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ELECTRON BEAM FLUE GAS TREATMENT PROCESS. REVIEW

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Electron beam flue gas treatment process. Review

The basis of the process for electron beam flue gas treatment are presented in the report. In tabular form the history of the research is reviewed. Main dependences of SO_2 and NO_x removal efficiences on different physico-chemical parameters are discussed. Trends concerning industrial process implementation are presented in the paper, finally.

Oczyszczanie gazów odlotowych z wykorzystaniem wiązki elektronów. Przegląd metod

Raport omawia podstawy procesu oczyszczania gazów odlotowych przy użyciu wiązki elektronów. W formie tabelarycznej zestawiono główne, niekiedy już historyczne, prace prowadzone na instalacjach laboratoryjnych i pilotowych. Prezentowane są zależności różnych fizykochemicznych parametrów na wydajność usuwania SO_2 i NO_x. Przedstawiono trendy związane z zastosowaniem procesu w pełnej skali przemysłowej.

CONTENTS

1. INTRODUCTION *7*

2. HISTORICAL REVIEW *7*

3. CHEMISTRY OF THE PROCESS *11*

   3.1. INFLUENCE OF ELECTRONS *11*

   3.2. OXIDATION OF NO_X AND FORMATION OF NITRIC ACID *12*

   3.3. OXIDATION OF SO_2 AND FORMATION OF SULPHUR ACID *14*

   3.4. INFLUENCE OF AMMONIA ADDITION *14*

4. CURRENT STATUS OF THE E-BEAM FGT PROCESS *17*

5. FUTURE TRENDS *18*

6. CONCLUSIONS *20*

REFERENCES *20*

1 INTRODUCTION

A great deal of worlds electricity production is based on fossil fuels; mainly on combustion of oil and coal. This produce a large amount of different kind of emissions which are harmfull for environment. Amount of these effluents have been researched in many local and global inventories (*e.g.* [1, 14, 18, 38, 69, 31]). In the developed countries the total emissions of SO_2 decreased about a quater and the total emissions of NO_X increased few percents from 1980 to 1990 [31]. In fast developing areas at Asia (ex. China and India) both SO_2 and NO_X emissions continued their strong increas [1].

Several different national and international institutions have ongoing efforts to project future energy use. Althought the scenarios incorporate slightly different assumptions they predict a substantial increase in energy use over the next 20 to 30 year. In World Energy Councils reference case demand rises more than 50% by 2020. The International Energy Agency and the U.S. Department of Energy (DOE) projections show a 34 to 44% rise in demand by 2010. Much of the growth of demand will occure in Asia and Latin America, where the electricity is produced mainly in the coal power plants [31].

The fossil fuel sources of both NO_X and SO_2 constitute the largest contribution to their atmospheric budgets [14]. The main environmental effect of raised atmospheric SO_2 concentration is acidification of soils and waters. The effects of NO_X are severe. NO_X affect not only acidification but also eutrophication and photochemical production of regional ozone and free tropospheric ozone [30]. Nitrogen oxides are the most important sources of tropospheric nitric acid. Kulmala *et al.* have studied that the presence of acid vapor can cause considerable changes to the radiative properties of low clouds [36]. Acidification of atmosphere and environment and increased content of aerosol make also health problems.

Public health and acid rain control consideration have led to worldwide action for reducing SO_2 and NO_X emissions from coal-fired boilers and for use of flue gas cleaning. There is a global need for good simultaneous SO_2 and NO_X removal methods. In this review we present one of the most promising methods - electron beam flue gas treatment, shown in Fig. 1 and Fig. 2. In the next section we will give a short historical survey. Then we explain the chemistry of process. In the sections four and five we discuss the current and the future status of method.

2 HISTORICAL REVIEW

The history of electron beam flue gas treatment begins in Japan in 1970 and 1971. Then Ebara Corporation initiated the studies for using radiation to prevent pollution. The first batch test studies (see Fig. 3) defined the radiation chemical reactions of SO_2 and NO_X originating from the irradiation of flue gases. The results were successful and development of the process continued with a joint effort by Ebara and Japan Atomic Energy Research Institute (JAERI) in 1972. At these tests it was noticed for example that [32, 25]

- Both SO_2 and NO_X can be extracted simultaneously when the flue gases are irradiated with an electron beam.
- Uninterrupted dry treatment of flue gases is realizable by irradiating the gas for a few seconds.

