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TRANSACTIONS OF THE INSTITUTE OF FLUID-FLOW MACHINERY

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INSTYTUTU MASZYN PRZEPŁYWOWYCH

107

Selected papers from the First Polish-Japanese Hakone Group Symposium on Nonthermal Plasma Processing of Water and Air, Sopot, Poland, May 29-31, 2000

WYDAWNICTWO IMP PAN

GDAŃSK 2000

THE TRANSACTIONS OF THE INSTITUTE OF FLUID-FLOW MACHINERY

exist for the publication of theoretical and experimental investigations of all aspects of the mechanics and thermodynamics of fluid-flow with special reference to fluid-flow machines

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poświęcone są publikacjom naukowym z zakresu teorii i badań doświadczalnych w dziedzinie mechaniki i termodynamiki przepływów, ze szczególnym uwzględnieniem problematyki maszyn przepływowych

Wydanie publikacji zostało dofinansowane przez PAN ze środków DOT uzyskanych z Komitetu Badań Naukowych

EDITORIAL OFFICE – REDAKCJA

Wydawnictwo Instytutu Maszyn Przepływowych
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ISSN 0079-3205

TRANSACTIONS OF THE INSTITUTE OF FLUID-FLOW MACHINERY

No. 107, 2000, 17-31

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Electron beam technology for multicomponent air pollution control – a review of Polish activities

The electron beam flue gas treatment technology has proved succesful in a pilot plant scale and came into industrial applications. Industrial installation is under operation at Chengdu Electric Power Station in China, another is under construction at EPS Nagoya in Japan. The Polish industrial installation is being examined for continuous operation. In this installation 270,000 Nm^3/h will be purified with the efficiencies of simultaneous removal of 70% of SO_2 and 80% of NO_x. In the paper Polish experiences in design and construction of EB plants are presented. Recent year brought new regulation and international obligation considering the emission of organic compounds, and hence there is a need to look for new, effective methods to control these pollutants. The results of preliminary experiments in a pilot plant for volatile organic compounds removal by electron beam (EB) are presented as well.

1. Introduction

The electron beam flue gas treatment technology was invented many years ago [1]. Research on the process has been conducted in Japan [2], USA [3], Germany [4] and Poland [5]. However, the recent findings, based on the experiments performed at a pilot plant at Electric Power Station Kawęczyn, led to developments which made the process mature just at the dawn of the XXI century [6]. The process is being implemented in the full industrial scale at Electric Power Station Pomorzany (Dolna Odra EPS Group) [7]. Other developments are reported in Japan. Following the Shin Nagoya's pilot plant experiments [8] and industrial demonstration plant built in China [9], another one is under construction in Japan [10]. There are remarkable differences in technological and design solutions applied in Japanese and Polish installations. Developments achieved at EPS Kawęczyn pilot plant [11] and INCT laboratory unit [12] are presented in the paper.

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2. Laboratory installation at INCT

A laboratory unit (Fig. 1) with a gas flow capacity up to 400 Nm^3/h has been constructed at INCT. The composition of flue gases from two natural gas fired boilers can be adjusted by a dosage of different gaseous pollutants (SO_2, NO_x, VOC). An irradiator, accelerator ILU-6, is used with electron beam energy in the range 600-1000 keV [13].

The system has been used for investigation of SO_2 removal from off-gases with the high SO_2 concentration (up to 15%). This range of SO_2 concentration is observed in the case of off-gases emitted in processes found in the copper industry [12].

Flue gas with high SO_2 content can be effectively cleaned by NH_3 and electron beam treatment, even in higher than optimal temperatures. Under such conditions removal efficiency of SO_2 in the range of 90-95% was obtained. The ammonia stechiometry, humidity and dose up to 7 kg·a have a significant influence on the SO_2 removal efficiency. The product formed in the process is the ammonium sulfate.

The described laboratory installation can be used for engineering development of new electron beam technology as well as for investigation of the influenced process physical and chemical parameters on the efficiency of removal of SO_2.

However this unit, and other units of this size used in different laboratories all over the world, can not be used for process upscaling. Dimensions of laboratory irradiation chambers do not allow to study the effect of electrons (with a limited range of penetration) interaction with separate fragments of flue gas streams flowing inside large ducts. Industrial pilot plants (size of 10,000 Nm^3/h and bigger) are necessary to be constructed to investigate such phenomena.

3. Pilot plant at electric Power Station Kawęczyn

The pilot plant with a capacity of 25,000 Nm^3/h was constructed at the Electric Power Station Kawęczyn. It was installed on a bypass of the main flue gas stream from the boiler WP-120. The scheme of the installation is given in Fig. 2. Two accelerators ELV-3a (50 kW, 700 keV) were installed in series on a reaction vessel. For the first time in the case of industrial pilot installation a two-stage gas irradiation cascade was applied [5, 11, 14, 15].

