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DEVELOPING WET VARIANTS OF ELECTRON BEAM REMOVAL OF NO_x, SO_2, AND PARTICULATE FROM FLUE GAS

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8.1 INTRODUCTION

The Energostal Institute (Kharkov, Ukraine), together with the Physical and Technical Centre of the Nuclear Physics Institute, Siberian Department of Russian Academy of Sciences (Lipetsk, Russia), is carrying out a program of research, testing, and industrial application of electron-beam (EB) technology. Since 1988, investigations were carried out at the facilities of the Novolipetsk Metallurgical Plant (Lipetsk, Russia). Since 1991, the program has been sponsored by the Donbassenergo Company (Donbass, Ukraine), and joined by the following organizations: Nuclear Physics Institute of Siberian Department of Russian Academy of Sciences (NPI of SD of RAS) (Novosibirsk, Russia), Kharkov Polytechnic University (Kharkov, Ukraine), and Russian Research Centre Kurchatov Institute (Moscow, Russia).

The complex of investigations are being carried out in the following directions: research and testing of the technology at a small-scale industrial facility; investigation of physical, chemical, and crystallophysical aspects of EB processes at this facility; and research and development of a technology to utilize the EB byproducts—sulfate and nitrate of ammonia (SNA)—in farming as fertilizers, engineering of large-scale EB pilot demonstration plants for power and metallurgical industry, development, and manufacturing of special-purpose high-power electron accelerators for EB technology.

8.2 A COMPLEX OF INVESTIGATIONS OF EB GAS TREATMENT ON A SMALL-SCALE INDUSTRIAL INSTALLATION

This section covers the investigations carried out in 1989–1992 on a small-scale industrial installation at Novolipetsk Metallurgical Plant (Lipetsk, Russia) (EB

processes and their physical, chemical and crystallophysical aspects), as well as the research and development of a technology to utilize the EB byproducts: SNA salts.

8.2.1 First Round of Experiments

The first round of experiments (1989–1990) [1, 2] was aimed at studying the basic processes of SO_2, NO, and NO_x conversion in real flue gases without ammonia addition prior to irradiation.

Real flue gases (low-grade coal and coke combustion products), after cooling and dedusting in a wet dust collector, were irradiated with the help of an electron accelerator ELV-4 (E = 1.5 MeV, 45 kW) designed by NPI of SD of RAS; the electron beam was let out through a foil. The reaction chamber was designed as a gas duct of a rectangular cross section; its bottom and top horizontal walls were equipped with foil windows. The chamber height was 200 mm; the distance between the sidewalls was 80 mm. Thus, the whole scanned electron beam passed through the chamber from top to bottom with the gas flowing in the horizontal direction. This chamber design ensured uniform irradiation of gas, minimized the heating of the chamber sidewalls, and enabled determination of the irradiation dose absorbed by the flue gas with fairly high accuracy, in contrast with those experiments where the dose distribution is very nonuniform and in which one usually measures a certain averaged dose absorbed by the gas being treated. In our experiment, the dose was measured by a calorimetric method.

The main results of these experiments (Figs. 8.1–8.4) present relationships between SO_2, NO, and NO_x conversion and NO_2 generation on one hand and the specific dose, on the other hand, the specific dose being the dose D divided by the initial concentration of SO_2 ($[SO_2]_0$), NO ($[NO]_0$) and NO_x ($[NO_x]_0$).

The SO_2 conversion energy consumed for one molecule made up E_0 = 10.91 eV for humid gas and E_0 = 15.9 eV for dry gas. The gas composition in these experiments was mainly as follows: SO_2—100–600 mg/m^3, NO_x—up to 50 mg/m^3, CO—not more than 0.01%, dust loading—up to 270 mg/m^3, T = 60–105°C. Figures 8.1–8.4 clearly show the dependence of the conversion degree on the specific dose and gas humidity [3, 4]; a comparison with the results [3] shows the impact of ammonia feed on NO_2 generation.

8.2.2 Second Round of Experiments

The second round of experiments (1991–1992) was aimed at investigating and testing a method of flue-gas irradiation using a concentrated outlet of the electron beam from the accelerator without scanning.

In these experiments, ammonia vapor was fed into the flue gas before the irradiation, which resulted in formation of ammonia salt particles in the process of irradiation. Special methods were used to investigate the growth of these particles in the different sections of the gas-handling system after irradiation.