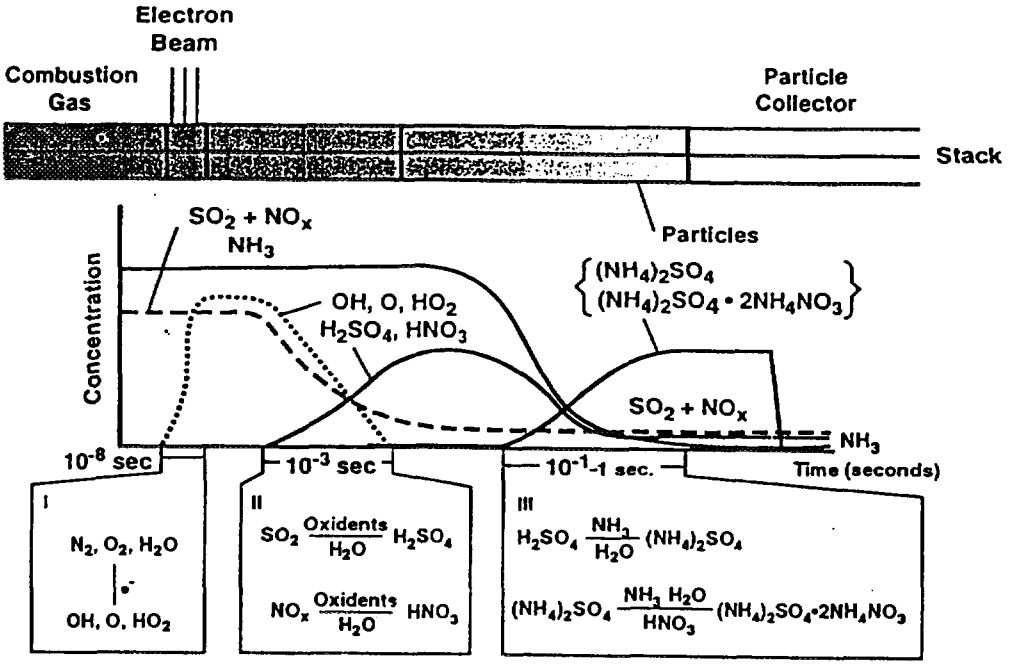


Figure 1: Schematic figure of physics and chemistry of electron beam dry-scrubbing process.

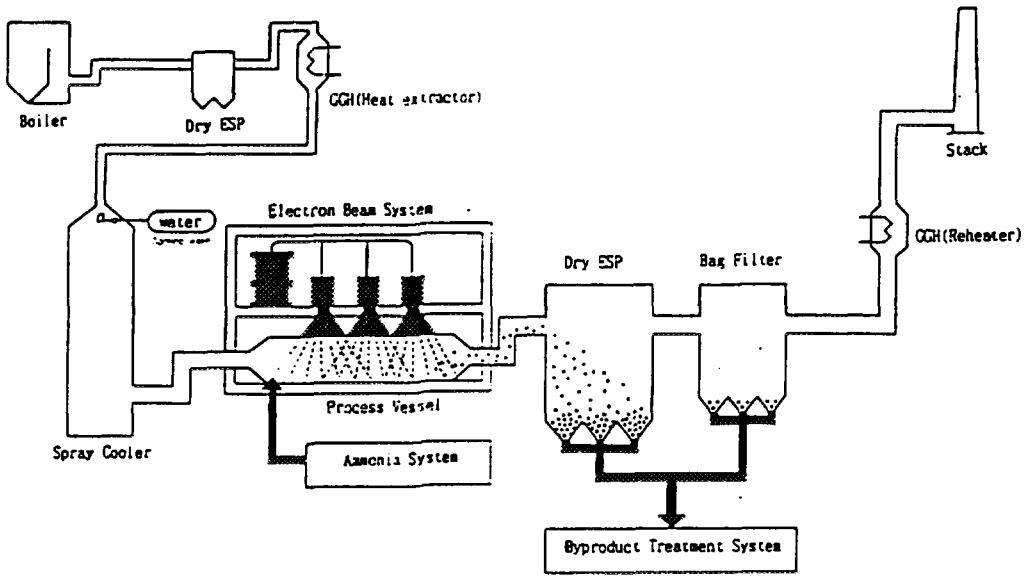


Figure 2: Process flow diagram.

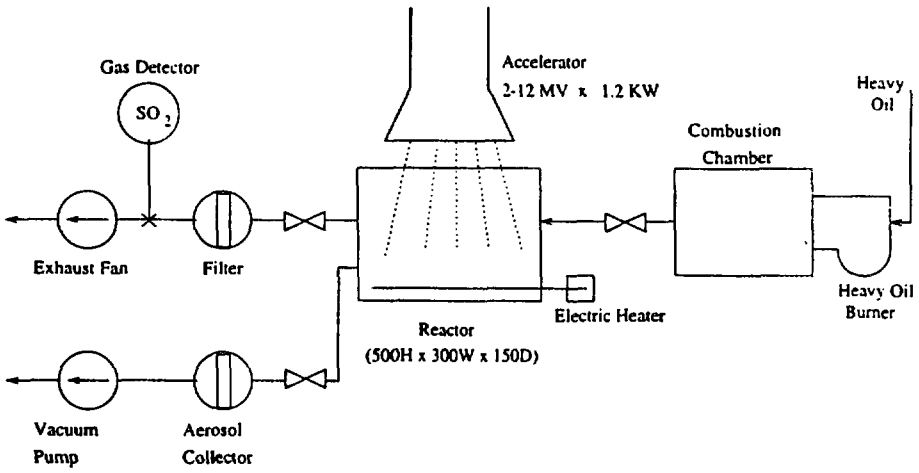


Figure 3: Schematic view of batch test in Japan in 1970-71 [25].

- The aerosol particles yielded with irradiation can be accumulated by an electrostatic precipitator mounted downstream of the reactor.

After this joint effort Ebara build a pilot plant in 1972. The purpose was to further study the process and to obtain basic data for commercialization of the process for treating flue gases with electron beam irradiation. This was the first test of the process using ammonia injection. The by-products were a mixture of ammonium sulfate and ammonium nitrate-sulfate, and were easier to capture than the aerosol without ammonia [40, 25].

Based on good scores of the earlier pilot plant in 1977 the Research Association for Abatement and Removal of NO_X in Steel Industry constructed a 10 000 Nm^3/h flue gas treatment plant to remove SO_2 and NO_X from the exhaust gas of a steel sintering plant at Yawata Works of the Nippon Steel Corporation in join research with Ebara. The pilot plant was operated for one year. During one month continuous operation the removal rates of 95% for SO_2 and 80% for NO_X was achieved [35, 40].