The flue gases are cooled in a spray cooler working under "dry-bottom" conditions and then irradiated in two stages in the reaction vessel, which is 7 m long cylinder and 1.6 m in diameter. A double titanium window system was employed. An "air curtain" was used to separate the secondary window from the corrosive flue gas. Gaseous ammonia is injected by nozzles upstream of flue gas irradiation. Aerosols formed in the process (ammonium sulfate and nitrate) were determined using the Andersen impactor and then captured in filters: particles size distribution is given in Fig. 3. Formed particles are small with a diameter less than 1

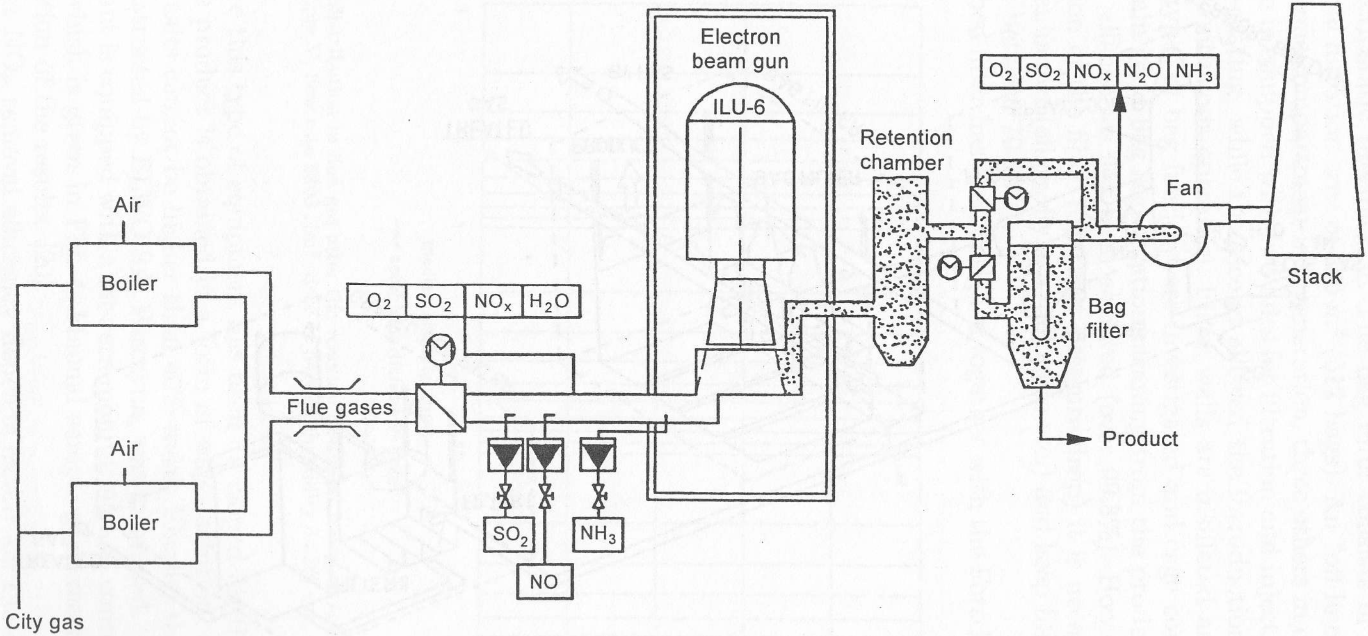

Fig. 1. Laboratory EB installation at INCT.