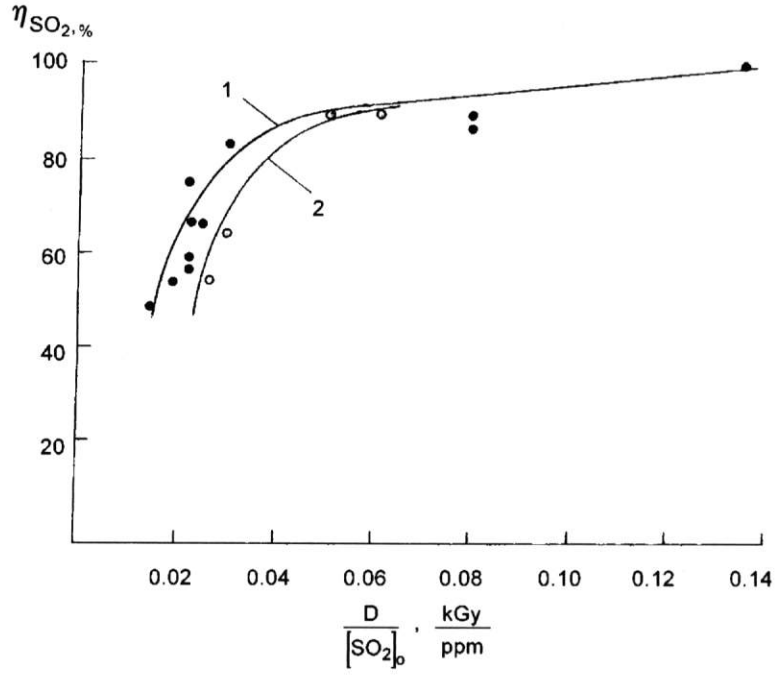

Figure 8.1 SO_2 conversion efficiency η_{SO_2} versus specific dose of irradiation $D/[SO_2]_0$ (1—high moisture content; 2—low moisture content).

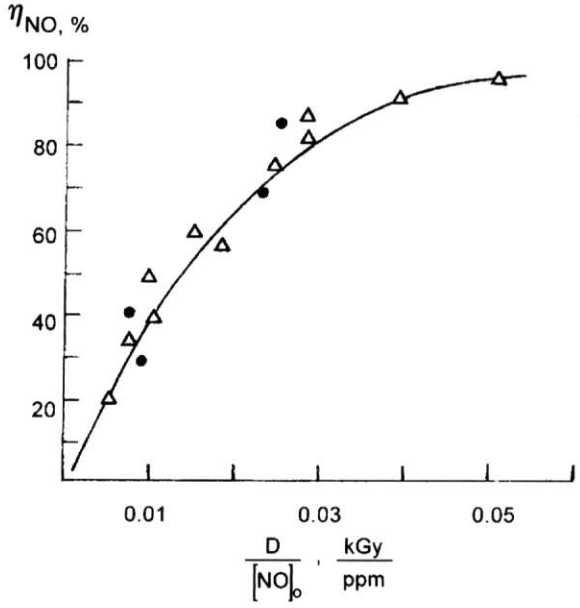

Figure 8.2 NO conversion efficiency η_{NO} versus specific dose of irradiation $D/[NO]_0$ [△—TU, Karlsruhe (Germany) [3]; •—Energostal (Ukraine)].

