at an EBFGT industrial pilot plant in Bulgaria have demonstrated feasibility of application of this technology for treatment of high sulfur and high humidity lignite fired boilers. Further laboratory tests have been performed for model flue gases similar to those emitted from a copper smelter and flue gases originated from different types of high sulfur heavy fuel oils. In all cases, dry-scrubbing process with ammonia addition has been tested. The removal efficiency of pollutants is as high as 95% for SO₂ and 70–80% for NO_x. The by-product of this process is a high quality fertilizer component. Additional laboratory studies have shown that volatile organic compounds (VOCs) emitted during combustion of fossil fuels, can be degraded as well. Therefore, EBFGT can be considered as a multicomponent air pollution control technology which can be applied to flue gases treatment from coal, lignite and heavy fuel oil-fired boilers. Other thermal processes like metallurgy and municipal waste incinerators are potential candidates for EBFGT technology application.

Key words: fossil fuel combustion • heavy fuel oil • electron beam irradiation • plasma processes • flue gas treatment • SO₂ and NO_x removal

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Introduction

Most the world-wide of energy (88%) is produced by the combustion of fossil fuels like oil, natural gas and coal [2]. All of these fuels are composed of major constituents such as carbon, hydrogen and oxygen and other components including sulfur and nitrogen compounds and metals. During the combustion process, different pollutants are emitted such as fly ash (containing diverse trace elements (heavy metals)), SO_x (SO₂ and SO₃), NO_x (NO_x = NO + NO₂), Hg and VOCs. Air pollution caused by particulate matter and other pollutants not only directly impacts the atmospheric environment but also contaminates water and soil, leading to their degradation. The exhaust gases released into atmosphere are irradiated by ultraviolet (UV) sunlight and undergo photochemical transformations. Wet and dry deposition of inorganic pollutants leads to acidification of the environment. These phenomena affect human health, increase corrosion and destroy crops and forests. VOCs emissions to the atmosphere can lead to ground level photochemical ozone formation and can have toxic or carcinogenic effects on human health.

Among conventional technologies for flue gas treatment aimed at removal of SO₂ and NO_x are wet, dry and semi-dry flue gas desulfurization (FGD) and selective catalytic reduction (SCR) of NO_x. VOCs are usually adsorbed on active carbon, but this process is rarely used for lean hydrocarbon concentrations. All

these technologies are complex chemical processes, and wastes, like wastewater, gypsum and used catalyst or adsorber, are generated [20].

EBFGT technology is among the most promising advanced technologies of new generation. This is a dry-scrubbing process of simultaneous SO_2 and NO_x removal, where no waste, except a by-product, is generated. The application of electron beam irradiation to initiate chemical reactions to remove SO_2 and NO_x was first investigated by joint research of the Japan Atomic Energy Research Institute (JAERI) and Ebara Corporation in the early 1980s. The method has been developed since then from the laboratory to pilot and a large demonstration scale by research and development projects in Japan, USA, Germany, China and Poland [11]. The final engineering design technology for industrial applications was achieved at pilot plants operating in Nagoya, Japan [17] and at Kawęczyn, Poland [6]. In the case of the latter, new engineering solutions were applied: double-longitudinal gas irradiation, an air curtain separating the secondary window from corrosive flue gases and modifications of humidification and ammonia injection system (high enthalpy water or steam injection, ammonia water injection) and others. A high irradiation dose is required for NO_x removal, while SO_2 is removed in proper conditions at low energy consumption [6].

Technology status

EBFGT technology for coal-fired boilers has been implemented on an industrial scale at the Thermal Power Plants (TPPs) at Chengdu [9] and Hangzhou [16] in China and at the EPS Pomorzany [4] in Poland. At present, the EBFGT installation in Poland is the only operational installation in the world. Table 1 presents the main parameters of these installations.