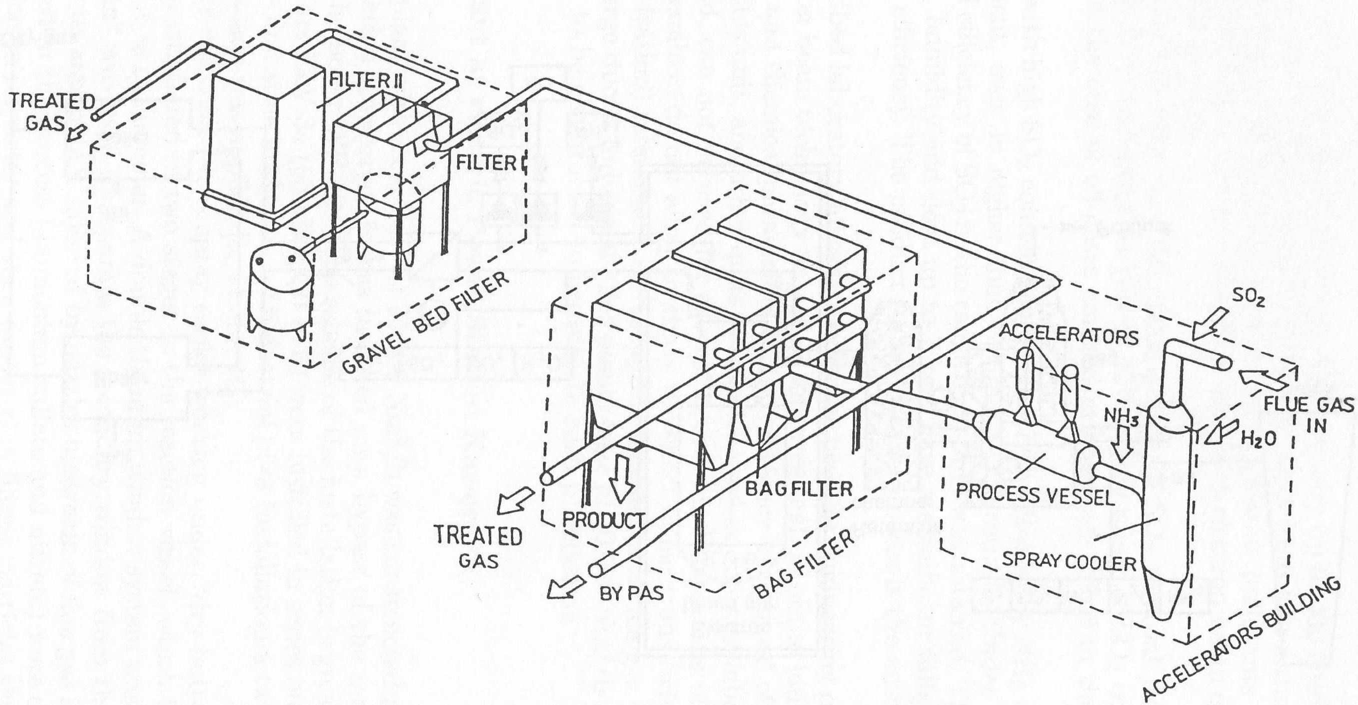

Fig. 2. EB pilot plant at EPS Kawęczyn.

micron and sticky. Three types of filters were applied – bag filter, gravel bed filter [16, 17] and electrostatic precipitator. The bag filter consists of four compartments with a total filtration area of 600 m^2 (512 bags). An "off line" regeneration mode was used (one compartment in regeneration, three others in operation) The bag filter system is equipped with two lines for filtration and injection. One could be used for pre-coating, while the second allowed the introduction of additional powder during continuous operation. Filter walls are insulated and bottom heated. Different types of bag fabrics were investigated and cage construction was modified to obtain good bag regeneration(cleaning from the product). Very high aerosol removal efficiencies has been achieved (over 99.8%). However, to obtain a stable operation of the filter (constant pressure drop) it is necessary to apply some filtration enhancer (silica, fly ash, dolomite etc.) and keep filter temperatures in the range between 70-80^{0}C.
The wet gravel bed filter has been designed together with the Forschungszentrum,

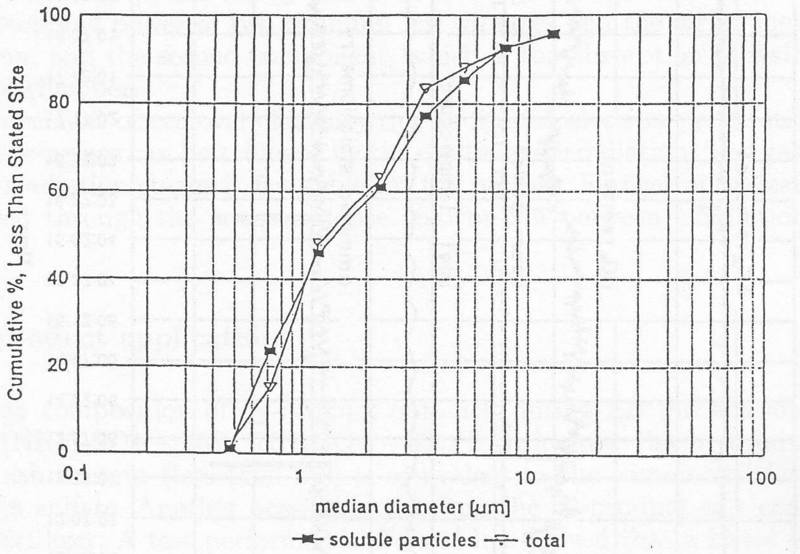

Fig. 3. Particle size distribution in flue gas after the reaction chamber. Initial SO_2 concentration 3000 ppm V, NO_x – 125 ppm V, flow rate 6800 Nm^3, dose 12.54 kg·a, humidity 6.52% vol., NH_3 stechiometry – 1.14.