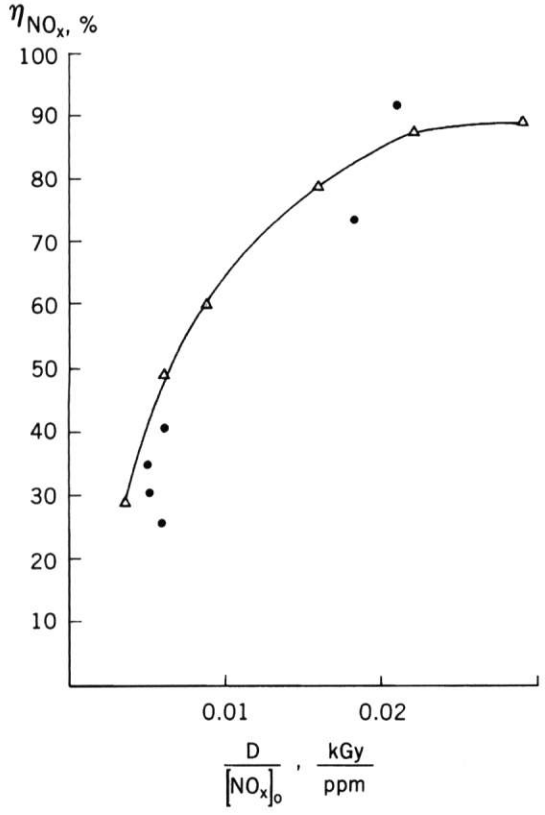

Figure 8.3 NO_x conversion efficiency η_{NO_x} versus specific irradiation dose $D/[NO_x]_0$ [△—Ebara (Japan) [4]; •—Energostal (Ukraine)].

The main results of these investigations are as follows:

1. The method of releasing the electron beam out into the reaction chamber from the accelerator ELV-4 by means of a concentrated outlet without scanning proved to be fairly efficient. In this case, the reaction chamber had a conical shape with apex angle 30° and height about 1.8 m [5]. A system of special probes was used to measure the electron current density in the radial cross section of the chamber 1 m below the concentrated outlet; the nonuniformity of the gas irradiation did not exceed 58%.

2. A relationship of SO_2 conversion efficiency versus specific dose of irradiation similar to that shown in Figure 8.1 was obtained; the influence of the gas moisture content was also clearly manifested. The conditions under which the curves were obtained are as follows: SO_2 and NO_x inlet concentrations are approximately the same as those given in Section 8.2.1: R_{NH_3} = 0.12–0.35, T = 64–95°C, gas moisture content—47.3 g m^{-3}.

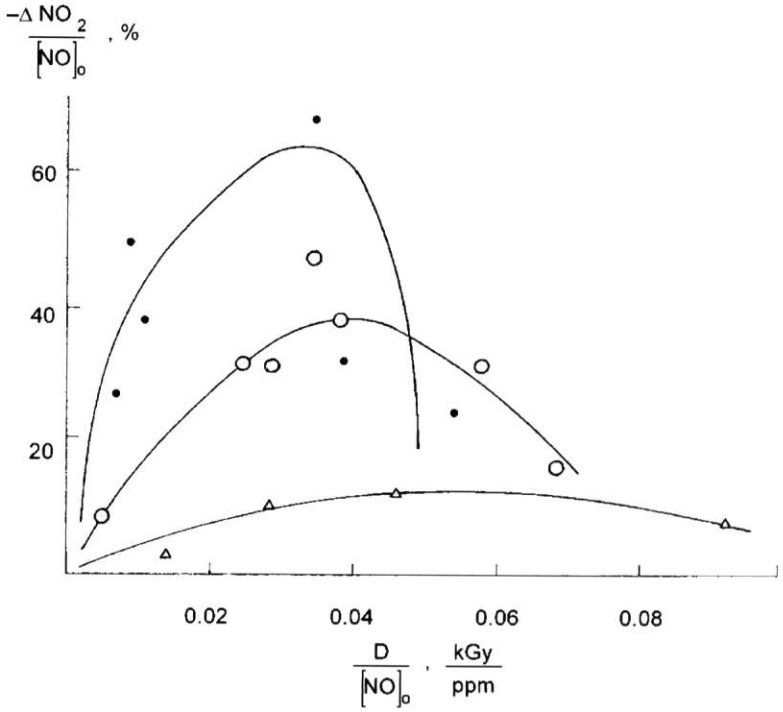

Figure 8.4 NO_2 generation rate $-\Delta NO_2/[NO]_0$ versus specific irradiation dose $D/[NO]_0$ [•—Energostal (R_{NH_3} = 0; $[SO_2]_0$ = 200 ppm); ○—TU, Karlsruhe (R_{NH_3} = 0.6–0.8; $[SO_2]_0$ = 0 ppm [3]); △—TU, Karlsruhe (R_{NH_3} = 0.6–0.8; $[SO_2]_0$ = 400 ppm [3])].