The plants in China were designed mainly for SO_2 removal, while the plant in Poland was designed for simultaneous SO_2 and NO_x removal. The flue gases from two Benson boilers (65 MW(e) and 100 MW(th) each) are purified at the EPS Pomorzany in Poland (see Fig. 1). The maximum flow rate of the gases is 270 000 Nm^3/h and the total electron beam power is equal to 1 MW. There are two process vessels over which two electron accelerators are installed in series (700 keV, 260 kW each). The applied dose is in the range 7–12 kGy (kJ/kg). At these doses, the removal efficiency approaches 85–95% for SO_2 and 50–70% for NO_x . The by-product is collected by an electrostatic precipitator and shipped to a fertilizer plant.

The most often applied conventional technology for flue gas treatment is a combination of wet FDG and SCR processes. A comparison of the costs of various emission

Table 1. Main parameters of industrial EB installations

Parameter	Unit	Chengdu TPP, China	Hangzhou TPP, China	Pomorzany EPS, Poland
Flue gas flow rate	Nm^3/h	300 000	305 400	270 000
Inlet flue gas temperature	$^{\circ}\text{C}$	150	145	140
Inlet SO_2 concentration	mg/Nm^3	5150	2770	2000
Inlet NO_x concentration	mg/Nm^3	820	410	600
SO_2 removal efficiency	%	80	85	90
NO_x removal efficiency	%	18	55	70
EB		800 keV, 320 kW \times 2	800 keV, 320 kW \times 2	700 keV, 260 kW \times 4

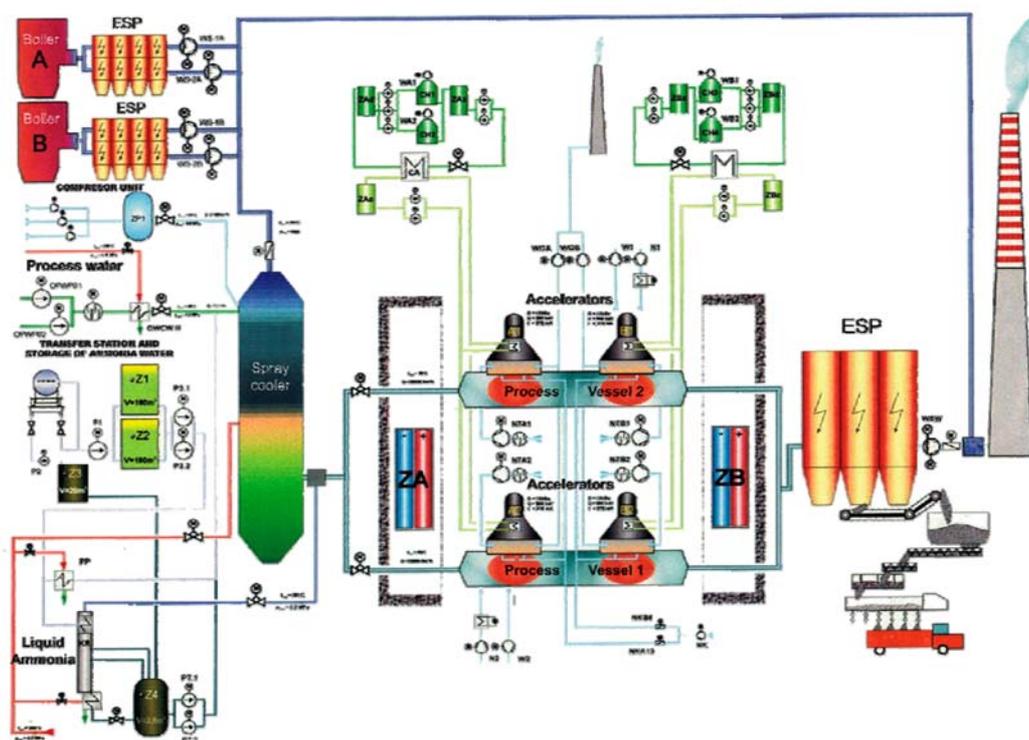


Fig. 1. Flow diagram of the industrial plant at the EPS Pomorzany, Poland.