Karlsruhe, where this type of equipment has been developed for the EB process. In this case, the product is obtained in a form of water solution. Concentration of the nitrogen salts cannot be higher than 40% mass. Finally, the electrostatic precipitator constructed by ELWO SA, Pszczyna, was examined.

The pilot plant is equipped with a modern monitoring and control system [18, 19], scheme of which is given in Fig. 4. Manual sample grab methods were also used for verification of the results, [20].

The fact that NO_x removal efficiency depends mostly on the dose and inlet NO_x concentration, SO_2 is mostly removed via thermal reaction, which renders

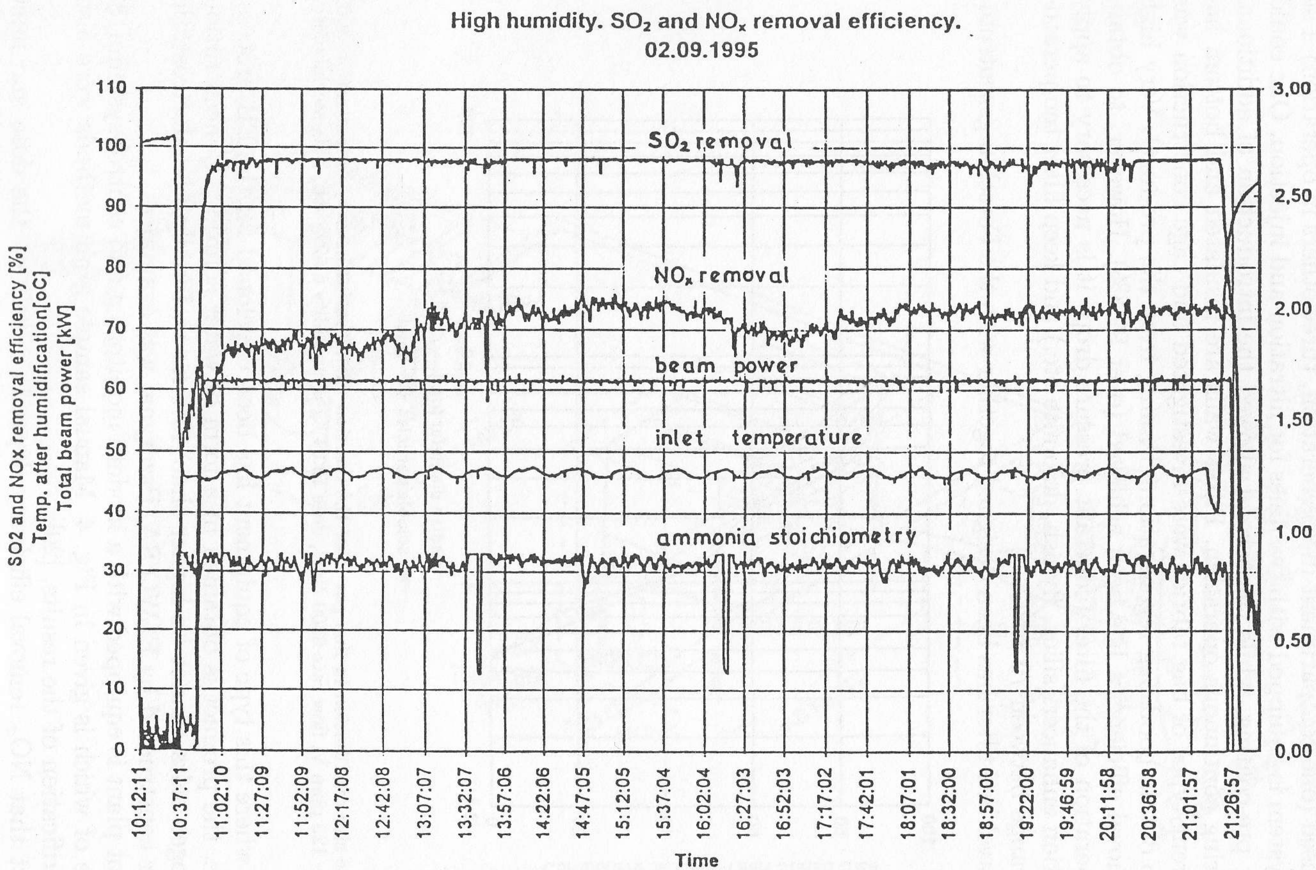

Fig. 4. Gas analyses system for electron-beam flue gas treatment process.

interesting observations concerning the ammonia consumption in both reactions.