During the first decade of research of e-beam flue gas treatment process basic research was concentrated mainly in Japan. In 1979 started an early DOE-sponsored research done by Research Cotrell. Project continued through 1985. Research Cotrell adjusted technical and economical potentiality of e-beam process to clean SO_2 and NO_X from a typical electric utility flue gases. In 1984 Research Cotrell built a pilot plant where they tested the effects of an alkali-slurry spray of hydrated lime to neutralize the formed sulfuric and nitric acids (see sections 3.2 and 3.3). The end products were calcium sulfite, calcium sulfate and calcium nitrate and removal rates were grater than 90% and 80% for SO_2 and NO_X, respectively [25].

In 1983 Ebara International Corporation (later Ebara Environmental Corporation) started to built a process development unit at the Indianapolis Power and Light Company's E.W. Stout plant in Indianapolis, Indiana. The purpose of these tests were to evaluate e-beam flue gas treatment in a coal fired utility plant. The results of this project are represented in the detailed report done for DOE [16] and in articles [20, 21, 22].

In Europe e-beam flue gas treatment was first investigated in Germany where three installations were built in 1984. In the Nuclear Research Center in Karlsruhe (KfK) and in the University of Karlsruhe the study concentrated mainly to the reaction mechanism of the process and to improving the process [58, 56, 33, 57, 28, 47, 73, 70, 71]. In the 20 000 Nm^3/h pilot plant of Badenwerk AG RDK-7 plant the testing mainly addressed NO_X

Table 1: Research places of e-beam FGT process during the years [23, 55, 43, 37, 61, 19, 17, 63].

Year	Institution		Flow	Accelerator	Type of flue gas
	name	country	[Nm^3/h]	[kW]/MeV	
1970-71	Ebara	Japan	0.02	1.2/2-12	simulated
1972-74	JAERI	Japan	60	15/1.5	simulated
1974-77	Ebara	Japan	1 000	30/0.750	heavy oil
1974-78	University of Tokyo	Japan	84	0.12/1.0	simulated
1977-78	Ebara	Japan	10 000	90/0.750	sinter plant
1984-85	Research Cortell	USA	5300	80/0.800	coal
1981-91	JAERI	Japan	0.9	/1.5	
1984-88	Ebara	USA	24 000	160/0.800	coal
1984-91	KfK	Germany	1 000	3.6/0.300	heavy oil
1984-91	University Karlsruhe	Germany	1 000	22/0.220	nat. gas
1984-	SINR[a] Academia	China	25	/0.8	simulated
1985-89	Badenwerk	Germany	20 000	180/0.300	coal
1986-90	University of Helsinki	Finland	50	1.5/0.150	simulated & heavy oil
1988-1992	Energostal NPI[b]	Ukraine Russia		45/1.5	coal and
1989	KfK	Germany	1 500	16.5/0.550	light oil
1991	INCT	Poland	20 000	100/0.700	coal
1992	NKK/JAERI	Japan	1 000	50/<1.000	incinerator
1992	Ebara	Japan	50 000	80/0.800	tunnel
1992	Ebara/JAERI	Japan	12 000	108/0.800	coal
1994-	IPEN-CNEN[c]	Brazil	1 200	37.5/1.5	simulated
1997	Ebara	China	300 000	600/0.800	coal
1998	INCT (Pomorzany)	Poland	270 000	1200/0.800	coal
2000	Research Cotrell & VAC[d]	Estonia	(boiler 100MW_e)	800/1.25	oil shale rock

(a) SINR=Shanghai Institute of Nuclear Research

(b) NPI=Nuclear Physics Institute, Siperian Department of Russian Academy of Sciences

(c) IPEN-CNEN=Instituto de Pesquisas Energéticas e Nucleares

(d) VAC=Virginia Accelerators Corporation

removal [26, 27, 2, 60, 65, 64]. Later in 1989 in KfK a second pilot plant was added to continue work on mechanisms, aerosol formation and filtration [55, 25].

In Finland was a project from 1985 to 1990 to study the feasibility of irradiation for flue gas treatment and the aerosol formation of sulphuric acid [62, 42, 41, 43].

In 1989 the Institute of Nuclear Chemistry and Technology (INCT) in Warsaw, Poland built a laboratory test unit which was used to supply information to build a demonstration facility at the Electric Power Station Kawęczyn. In the INCT the research has centered to optimization of energy consumption, to by-product filtration and to building of efficient accelerators [10, 9, 7, 11, 6, 13].

During the beginning of this decade the e-beam process has got a great interest. Ebara built a test facility in 1991 [25]. In 1992 a pilot plant was started for treating incinerator gases from the Matsudo City [15, 54, 66]. Also in 1992 was started a 12 000 Nm^3/h coal burning facility to demonstrate the zone irradiation (see section 3.2) [3, 66] and a 50 000 Nm^3/h plant to treat ventilation gases from automobile tunnels [25, 66].

In Table 1 is a summary of institutions which have been researching and using electron beam flue gas treatment.