3. Glass substrates located in the gas duct immediately after the irradiation chamber (location 1) and further downstream were used to study the morphology of the forming particles. A high-resolution optical and electron microscopy showed the following.* Immediately after irradiation, under the conditions mentioned in paragraph 2 (above), fine particles of submicrometer–micrometer dimensions grow in the gas on the substrates (Fig. 8.5). The nature of the crystal growth (dendrite growth; growth of ~1-μm-diameter, ~100-μm-long whiskers; high nucleus density) suggests that the crystallization in the duct at the outlet of the reaction chamber proceeds under conditions of high supersaturation. As the gas flows further downstream along the duct (within a time interval of 0.5–1 s) (location 2) away from the reaction chamber, particles continue to grow in the gas on the substrates. These particles have a conspicuous crystalline form; the crystals measure about 100 μm (Fig. 8.6). Under a microscope they display an optical anisotropy typical for crystals of ammonia salts of sulfuric

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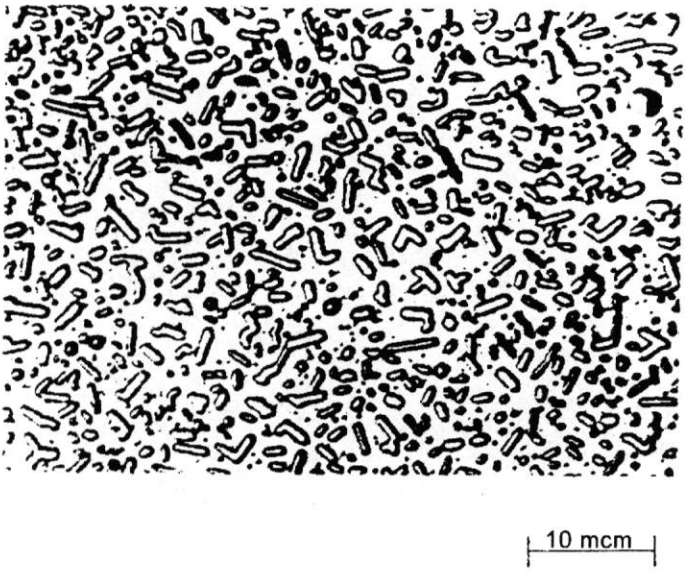

Figure 8.5 Initial stage of crystalline deposit formation on a glass in location 1.

and nitric acids. The forms of growth of these crystals turned out to be very similar to those of ammonia salts of sulfuric and nitric acids and their double salts grown in water solutions. An analysis of the results obtained suggests that the most probable mechanism of forming crystalline aggregates during EB gas treatment in the presence of moisture and ammonia is the growth of ammonia salts from the liquid film of their water solution, which is wetting the faces of the growing crystals (Fig. 8.6). A phase x-ray diffraction analysis of the crystalline aggregates obtained under the conditions described above has shown that these aggregates consist mainly of ammonia sulfate with insignificant admixtures of the double salt $(NH_4)_2SO_4 \cdot 2NH_4NO_3$ and ammonia nitrate, which is natural for the composition of the gas irradiated at the experimental facility.

8.2.3 Electron-Beam Byproducts

Much attention was paid to effective utilization of EB byproducts—ammonia sulfate, ammonia nitrate, and double salt $(NH_4)_2SO_4 \cdot 2NH_4NO_3$ (SNA)—as fertilizers for farming. These investigations have been carried out since 1991 in two directions: (1) development of an effective technology of SNA granulation adapted to EB treatment technology and (2) study of efficiency of SNA with admixture of fly ash as a fertilizer and the impact of this mixture on the quality of the farming produce. The latter problem is of particular importance since EB treatment of a utility flue gas yields a byproduct consisting of SNA with fly-ash admixtures. The fly ash, in turn, contains heavy metals, and the issue of what the allowable ash content in fertilizers is becomes very critical.