Table 2. Costs of various emission control methods for retrofit 120 MW(e) unit [14]

Emission control method	Investment cost USD/kW(e)	Annual operational cost USD/MW(e)
Wet flue gas desulphurization	120	3000
Selective catalytic reduction	110	4600
Wet FGD + SCR	230	7600
EB treatment EBFGT	160	7350

control methods for a 120 MW(e) unit is presented in Table 2.

The operation of the EBFGT industrial plants in China and Poland for the last few years has demonstrated advantages of using this technology for simultaneous removal of SO_2 and NO_x from flue gas under varying conditions. EBFGT has to be competitive with the conventional ones both from economic and technical point of view. Therefore, any development concerning cost reduction and technological improvements regarding technical components, especially reliability of electron accelerator plays a very important role for further EBFGT process implementation.

Potential applications for industrial off-gases

Combustion of fuel oil

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, north-eastern USA. No. 2 and no. 6 oils are most commonly used liquid fuels. Heavy fuel oils (HFO) are 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, refineries, industrial boilers, pulp and paper industry, marine applications and metallurgical operations which generally preheat the fuel oils. Depending on the source, the sulfur content in the HFO could be as high as 4.0 wt.%. Sulfur is converted to sulfur dioxide when oil is burnt. Flue gas emitted from such heavy fuel oil-fired boiler contains high SO_2 and NO_x concentrations, many times exceeding the permissible emission limits, which necessitates the use of add-on control devices for the reduction of SO_2 and NO_x emissions.

A study of EBFGT on flue gas from combustion of high sulfur heavy fuel oils was performed at the laboratory plant at the Institute of Nuclear Chemistry and Technology (INCT) in Warsaw [1]. Figure 2 presents the flow diagram of the laboratory plant. Three types of high sulfur (about 2.9 wt.% of sulfur) heavy fuel oils with different viscosities were combusted. The flue gases contain high concentrations of SO_2 , above 1100 ppmv and NO_x above 150 ppmv. These flue gases were irradiated by an EB from an accelerator (800 keV, 20 kW) in the presence of gaseous ammonia. Figure 3 presents the dose dependence of SO_2 and NO_x removal efficiencies