3 CHEMISTRY OF THE PROCESS

3.1 Influence of electrons

Electron beam initiate neutral chemistry by depositing energy into the flue gas. The produced active species consist of a combination of ions, atoms, radicals and excited atoms and molecules. Because the acceleration energy of electrons is typically 300-800 keV there is practically no close electron-nuclei interaction. Electrons transfer their energy to the electron shells of the gas molecules by numerous successive inelastic collisions and they become scattered throughout the whole reaction chamber in which the gas flows. The incident electron energy is distributed in the flue gas components statistically; proportionally to their mass fraction. So main flue gas components (N_2, O_2, H_2O and CO_2) absorb more than 99% of the incident energy. The primary processes can be schematically represented by:

$$4.43\,N_2 \overset{100eV}{\sim\!\sim} 0.29N_2^* + 0.885\,N(^2D) + 0.295\,N(^2P) + 1.87\,N(^4S) + 2.27\,N_2^+ + 0.69\,N^+ + 2.96\,e^- \quad (1)$$

$$5.377\,O_2 \overset{100eV}{\sim\!\sim} 0.077\,O_2^* + 2.25\,O(^1D) + 2.8\,O(^3P) + 0.18\,O^* + 2.07\,O_2^+ + 1.23\,O^+ + 3.3\,e^- \quad (2)$$

$$7.33\,H_2O \overset{100eV}{\sim\!\sim} 0.51\,H_2 + 0.46\,O(^3P) + 4.25\,OH + 4.15\,H + 1.99\,H_2O^+ + 0.01\,H_2^+ + 0.57\,OH^+ + 0.67\,H^+ + 0.06\,O^+ + 3.3\,e^- \quad (3)$$

$$7.54\,CO_2 \overset{100eV}{\sim\!\sim} 4.72\,CO + 5.16\,0(^3P) + 2.24\,CO_2^+ + 0.51\,CO^+ + 0.07\,C^+ + 0.21\,O^+ + 3.03\,e^- \quad (4)$$

The equations come from G-values[1] reported by Willis and Boyd [72]. N_2^* and O_2^* represent the sum of all long-lived excited state molecules and O^* denotes a highly excited O atom above the $O(^1S)$ level.

[1]The overall gain of excited state molecules, direct dissociation into neutral radicals and dissociation into ion pairs is described by G-values [72]

The traces of these primary radiolytic reactions get lost quickly. Fast charge transfer reactions redistribute the absorbed energy and actuate to the relaxation of the system toward the stable final state. Many charge transfer reaction lead to a partial dissociation of the reactants and generate neutral radicals thereby. For example, the main sources of the OH radical are the ion-molecule reactions:

$$O_2^+(H_2O) + H_2O \rightarrow H_3O^+ + OH + O_2 \quad (5)$$
$$O_2^+(H_2O) + H_2O \rightarrow H_3O^+(OH) + O_2 \quad (6)$$
$$H_3O^+(OH) + H_2O \rightarrow H_3O^+ + OH + H_2O \quad (7)$$

According Mätzing about 90% of the OH results from positive ion-molecule reactions, while only 10% originates from direct radiolytic decomposition of water vapour [45].

Absorbing of energy and ion-molecule reactions are participated mostly by the major components of flue gases. Instead radicals react with the trace components to be removed from the flue gase. The OH radical reacts essentially with all trace components and is responsible for most of the simultaneous oxidation of NO_X and SO_2 to nitric and sulfuric acid [45, 46].

3.2 Oxidation of NO_X and formation of nitric acid

In the chemical models of electron beam flue gas treatment the oxidation of nitrogen oxides (NO and NO_2) to nitric acid is not a simple straightforward process. Oxidation has to compete with backward reactions in which amount of partially oxidized intermediates reduce [59]. The main reaction of nitrogen oxide (NO) oxidation are:

$$NO + HO_2 \rightarrow NO_2 + OH \quad (8)$$
$$NO + O_3 \rightarrow NO_2 + O_2 \quad (9)$$

(Ozone is formed from the termolecular conjunction of O_2 and O [44]). NO_2 reacts considerably with O atoms and regenerate NO:

$$NO_2 + O \rightarrow NO + O_2 \quad (10)$$

This means that the intermediata NO_X is trapped in the reaction cycle of oxidation and reduction until it arrives at the product channel to form nitric acid with reaction

$$NO_2 + OH + M \rightarrow HNO_3 + M \quad (11)$$

or molecular nitrogen with reactions

$$N + NO \rightarrow N_2 + O \quad (12)$$
$$NO_2 + N \rightarrow O_2 + N_2 \quad (13)$$

or nitrous oxide with reaction

$$NO_2 + N \rightarrow N_2O + O \quad (14)$$

In the reaction equation (11) M is "a what-ever-molecule" (ex. H_2O, N_2, O_2). In addition to reactions (13) and (14) N atom can reduce N_2O with reaction

$$NO_2 + N \rightarrow 2\,NO \quad (15)$$

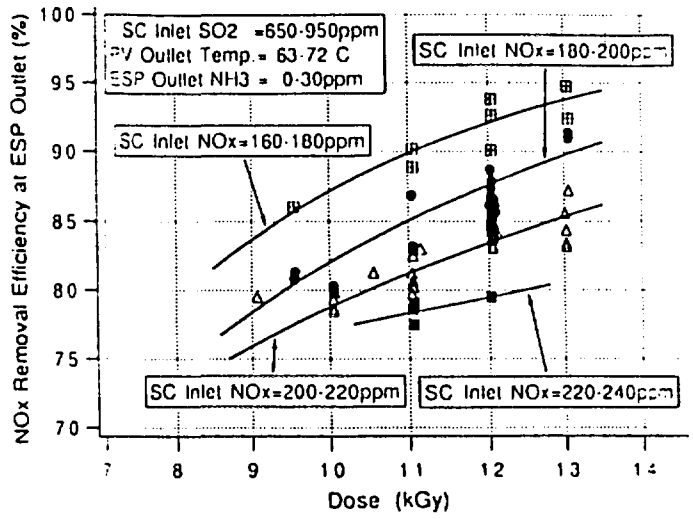


Figure 4: Removal of NO_X as a function of irradiation dose [51].