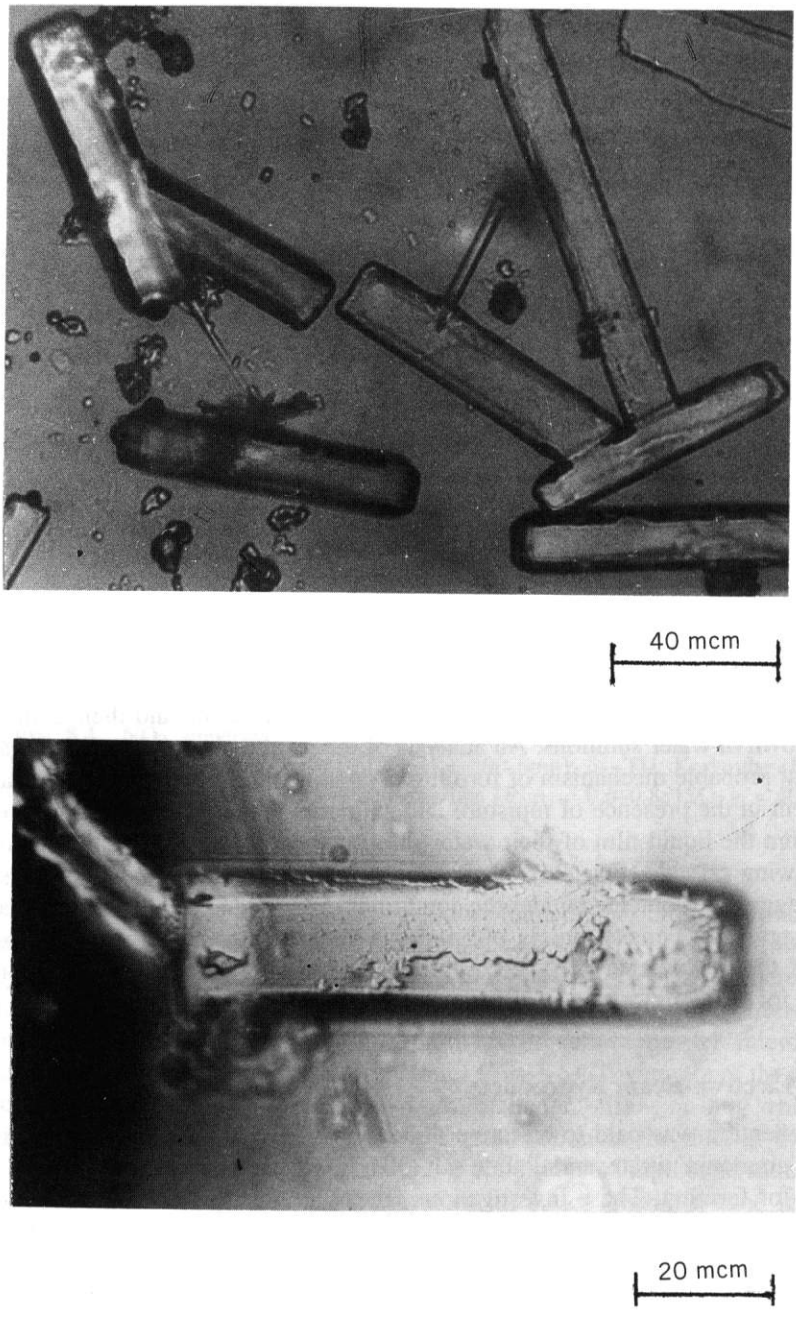

Figure 8.6 Steplike structure of surface of crystals growing on a substrate in location 2; photographs taken in transit light: (a) nonpolarized; (b) polarized.

Model investigations of SNA efficiency as a fertilizer are being carried out according to the standard procedures of long-term tests practiced when studying fertilizers and their effect on the vegetable produce quality. These investigations showed that the mixture of ammonia sulfate, ammonia nitrate, and 2–6% fly ash is a very effective fertilizer. The use of this mixture as a fertilizer does not impair the quality of the produce and does not result in accumulation of heavy metals in it.

An optimal technology of SNA granulation was developed using standard Ukrainian equipment adapted to the wet variant of EB technology.

8.2.4 Electron-Beam Technology From the Point of View of Physics and Chemistry

The investigations mentioned above suggest that EB technology is basically a combination of two physical and chemical processes: (1) radiation and chemical transformations and (2) phase transitions (i.e., the formation of solid particles of SNA growing from gaseous or liquid phase). The supersaturation of SNA vapors in the gas is highest immediately after irradiation; it gradually decreases as the gas flows along the gas-handling system, remaining, however, always above zero even at the outlet of the smokestack. Thus, favorable conditions are created for the growth of SNA crystals, that is, for accumulation of deposits on the surfaces of the ducts and equipment, such as inside the fabric of bag filters or on the elements of electrostatic precipitators.

From this point of view, a dry variant of the technology looks less reliable as it may entail serious problems in terms of deposits and clogging. In contrast to this, a wet variant of the technology appears to be more reliable and serviceable, taking into consideration a good solubility of SNA as well as the fact that in many countries the technology of fertilizer granulation is also wet.