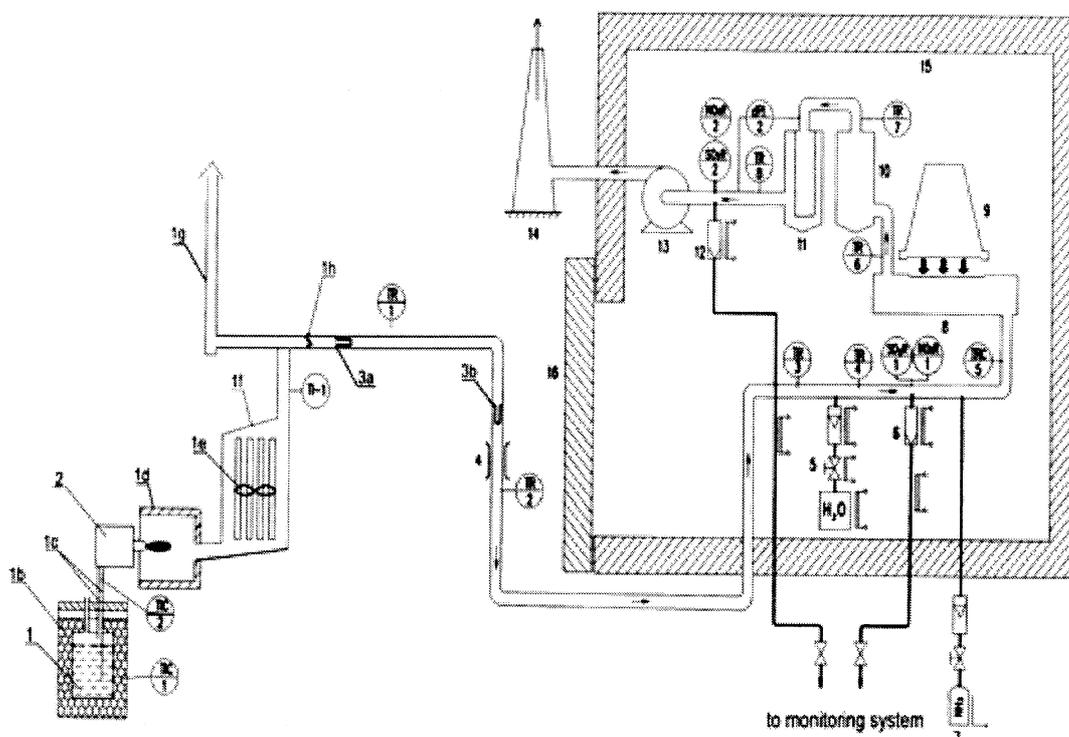


Fig. 2. Schematic diagram of laboratory EB installation. 1 – thermostated fuel oil; 2 – oil burner; 3 – particulate and soot filters; 4 – orifice; 5 – dosage of water vapor; 6 – gas sampling point-process inlet; 7 – ammonia injection; 8 – process vessel; 9 – electron beam accelerator; 10 – retention chamber; 11 – bag filter; 12 – gas sampling point-process outlet; 13 – induced-draught fan (ID fan); 14 – stack; 15 – concrete shielding wall; 16 – concrete shielding door.

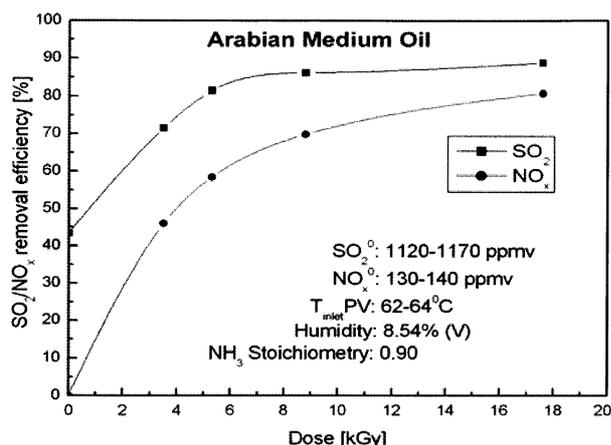


Fig. 3. Effect of irradiation dose on SO_2 and NO_x removal from flue gases formed during combustion of Arabian Medium Oil with 2.81 wt.% of sulfur.

from flue gases from combustion of Arabian Medium Oil with 2.81 wt.% of sulfur. High removal efficiencies up to 98% for SO_2 and 80% for NO_x were simultaneously obtained under optimal irradiation conditions. All these flue gases after adequate electron beam irradiation meet the stringent emission regulations for both pollutants. The by-product, which is a mixture of ammonium sulfate and nitrate, can be used for production of commercial agricultural fertilizers, like NPK (i.e. fertilizer containing nitrogen, phosphorous and potassium as nutrients) or NPKS (i.e. fertilizer containing nitrogen, phosphorous, potassium and sulfur as nutrients).

Combustion of high-sulfur lignite

In many countries large amounts of low-quality, high-sulfur coal is used in the power industry. The TPPs in the Maritsa-East region (Bulgaria) are supplied by low calorific with high sulfur content Thracian lignite. This is a low-grade Bulgarian lignite with the following quality parameters: ash content (as received base) up to 20 wt.%, moisture up to 52.5 wt.%, combustible sulfur up to 2.7 wt.%, carbon up to 20 wt.%, and low calorific value up to 6711 kJ/kg. The 215 MW unit of TPP Maritsa-East 2 consumes 350 ton/h of lignite. The pilot plant for EBFGT of the flue gas from lignite-fired boiler was constructed in the TPP Maritsa-East 2 [10]. Table 3 presents the main technological parameters of the plant. High simultaneous removal efficiency of SO_2 up to 99% and NO_x up to 90% was achieved at low doses