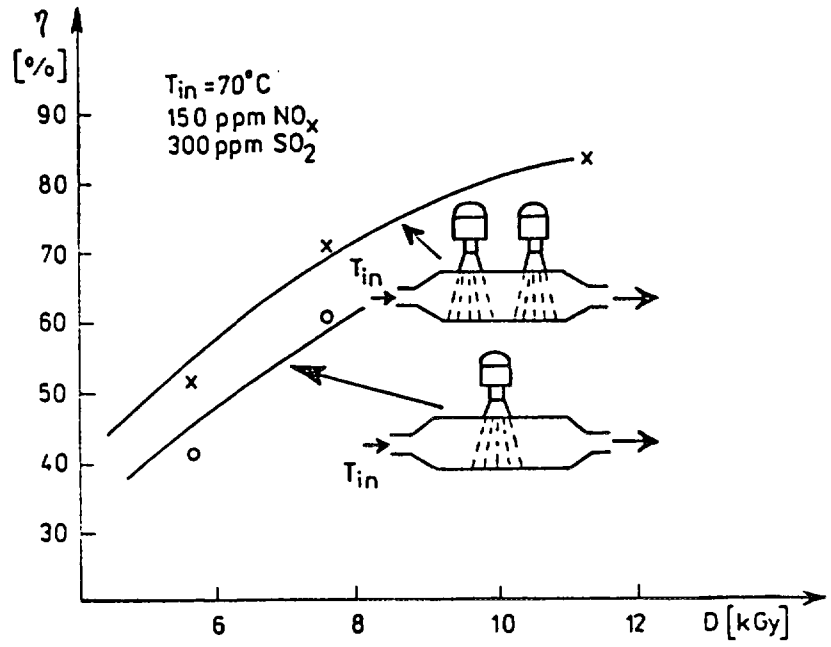


Figure 5: Effect of double irradiation to removal efficiency of NO_X [13].

From the equations above the reaction (10) is the major reverse reaction in oxidation of NO_X. Because of these reverse reaction the removal efficiency of NO_X is not linear functio of irradiation (see Fig. 4).

Removal efficiency can be enhanced with multi stage irradiation [48, 49, 10, 11, 8, 13]. Chmielewski *et al.* has studied that double irradiation raises the NO_X removal efficiency about 10% higher than in the single irradiation process (see Fig. 5). Energy savings of two stage irradiation will be nearly 20% if same purification effect is wanted as in single irradiation. Improvement of efficiency may come from the conversion of some intermediate products to stable forms during the interruption of irradiation [67]. Efficiency can be improved further by optimizing the energy consumption between the irradiation zones [12].

The removal efficiency of NO_X is not affected only by irradiation dose and multi stage irradiation but also the temperature. The dependence removal efficiency of NO_X in double

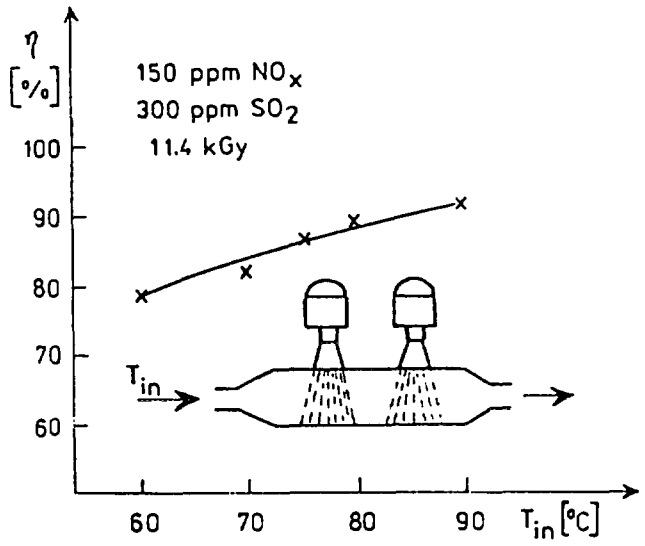


Figure 6: Removal efficiency of NO_X as a function of temperature [13].

irradiation process from temperature is presented in Fig. 6.

3.3 Oxidation of SO_2 and formation of sulphur acid

Sulphur dioxide withdraws from flue gases through two routes: with thermal reactions (see section 3.4) or with radical reactions. According to Namba *et al.* SO_2 removal is mainly occured through thermal reactions [48, 49].

The most important oxidizer of SO_2 in the gas phase is the OH radical. The process has the steps:

$$SO_2 + OH + M \rightarrow HSO_3 + M \quad (16)$$

$$HSO_3 + O_2 \rightarrow SO_3 + HO_2 \quad (17)$$

which are practically immediately followed by

$$SO_3 + H_2O \rightarrow H_2SO_4 \quad (18)$$

The removal efficiency of SO_2 increase with increasing radiation dose, with decreasing temperature of flue gas, with increasing relative humidity and increasing concentration of ammonia [68, 16, 33, 60].

3.4 Influence of ammonia addition

Amount of ammonia addition used in flue gas treatment in practical to be proportion to the input NO_X and SO_2 concentrations with the equation

$$[NH_3] = s \cdot ([NO_X] + 2 \cdot [SO_2]) \quad (19)$$

in which the s is the ammonia stoichiometry. Normally the value of s is near 1. (Thus the ammonia is the most abundant harmful component of flue gas after carbon dioxide!)