8.3 ENGINEERING OF A 100,000-m^3/h EB DEMONSTRATION PILOT UNIT TO REMOVE NO_x, SO_2, AND PARTICULATE FROM FLUE GAS OF SLAVYANSKAYA POWER PLANT (DONBASS, UKRAINE)

With all the advantages of the EB technology of flue-gas purification that are now quite evident, for wide application of the technology in industry it is still necessary to solve several important problems, namely: (1) essentially reduce the power consumption for the EB gas treatment, (2) ensure reliable and effective collection and utilization of the byproduct (SNA), and (3) organize development and serial production of powerful electron accelerators. Development of EB technology also requires its testing on large-scale demonstration units of about 100,000-m^3/h capacity or more. An essential factor for development and wide application of EB technology is the prevailing conditions in different countries, in particular the level of development and the current shape of the

economy, the availability of pollution-control installations in industry, and the general ecological situation in a particular country. From this point of view, strange as it may seem, the most intensive application of EB technology can be expected in the countries with a generally poor ecological situation and a low level of availability and efficiency of pollution-control devices, but with modern nuclear physical technologies. Therefore, it is not by chance that construction of the largest EB installations is planned for the Ukraine and Poland, and in China such an installation has already been built.

The Energostal Institute (Ukraine, Kharkov), together with the coauthors (see the list of authors of this chapter), has developed a working design of a 100,000-m^3/h EB demonstration unit to treat the flue gas of boiler 9 at the Slavyanskaya thermal power plant (Donbass, Ukraine) [6].

When engineering the unit, a wet method of ash and particulate collection was chosen. An analysis of EB technology development reports, as well as analysis of a complex of physical and chemical processes that both underlie this technology and take place in the environment, show that, according to our opinion, the wet variant is a very promising way for development of this technology. In this variant, the following main problems of EB technology are solved in the best way: reduction of power consumption for irradiation, efficiency of ammonia salt collection, and effective utilization of EB byproducts in the form of granulated fertilizers. It was obviously not by chance that the Mother Nature uses both wet and dry variants for realization of the natural radiation and chemical processes of SO_2 and NO_x transformation in the atmosphere and the subsequent cycle of the converted chemical substances in the environment. Obviously, both of these technology variants can be applied in engineering to advantage.

The demonstration unit is designed to identify and test, under real industrial conditions, the operating parameters to reduce the power consumption due to different variants of gas conditioning before irradiation, as well as to determine the most effective operating conditions for collecting the ammonia salts. After completion of these pilot-plant tests, construction of an ammonia salt granulation unit is planned to begin at the same time as construction of a full-scale EB installation to treat the whole amount of the flue gas from boiler 9 of the Slavyanskaya power plant.

For collecting the SNA particulate matter, high-pressure Venturi scrubbers with variable throat were selected for the following reasons: (1) this dust is known to be fine and (2) no information is available in the literature on the required operating conditions of wet control systems collecting this dust.

Application of these Venturi scrubbers is expected to enable identifying, in the process of investigations, the optimum conditions of collecting SNA aerosols. A similar control system is designed for the gas pretreatment (fly-ash removal) before EB treatment proper. The SNA salt collecting unit has a closed-loop recycled-water supply system to enable buildup of the SNA salt concentration in the solution to the level required for granulation. A service water makeup will compensate for water loss with the treated gas and suspension blowdown.

A schematic of the demonstration unit is shown in Figure 8.7. After the existing fly-ash collector (not shown), the boiler flue gas passes to the flue of the boiler (1) to be emitted into the atmosphere through a smokestack. By means of a VVN-20 fan, part of the flue gas (up to 100,000 m^3/h) is tapped to the demonstation unit. The flue-gas flow rate is adjusted according to the technical characteristics of the ELV-6 electron accelerator to ensure the consumed irradiation dose required for the effective gas treatment.

First, the gas passes through a gas pretreatment unit comprising a variable-throat Venturi scrubber (2), a separator tank (3), and a droplet separator (4), where the gas is dedusted to the required level. After exiting the pretreatment unit, water is discharged into the existing hydraulic ash removal system. If necessary, the pretreated gas can be heated to the required temperature in the heater (5). Then the required amount of ammonia is injected into the gas in the form of an ammonia–steam mixture generated with the help of an ammonia desorber (6) or by means of pneumatic atomization of the ammonia water in the ammonia water injector (7). In both cases, ammonia water is pumped from the ammonia water tank (9).