Table 3. The main technological parameters of the Maritsa-East 2 pilot plant

Parameter	Unit	Value
Flow rate of flue gas	Nm^3/h	10 000
Inlet gas temperature to PV	$^\circ\text{C}$	66
Ammonia stoichiometry		0.90
Inlet SO_2 concentration	mg/Nm^3	17 190
Inlet NO_x concentration	mg/Nm^3	217
EB parameters	800 keV, 35 kW \times 3	
Absorbed dose	kGy	3.94
Obtained removal efficiency		
SO_2	%	97.8
NO_x	%	86.3

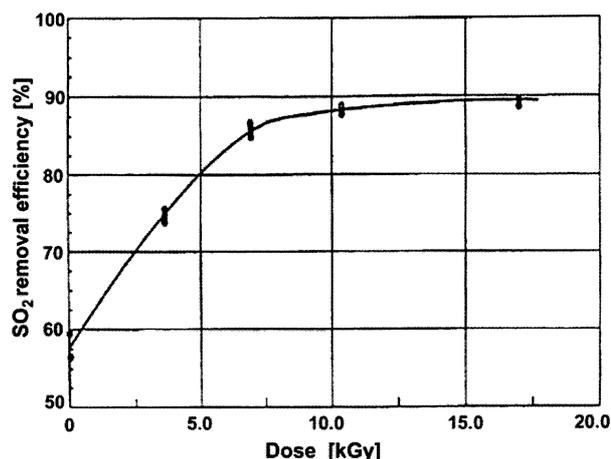


Fig. 4. Dose dependence of SO_2 removal from model gases with extremely high SO_2 (15 vol.%). The experimental conditions: gas humidity (14.5 vol.%), ammonia stoichiometry (0.90) and gas temperature (105–114°C).

4–6 kGy. The by-product collected at the electrostatic precipitator, is a mixture of ammonium sulfate and nitrate with a moisture content of about 0.8 wt.% can be used directly as fertilizer or as component for mixed fertilizers used in agriculture. The positive results of the plant operation were the basis for preparation of a feasibility study for an industrial plant in Bulgaria.

Purification of exhaust gases from copper smelter

Exhaust gases with high SO_2 concentration up to 15 vol.% are emitted from a copper smelter [21]. In conventional worldwide smelter practices, these gases with an SO_2 content higher than 4 vol.% are fed to sulfuric acid plants that provide SO_2 abatement. In the metallurgical industry, there is no consistent trend for the treatment of flue gas with low SO_2 content (smaller than 4 vol.%). The gases with a low SO_2 content are released from varied smelter sources such as: fugitive gases from different stages of the process, gases from secondary process (electric furnaces, etc.) and tail gas from sulfuric acid plants. The applicability of the EB-FGT for controlling copper smelter off-gases has been demonstrated at two plants in Poland: the EPS Kawęczyn pilot plant for flue gas with concentration of SO_2 up to 3200 ppmv (an additional SO_2 was dosed to the flue gases from a coal fired boiler) and the INCT laboratory installation with a model gas with an SO_2 content up to 15 vol.% SO_2 removal efficiency up to 90% was achieved (see Fig. 4) in the INCT

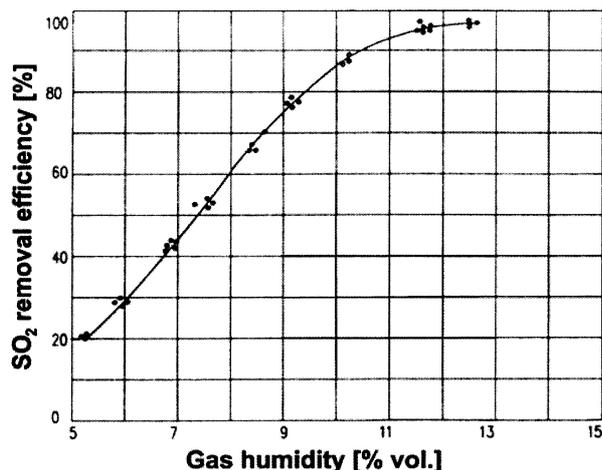


Fig. 5. Effect of gas humidity on SO₂ removal. The experimental conditions: inlet SO₂ concentration (1000–3200 ppm), dose (11.5 kGy), gas temperature (60–64°C), NH₃ stoichiometry (0.85–0.95).

laboratory installation at a dose of 17.5 kGy, when the temperature and humidity (see Fig. 5) of irradiated gases and ammonia stoichiometry were properly adjusted [15]. Both exhaust gases with a weak SO₂ content as well as with high SO₂ content can be desulfurized in the EBFGT process with high efficiency.