In the electron beam treatment system the ammonia is added to flue gas to neutralize the nitric and sulphuric acids born by irradiation. However significant amounts of ammonia also take part to the process other ways. It has been found that the NO concentration

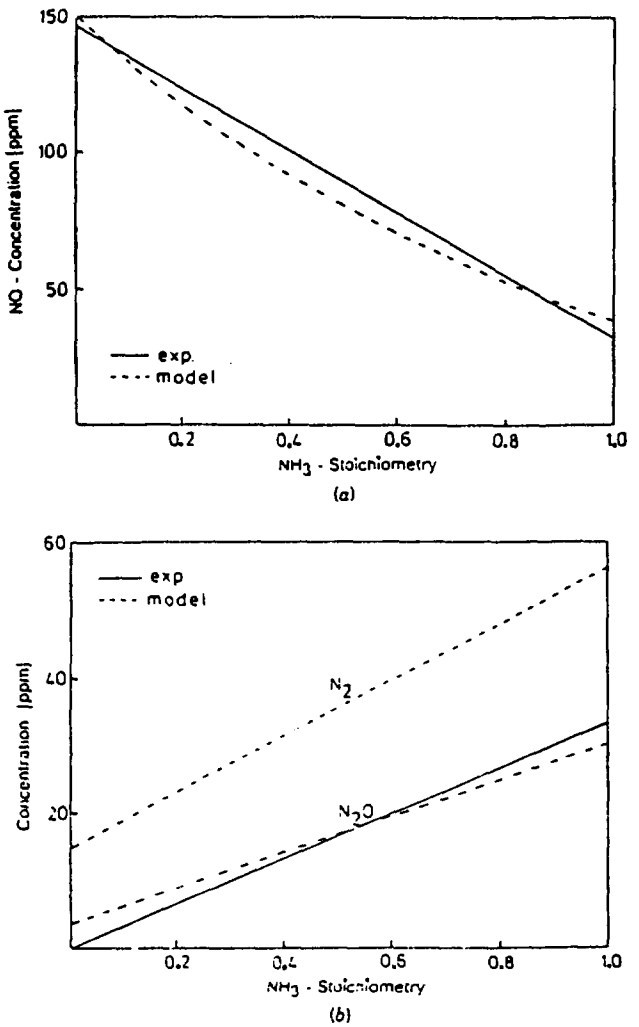


Figure 7: (*a*) NO removal as function of ammonia stoichiometry. Initial conditions: 360 ppm NO, 540 ppm SO_2, r.h.=27%, D=12 kGy. (*b*) N_2 and N_2O formation as function of ammonia stoichiometry. (Conditions same as in part *a*)[46].

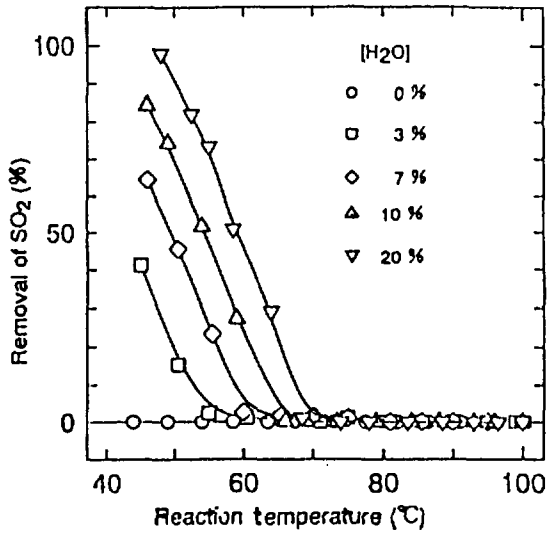
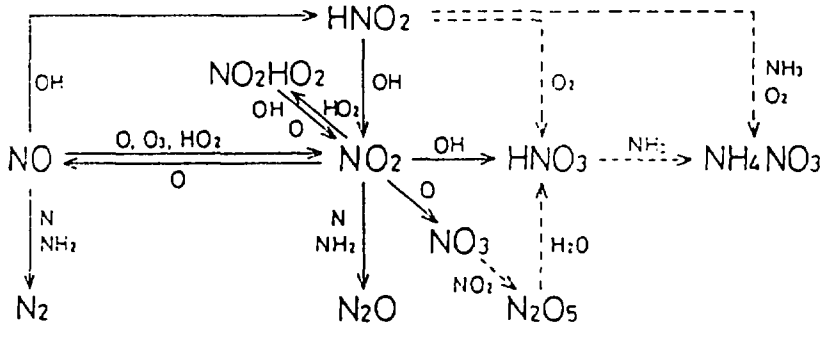
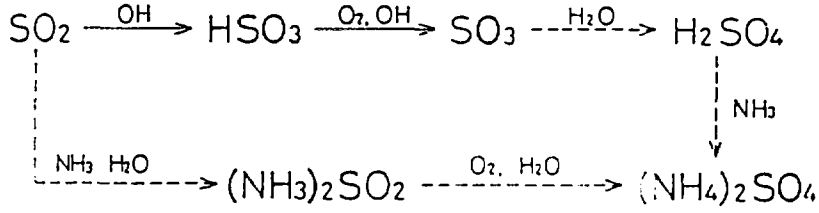


Figure 8: SO_2 removal as a function of temperature with different water vapour concentrations. Initial conditions: NH_3 (1200 ppm), SO_2 (600 ppm), NO (225 ppm), oxygen (10%), rest nitrogen [67].