To desorb ammonia from ammonia water, a desorber based on mixing ammonia water with water steam is used. Ammonia is injected into the gas duct upstream of the reaction chamber through a special spraying device.

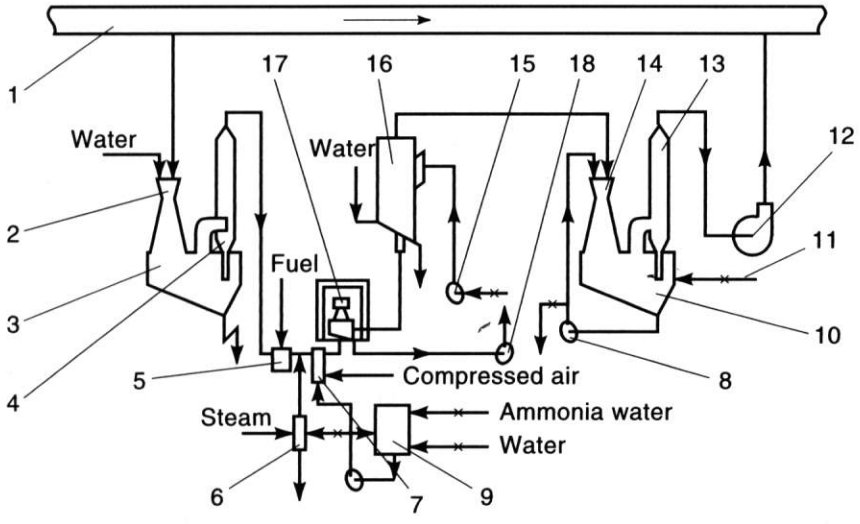

Figure 8.7 Schematic of EB demonstration unit at Slavyanskaya thermal power plant (1—boiler flue; 2—variable-throat Venturi scrubber; 3—separator tank; 4—droplet separator; 5—gas heater; 6—ammonia desorber; 7—ammonia water injector; 8—circulation pump; 9—ammonia water tank; 10—separator tank; 11—water makeup; 12—induced-draft fan VVN-20; 13—droplet separator; 14—variable-throat Venturi scrubber; 15—fan of DH10/Y type; 16—vikhr (vortex) apparatus; 17—electron-beam unit; 18—solution recirculation pump).

It is also possible to inject finely atomized ammonia water directly into the gas before the reaction chamber. Then the gas is passed to the electron-beam unit (17), where it is irradiated in the irradiation chamber by means of a modernized electron accelerator ELV-6 with beam power up to 150 kW designed by the Nuclear Physics Institute of the Siberian Department of Russian Academy of Sciences (Novosibirsk, Russia). Because of the experimental nature of the demonstration unit and the limited financial capacity of the Slavyanskaya power plant, only one accelerator ELV-6 is used. The demonstration unit can be operated with a gas flow rate of 30,000–100,000 m^3/h, with a maximum consumed irradiation dose of 1.80 and 0.54 Mrad, respectively. Considering the eventual reduction in energy consumption for the irradiation, due to special gas conditioning, the design removal efficiency in the experimental mode is 75% for NO_x and 80% for SO_2.

The accelerator ELV-6 has a double-outlet window protected with foil. The configuration of the reaction chamber and the arrangement of the gas streams ensure uniform irradiation and intensive mixing of the gas during treatment. Accelerated electrons are let into the gas reaction chamber through a foil double window. To remove the particulate matter sedimented in the reaction chamber, a flushing system is provided comprising a solution recirculation pump (18), which delivers these sediments into the water recycle system of the SNA salt collection device. After exiting the electron-beam unit, the gas is passed to a device to condition the gas prior to particulate collection—a *vikhr* (vortex) apparatus (16) with a fan DH10/Y (15) for the secondary air supply. The particles sedimented on the bottom of this apparatus are flushed off and dumped into the water recycle system of the SNA salt collection unit.

Then the gas is passed to this unit, which consists of a variable-throat Venturi scrubber (14), a droplet separator (13), a separator tank (10), and a closed-loop water supply system of the Venturi scrubber. This system consists of a circulation pump (8) and a water makeup component (11) to compensate for the water loss due to blowdown of the salt solution from the system when the SNA salt concentration in the solution reaches 25%. The purified gas is discharged by the induced-draft fan VVN-20 (12) into the flue and then into the atmosphere through the existing smokestack of the boiler.