Removal of polycyclic aromatic hydrocarbons (PAHs)

PAHs are emitted from coal or oil-fired boiler. Some of them are carcinogenic, mutagenic or both. The United States Environmental Protection Agency (US EPA) has fixed 16 PAHs as priority pollutants. The studies of PAHs removal during simultaneous SO₂ and NO_x removal from flue gas using EB irradiation have been carried out at EBFGT pilot plant at EPS Kawęczyn using coal as fuel [5]. Concentration of 16 PAHs in the flue gas, emitted from a coal-fired WP-120 boiler, was measured at the plant inlet and outlet by gas chromatography-mass spectrometry (GC-MS) technique. During the plant operation with irradiation dose of 8 kGy, two tests were performed as follows:

- The first test was with a gaseous ammonia addition to the flue gas before its inlet to the process vessel (PV). The ammonia stoichiometry was 0.90. It was a typical condition for simultaneous SO₂ and NO_x removal in the EBFGT process.
- The second test was without ammonia addition.

The removal efficiencies of 5 PAHs obtained in both tests are presented in Fig. 6. The results of both tests can be concluded as follows:

- Removal efficiencies of 14 PAHs ranged from 2% for the four-ringed benzo(b+k)fluoranthene up to 96.5% for the two-ringed acenaphthene were obtained in typical conditions for SO₂ and NO_x removal in the EBFGT process,
- The concentrations of the 2 three-ringed compounds: anthracene and fluoranthene increased after EB irradiation. Probably they are the products of decomposition of higher-ringed PAHs. This phenomenon is now under investigation.
- Ammonia addition to the flue gas has a positive influence on the removal efficiencies of 16 PAHs.

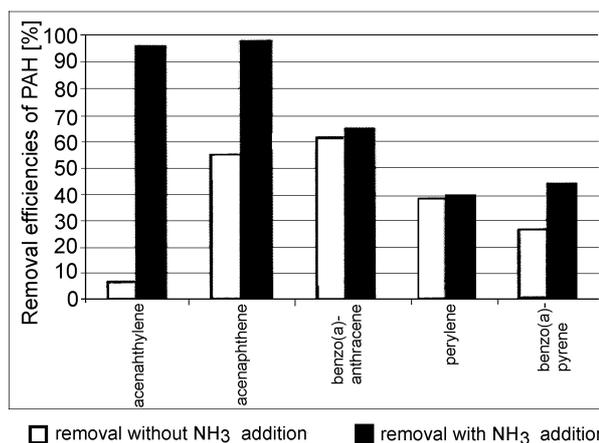


Fig. 6. Removal efficiencies of 5 PAHs obtained in the tests with and without ammonia addition to the irradiated flue gas. The experimental conditions: flue gas flow rate (10000 Nm³/h), dose (8 kGy), gas humidity (6.5 vol.%), ammonia stoichiometry (0.90), gas temperature at PV inlet (65°C). Simultaneous SO₂ removal (92%) and NO_x removal (75%) were obtained.

After EBFGT with an irradiation dose of 8 kGy, the total concentration of all 16 PAHs was lowered by 26.5%. The PAHs are characterized by their carcinogenic factors related to benzo(a)pyrene. Such factors are proposed by EPA in the USA [18]. The overall toxicity of flue gas was calculated as a sum of concentration and carcinogenicity factor product for each PAH measured in the flue gas. Such calculations indicate that after EBFGT with an irradiation dose of 8 kGy, the overall toxicity of flue gas was decreased by 47.5%. The concentration of organic pollutants in the solid by-product is several times lower in comparison to commercial ammonium sulfate being a by-product of caprolactam or coke production processes.