The first set of experiments was performed for low gas humidity and rather high temperatures. In the second set of experiments the humidification system at the Kawęczyn pilot plant has been upgraded. High enthalpy water was used, packing was introduced into the column and water re-circulation was applied. The humidity of flue gases can be increased to 15% per volume and the gas inlet temperature reduced to 45^0C. At this condition the removal efficiency of SO_2 as high as 98% can be achieved (Fig. 5). Due to thermal reaction a very low dose is required to obtain a high removal efficiency (Fig. 6). In the case when moderate NO_x removal efficiency is required the economical competitiveness of EB process with conventional technologies increases even further.

The most important problem to be solved in the process up-scaling is the construction of irradiation vessel. Longitudinal, double irradiation of the flue gases has been applied in the Kawęczyn's pilot plant .

The losses in the beam energy delivered to the gas consist of two components: one, the stopping power of two titanium foil windows and the air in the gap between them, and the second component, which is the absorption in walls of the irradiation chamber.

The improvement of removal efficiency due to limited electron penetration through the flowing gas can be achieved by the multistage irradiation. The gas mixing between irradiation stages is favorable for the process. Further optimization can be achieved through the adequate dose distribution between irradiation stages [21].

4. By-product application

Average composition of by-product obtained during EB purification of flue gases is: $(NH_4)_2SO_4$ – 95%, NH_4NO_3 – 5%. In principle, the by-product with small fly ash content (less then 2%) is equivalent to the commercial fertilizer – ammonium sulfate. Another possibility is to use the by-product as a component in NPK fertilizer. A test performed in Poland has proved that a blend obtained with the application of the by-product meets the standards established for this kind of fertilizers.

Agricultural tests have been performed using the pure by-product and a by-product with different fly-ash content. Results were very positive. The content of heavy metals is much lower than the values allowed for commercial fertilizers.

5. Industrial project in Poland

The experience gathered during laboratory and pilot plant tests has allowed for the preparation of the full scale industrial plant design.

The electron dry scrubbing process will be employed at the Pomorzany Electric Power Station – Szczecin in Poland for the simultaneous removal of SO_2 and

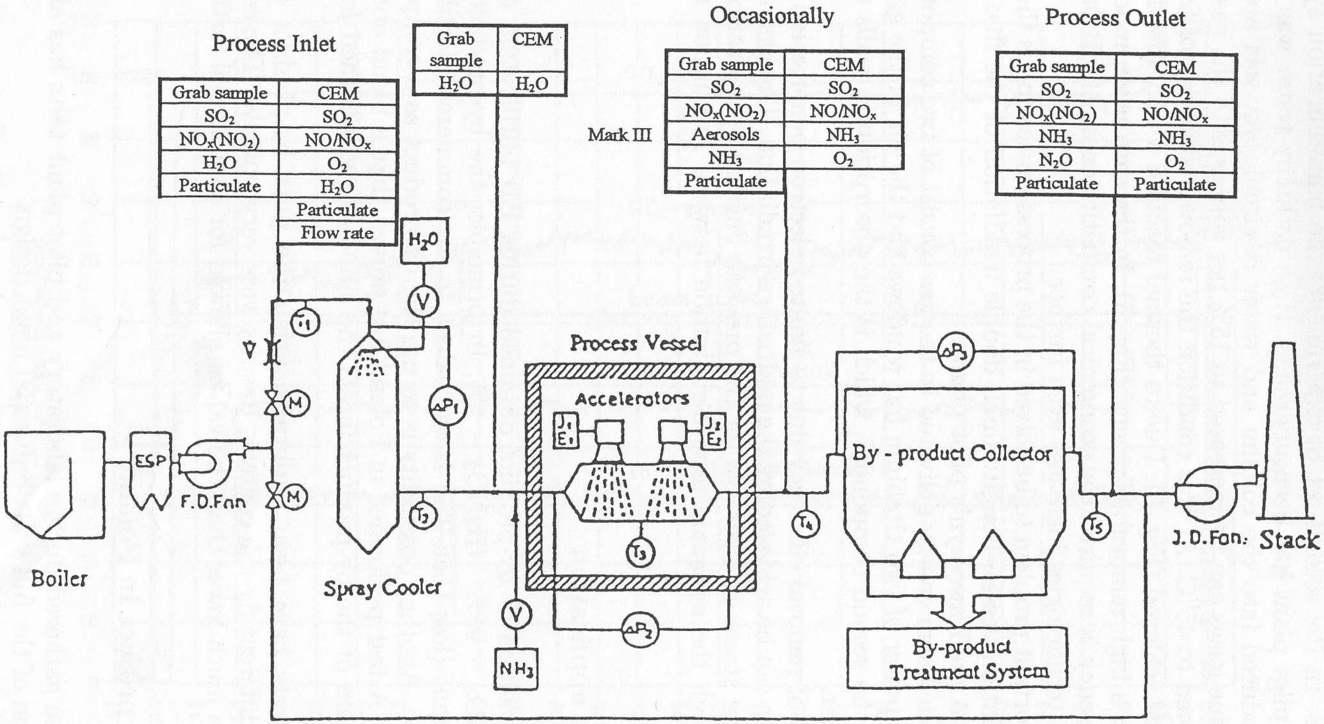

Fig. 5. SO_2 and NO_x removal in continuous operation.