(a) Removal of NO_X



(b) Removal of SO_2

Figure 9: Schematic view of chemical process of removal of NO_X (a) and SO_2 (b) from flue gases with electron beam treatment [67].

decrease linearly wiht rising ammonia stoichiometry. (see Fig. 7). This originates from decomposing of ammonia by OH radical:

$$NH_3 + OH \rightarrow NH_2 + H_2O \quad (20)$$

This reaction yields the amidogen radical which can change NO and NO_2 to molecular nitrogen and nitrous oxide:

$$NO + NH_2 \rightarrow N_2 + H_2O \quad (21)$$
$$NO_2 + NH_2 \rightarrow N_2O + H_2O \quad (22)$$

The reaction (22) is the major source of nitrous oxide in the electron beam flue gas treatment process although a small amount of nitrous oxide will yield for example from reaction (14). In Fig. 7b can be seen that at those conditions roughly about 15±5% of input NO is converted to molecular nitrogen at ammonia stoichiometry between 0.5 and 1. The molecular nitrogen which is produced at zero ammonia stoichiometry comes from the reaction (12). Namba *et al.* have researched the nitrogen balance of e-beam process by isotope-labeled ^{15}NO as input and then detecting the end products by mass spectrometer [50, 52].

Tokunaga *et al.* have investigated thermal reactions between SO_2 and NH_3 when simulated flue gas is not irradiated. Removal efficiency of SO_2 of thermal reactions depends strongly from reaction temperature and relative humidity (see Fig. 8). Furthermore important parameters are oxygen concentration and surface material of reaction chamber because thermal reaction between ammonia and sulphur dioxide take place at surface of materials (walls of the reaction chamber, ducts, surfaces of aerosol particles) [67]. Thermal reaction taking place at surfaces of materials explains why there has been accumulation of reaction material in ducts in some material balance tests [52, 34].

The whole chemical process of electron beam treatment process can be seen in Fig. 9 in a simplified and compressed form. In the Fig. 9 continuous line means chemical reaction caused by radicals and broken line means thermal reaction. The unsimplified process would be much more complicated, for example AGATE code developed at KfK involves 739 reactions and 95 different species [44, 46].

4 CURRENT STATUS OF THE E-BEAM FGT PROCESS

From the early begining in the Japan the commercializing has been one of the motivating forces behind many research projects of electron beam flue gas treatment. Thanks to those inquests electron beam process has nowadays lower or equivalent costs than other flue gas treatment methods if by-product credit is observed [24]. Reasons to this good economical competitivity are recent innovations in the zone irradiation and the improvement of apparatus.

If multi stage irradiation is used then accelerator power can be reduced significatively. It has been reported that power savings could be even higher than earlier in the section 3.2 mentioned 20% (see Fig. 10). In the recent tests Namba *et al.* got moderate results in the condition of triple irradiation with 7.7 kGy dose. The average removal rates of SO_2 and NO_X for 7 hours were 94% and 80%, respectively. In same tests it was found that there was no difference of NO_X concentration between dwelling time of 0.5s and 0.05s [53]. The last innovation in the multi stage radiation is to use non equal dose delivery between each stage. The highest dose should be applied in the first, lower at second and

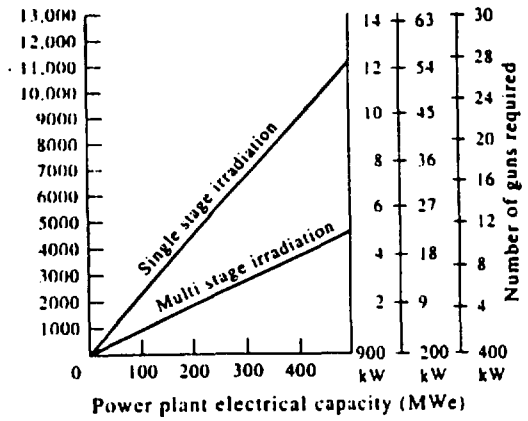


Figure 10: Electron beam power requirement versus power plant electrical capacity [24].

eventualy if applicable at third gas irradiation stage. For double irradiation the optimal dose delivery ratio is 0.6 at the first and 0.4 at the second stage [12]. In the commercial e-SCRUB^{TM} which is offered by Research-Cottrell and Virginia Accelerators Corporation and which use the double irradiation with optimal dose delivery the removal rates of SO_2 and NO_X for higher sulfur coal (> 2.5%) are 98% and 80%, respectively, and the needed dose is then under 10 kGy [29].

The most important and expensive part of apparatus in the electron beam treatment process are accelerators. For example they constituted 17% of investment cost and their electricity consumption constituted nearly 20% of total annual cost estimate for EPS Pomorzany flue gas treatment plant [5]. If the removal of NO_X is not very important (for example because of low NO_X burning) the process can be adjusted to (see Fig. 11) high SO_2 and low NO_X removal to save energy and investment costs. The dose needed to achieve the 98% removal rate of SO_2 is then about 2.5 kGy. This kind of process paramiters will be applied for example in China (see Table 1), where low about 10% NO_X removal and about 80% SO_2 removal with low dose rate (4 kGy) will be achieved [17, 39].

5 FUTURE TRENDS

At this very moment the electron beam flue gas processing has became to a commercial level. There are constructions going on in Poland and in China for large scale treatment plants. In the Estonia the plans for using e-SCRUB system are nearly ready [63].