This test confirms that the EBFGT process ensures simultaneous removal of SO₂, NO_x and PAHs with high efficiencies from coal-fired flue gas. Therefore, EBFGT is the multicomponent air pollution control technology.

Reduction of dioxin emission from municipal waste incinerators

Dioxins are very toxic to human body due to their endocrine disrupting character, carcinogenicity and mutagenicity. Dioxins include polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and coplanar polychlorinated biphenyls (PCBs). Dioxins are emitted from the incineration (especially of municipal solid waste and medical waste), metallurgical processes (steel and iron, copper, aluminum and zinc metallurgy), manufacturing of paper and pulp and combustion of fuels with high content of chlorine. In addition, dioxins are typically formed during cooling of the off-gases. The EBFGT was applied to reduce the emission of dioxins in flue gas from the municipal solid waste incinerator at the Takahama Clean Center (Japan) [12]. More than 90% decomposition of dioxins was obtained after EB irradiation with a dose of 14 kGy. The treated flue gas meets the stringent emission standards for dioxins

established by the Government of Japan, the USA and the EU. Low-toxicity of irradiated gas was confirmed in terms of endocrine disrupter using enzyme immunoassay.

VOCs removal from exhaust gases

VOCs are released into the atmosphere from various industrial processes. Their main emission sources are: the mobile combustion sector, organic solvent application, petroleum industry, combustion of fossil fuels (industrial and non-industrial), waste treatment and disposal. Several types of VOCs are very harmful to human health and the environment. When exhaust gases containing VOCs mixed with nitrogen oxides are released into the atmosphere and irradiated by UV component of sunlight, a complex chain of reactions converts them into photochemical pollutants (ozone, aldehydes, hydrogen peroxide, peroxyacetyl nitrate, organic and inorganic acids and fine particles) [7]. Among these pollutants, ozone is considered to be the most important because it is produced in high concentrations and its wide range of effects may have on human health, plant growth and material corrosion. Ozone is formed in the lower layer of the troposphere. The hydrocarbon reactivity is expressed in terms of photochemical ozone creation potential (POCP), units calculated with respect to ethylene as a reference compound (POCP = 100) [8]. The alkenes and aromatic hydrocarbons represent classes of VOCs with high POCP. VOCs released to the atmosphere affect large areas during long-distance redistribution through atmospheric pathways. The reduction of VOCs emission is a subject of growing interest in the international conventions.

EBFGT technology is a promising method for treatment of VOCs at low concentrations. Many studies on EBFGT of individual VOC in air have been performed at laboratory installations in Japan, Germany, the USA, South Korea and Poland [13]. It is very important to clarify how much EB energy is required to destroy VOCs in the flue gas from oil-fired boilers. Such studies are carried out at the INCT laboratory plant, Poland.

Equipment and process engineering

The main components of the process are: spray cooler, ammonia injection system, process vessel with accelerators and electrostatic precipitator (ESP). The process control system is based on industrial computer monitoring system. Most of the used systems are based on conventional process engineering design principles. However, they were adopted, tested and optimized at the EPS Pomorzany installation in Poland. The construction of PV was developed by the INCT, Poland [3]. Most crucial for plant operation in harsh industrial conditions is engineering and reliability of high power accelerators and their shut downs were the main drawback of recently constructed industrial plants. A new high power machine (400 kW) installed and operated at a wastewater plant, Korea, [19] is a breakthrough in accelerator construction which can solve the problems observed in EBFGT installations.

Conclusions

The EBFGT technology is very competitive in comparison with the conventional ones and usually is applied as a combination of wet FGD + ammonia based SCR. The process is dry and the by-product is a good quality fertilizer. EBFGT allows simultaneous removal of SO₂, NO_x and VOCs, and this cannot be achieved in other technologies. The technology can be applied for air pollution control in the case of different fuels; coal, lignite, heavy fuel oil, municipal wastes and to treat other industrial off-gases, e.g. lean gases emitted from copper smelters. Further development of high power electron accelerators is a crucial factor for technology implementation on industrial scale.

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