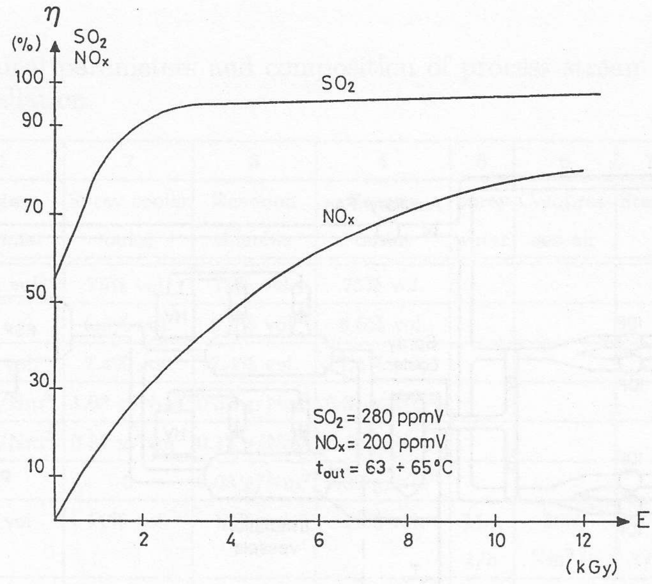

Fig. 6. Removal efficiency of SO_2 and NO_x vs. dose.

NO_x from flue gases emitted by Benson boilers [22].

After a detailed analysis it was decided to design and construct the electron-beam installation treating a maximum flow of 270,000 Nm^3/h of flue gas.

High concentration of NO_x and relatively low content of SO_2 in flue gas emitted from Benson boilers establish the specific conditions for flue gas treatment. The parameters of EB process are chosen so as to guarantee efficiency of removal up to 80% of NO_x and up 70% of SO_2 in continuous operation of the installation. For intermittent operation up to 80% removal of NO_x and 90% reduction of SO_2 are expected. The conceptual system arrangement of this situation is shown in Fig. 7.

The main technical parameters and composition of flue gases in the Pomorzany EB installation are shown in Table 1.

The flue gas will be humidified up to 10% by volume in a dry bottom spray cooler. Ammonia is then injected before two parallel irradiation vessels equipped with two 300 kW, 800 keV accelerators installed in series (Kawęczyn pilot plant's solution). Longitudinal gas irradiation will be applied. Formed aerosol will be collected in a dry electrostatic precipitator ESP with a flat heated bottom furnished with a scraping device. The manufacturer guaranteed a high removal efficiency (particulate at the outlet less than 20 mg/Nm^3) and corrosion resistivity of the equipment. The product will then be granulated for final distribution.

The ammonia consumption is estimated to be 180-200 kg/h and the by-product yield to be ca. 800 kg/h. Ammonia water will be used, as the component for preparation of gaseous ammonia.