The future of electron beam processing seems to be promising. The commercial electron beam treatment systems have lower levelized cost than flue gas desulfurization systems alone for high sulfur coal. Because of the world endless need of food the fertilizer by-product is valuable and it will cover the cost of ammonia needed in the process.

Although the sceneries look promising there is a lot of research to do. There are some research plans to resolve the accumulation of reaction materials in ducts. The principal goals to be achieved for accelerators applied at industrial scale for flue gas treatment are according to Zimek as follows [74]:

- high reliability for long time operation (6000-8000 h/y),
- electron beam cost reduction (1.5-2 $/W),
- electrical efficiency >80%,

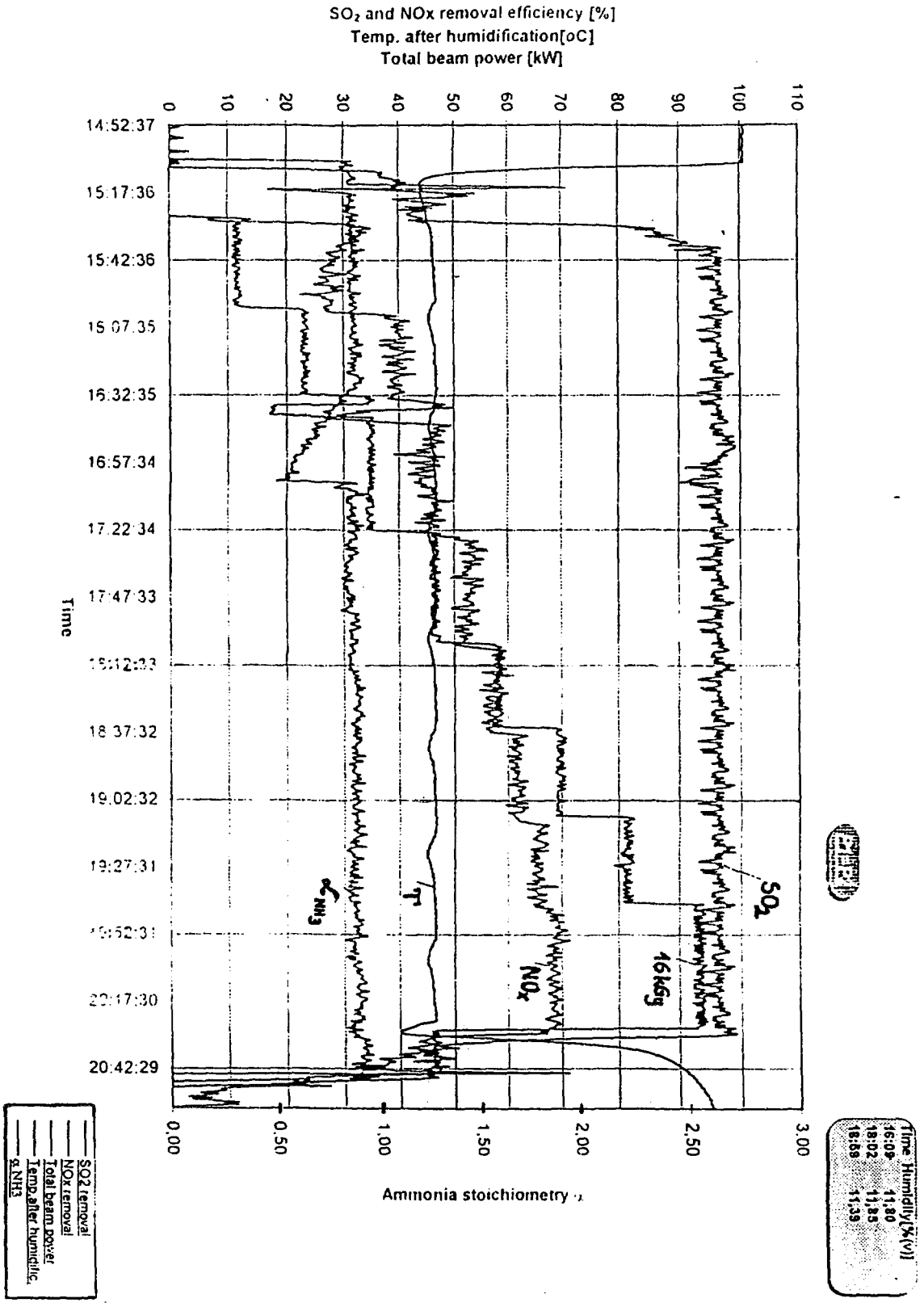


Figure 11: SO_2 and NO_X removal efficiency in high humidity test in Kawęczyn pilot-plant at 02.21.1995.

- low level losses and high current density windows,
- a fault protection system.

And there still are some optimization work to be done. For example Baranchicov *et al.* have shown that with proper reaction parameters (current density, humidity, particles size and so) the energy costs of oxidation could be reduced remarkably [4].

6 CONCLUSIONS

In this review we have referred most important points of electron beam flue gas treatment in the history, the chemical process during the treatment and the current status and some of future trends.

We have shown that electron beam flue gas treatment is reliable for simultaneous removal of SO_2 and NO_X with high removal rates; 98% and 15-80% (depending on ones necessity), respectively. The treatment is a dry process without waste waters. The by-product of the process is the valuable nitrogen fertilizer which have been tested in several growing tests.

Electron beam flue gas treatment plants will have lower levelized cost than competitave processes. This together with high performance and reliability makes it easy to say that in the near future electron beam flue gas treatment process will be the best selection for many power plants around the world.

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