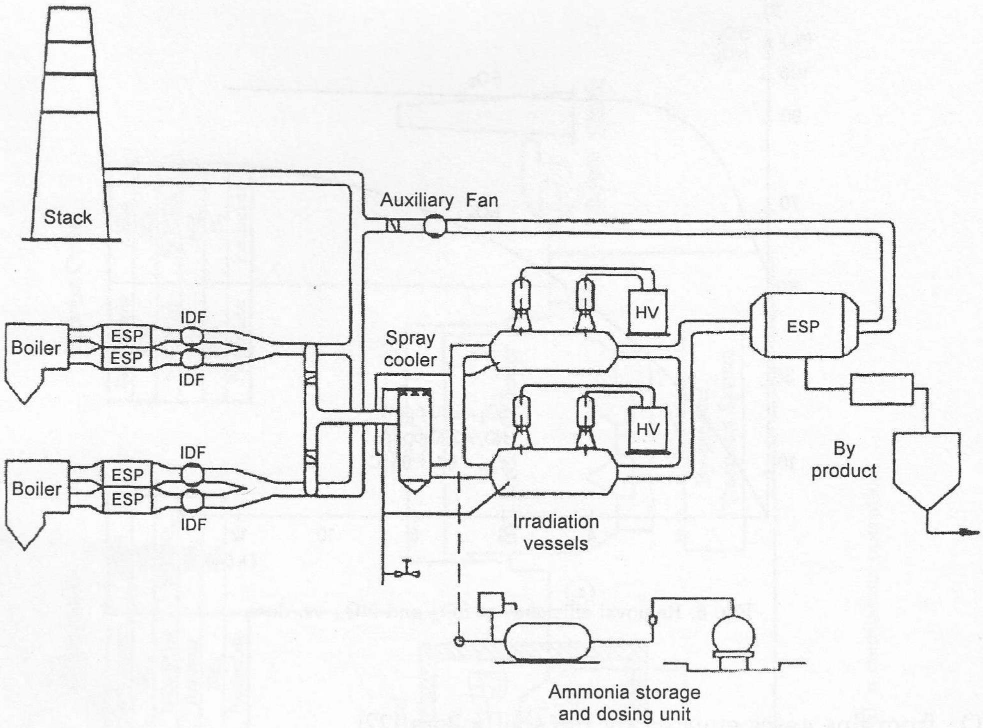

Fig. 7. Process flow diagram of EB installation at the EPS Pomorzany, Szczecin.

An EB industrial installation of similar capacity to the EPS Pomorzany has been designed and is now installed by Ebara Corporation at Chendu Electric Power (Sichuan Electric Power Administration) in China [9], primarily for the removal of SO_2. Design condition of this installation are following:

volumetric flow of flue gases approx.	300,000 Nm^3/h
SO_2 content in flue gas	1,800 ppm
efficiency removal of SO_2	80%
by product output rate	2,470 kg/h

The Chinese EB installation has been in operation since May 1998.

6. Volatile organic compounds (VOCs) removal

Combustion of fossil fuel is also one of the largest source of volatile organic compounds emission. Emitted VOCs belong to the aliphatic hydrocarbons, aldehydes, chlorinated hydrocarbons, aromatic hydrocarbons and the most dangerous polyaromatic hydrocarbons (PAHs) groups. Many of them are involved in the formation of photochemical smog as well as depletion of the stratospheric

Table 1. Technical parameters and composition of process stream at key points of the EB installation

Stream No.	1 System gas inlet	2 Spray cooler oulet	3 Reaction chamber	4 Process outlet	5 Spray water	6 Compres-sed air	7 Steam	8 NH_3	9 By-pro-duct
N_2	80% vol.	75% vol.	75% vol.	75% vol.					
O_2	7% vol.	6.6% vol.	6.6% vol.	6.6% vol.					
CO_2	8% vol.	7.4% vol.	7.4% vol.	7.4% vol.					
SO_2	1.1 g/Nm^3	1.03 g/Nm^3	0.31 g/Nm^3	0.31 g/Nm^3					
NO_x	0.6 g/Nm^3	0.57 g/Nm^3	0.12 g/Nm^3	0.12 g/Nm^3					
NH_3	0	0	0.03 g/Nm^3	0.03 g/Nm^3					
H_2O	5% vol.	11% vol.	11% vol.	11% vol.	11.1 t/h	3800 Nm^3/h	4.5 t/h	180 kg/h	695 kg/h
$(NH_4)_2SO_4$	0	0	415 kg/h	0.012 g/Nm^3					
NH_4NO_3	0	0	262 kg/h	0.007 g/Nm^3					
Solid particles	0.08 g/Nm^3	0.06 g/Nm^3	2.45 g/Nm^3	0.02 g/Nm^3					
Flue gas	270000 Nm^3/h	284000 Nm^3/h	284000 Nm^3/h	284000 Nm^3/h					

ozone. In addition some of them are toxic and carcinogenic. These compounds are emitted from the combustion process with various concentrations, ranging from few ppm up to hundreds ppm, the volume of emission depends simultaneously on the process conditions and kind of used fuels, [23]. The most popular method used in industry to control VOCs in flue gases is adsorption but in the case of combustion gases, the presence of other pollutants such as SO_2, NO_x and dust causes this process to be ineffective. Otherwise, the secondary pollution – used adsorbent is generated. In the present state of art, it seems that electron beam treatment is a single promising method to remove SO_2, NO_x and VOCs from flue gases simultaneously.

Decomposition of many VOCs by electron beam treatment has been investigated [24, 25], numerous laboratory experiments, especially for chlorinated aliphatic hydrocarbons, have been carried out [26]. Investigated compounds have been irradiated by electron beam with the dose ranging from 2 to 15 kg·a, products of radiolysis have been detected by gas chromatography/flame ionization (GC/FID) and gas chromatography/mass spectrometry (GC/MS) techniques. The major products of irradiated organic compounds are H_2O, CO_2, CO and HCl, Cl for chlorinated hydrocarbons, and the efficiencies of decomposition have ranged from 60% up to 98%.

Irradiation of the air containing vaporized water causes production of the activated species such as: HO_2, O, N, H and OH radicals. Mechanism of organic compounds decomposition is based on the oxidation process, OH radicals are proposed, [27] as the most effective oxidizing species, direct electron impact processes and negative ion dissociative attachments may contribute the process as well [28].

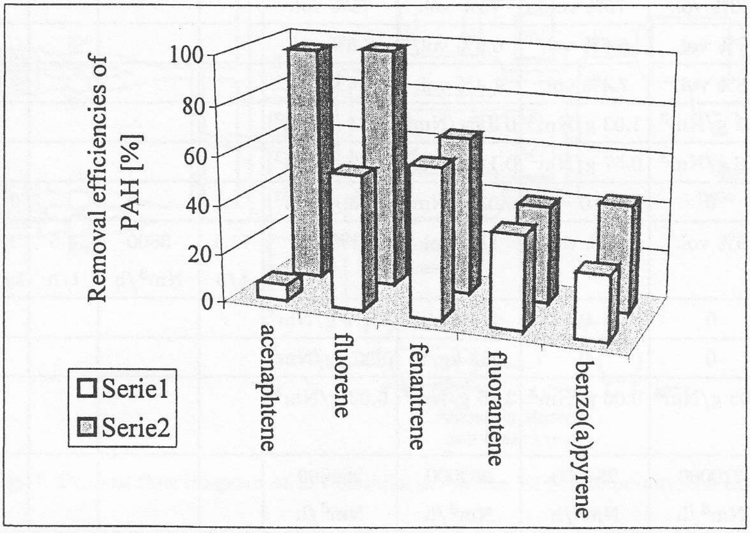

Fig. 8. Removal efficiencies of PAHs from flue gas.

The pilot plant tests for the influence of electron beam on the chosen polyaromatic hydrocarbons during simultaneous SO_2 and NO_x removal has been carried out at EPS Kawęczyn, equipped with 2 electron accelerators 50 kW/700 keV each. Concentration of 5 polyaromatic hydrocarbons have been measured before and behind the irradiation chamber. After filtration, the sample gas have passed through the adsorbents (activated coal and XAD – 2 resin). PAHs trapped on the adsorbent material have been extracted by solvents and analysed by GC/MS technique. The experimental work have been carried out in two series; irradiation dose in both was 7 kg·a, in the second series ammonia was injected into combustion gases. The purpose of carried out experiment with ammonia additive was to investigate the influence of EB on the PAHs during SO_2 and NO_x removal; in this process NH3 is a reagent. Removal efficiencies obtained in the pilot plant test are presented in Fig. 8, in the case of ammonia presence higher removal efficiencies for all PAHs are observed. Probably when ammonia is added, sulphur and nitrogen acids are neutralized and reaction with OH radical is promoted, or some products of VOCs with NH_3 are created. There is a necessity in the future work to carry out laboratory experiment of irradiation of single compounds with and without

NH_3 additive to recognize which kind of effect NH_3 causes. In a pilot plant test it is possible to check only the global influence of EB on the organic compounds stream. Simultaneously high removal efficiencies of SO_2 (89%) and NO_x (72%) were obtained.

7. Conclusions

The tests performed in the laboratory installations and the pilot plant resulted in the preparation of the industrial plant design. All the technical and economical analyses prove the advantages of this technology over the present day conventional technologies. The pilot plant test for polyaromatic hydrocarbons suggests that the electron beam treatment is also a promising technology for emission of organic compounds.

Received 15 August 2000

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Technologia wykorzystująca wiązkę elektronów do oczyszczania gazów – przegląd dotyczący Polski

Streszczenie

Technologia wykorzystująca wiązkę elektronów została sprawdzona w skali pilotowej i znalazła zastosowanie w skali przemysłowej. Instalacja przemysłowa tego typu pracuje w Elektrowni Chengdu w Chinach, kolejna jest budowana w EC w Nagoi (Japonia). Pierwsza polska instalacja tego typu jest testowana przed rozpoczęciem pracy ciągłej. Podczas pracy instalacji 270.000 Nm^3/h gazu będzie oczyszczane z efektywnością usuwania 70% SO_2 i 80% NO_x jednocześnie. W publikacji omówiono polskie doświadczenia w projektowaniu i konstruowaniu instalacji. Ostatnie lata przyniosły nowe regulacje dotyczące emisji lotnych związków organicznych, w związku z tym istnieje potrzeba poszukiwania nowych, efektywnych metod usuwania również tych związków. Przedstawiono również wyniki wstępnych badań napromienia wiązką elektronów lotnych związków organicznych w skali pilotowej.