Table 8.1 presents the initial data and basic technical characteristics of the demonstration unit. The unit is engineered in compliance with valid engineering standards and regulations. The design is approved by the supreme sanitary and environmental authorities of the Ukraine. The key element of the unit—an electron accelerator ELV-6—has already been purchased. The International Atomic Energy Agency has decided to render financial and technical assistance to the demonstration unit project.

The approximate cost of the EB demonstration unit is shown in Table 8.2. It should be noted that the specific capital investments for the conditions of the Slavyanskaya power plant are $43,000 per 1000 m^3/h of the gas being treated, which is much less than similar figures for EB designs developed in other countries.

TABLE 8.1 Initial Data and Basic Technical Characteristics of Electron-Beam Demonstration Unit

Parameter	Value
Boiler type	T-230
Steam-generating capacity (t/h)	230
Consumption of coal by the boiler (t/h)	25
Amount of boiler flue gas (m^3/h)	≤400,000
Gas temperature after existing pollution control installation (°C)	80–95
Ash concentration after existing pollution-control installation (g/m^3)	1.5
Concentration of sulfur dioxide before demonstration unit (g/m^3)	≤5.7
Concentration of nitrogen oxides (g/m^3)	≤1.0
Gas–water moisture content (g/m^3)	30.0
Gas flow rate (m^3/h)	30000–100,000
Ash concentration after gas pretreatment before EB unit (mg/m^3)	100-1000
Ash removal efficiency (%)	≤90
Ammonia salt concentration before salt collection devices (g/m^3)	≤11.4
Ammonia salt concentration after collection devices (mg/m^3)	100–700
Pressure drop across gas-handling system (Pa)	14,500
Fan head (Pa)	15,930
SO_2 removal efficiency (%)	≤80
NO_x removal efficiency (%)	≤75
Ammonia salt removal efficiency (%)	≤94
Consumption of recycle water by Venturi scrubbers, for flushing sediments and agitation of hydraulic seals (m^3/h)	150
Consumption of chemically treated water by accelerator (P = 0.4–0.5 MPa, T = ≤25°C) (m^3/h)	1.5
Service water consumption for cooling fan VVN-20 (P = 0.4 MPa) (m^3/h)	2
Consumption of steam by ammonia desorber (t/h)	1.5
Consumption of natural gas for flue-gas heating ($m^3\ h^{-1}$)	250
Natural-gas pressure (mmH_2O)	400–600
Consumption of compressed air by pneumatic sprayers ($m^3\ h^{-1}$)	1000
Compressed-air pressure (MPa)	0.5
Consumption of ammonia water (25% solution) by desorber (m^3/h)	1.2
Accelerator type	ELV-6
Energy of accelerated electrons (MeV)	1.0
Average density of electron beam current (A/cm^2)	≤5 × 1 0^{-5}
Maximum beam power (kW)	150
Maximum irradiation dose at maximum gas flow rate (Mrad)	0.54
Maximum gas irradiation dose at gas flow rate 30,000 m^3/h (Mrad)	1.80
Power instability (%)	±5
Accelerating voltage ripple (%)	±2
Dimensions of double outlet window (mm × mm)	1700 × 400

TABLE 8.2 Approximate Cost of Electron-Beam Demonstration Unit at Slavyanskaya Power Plant

Part of Project	Ukrainian Prices[a] ($U.S. million)	World Prices ($U.S. million)
Accelerator	0.8	0.8
Process equipment	2.578	3.71
Instrumentation and automatic control	0.256	0.72
Construction materials and erection work	0.616	0.75
Total cost	4.25	5.98

[a]The value of Ukrainian prices in U.S. dollar equivalent continues to change.

8.4 CONCLUSION

In the developed countries, EB gas treatment technology has been developed mainly as a dry process. However, after encountering some difficulties associated with collecting salt particles and fouling gas-handling system, there was some effort to use wet particulate collectors. An analysis of the dry and wet variants of the technology shows that both of them have advantages as well as disadvantages. At first consideration, the dry variant appears to be preferable (no wastewater, no additional energy for product crystallization, etc.). Still, our choice was a wet variant because of the following advantages, which seem to be essential:

1. A higher reliability of EB technology in terms of (a) clogging of gas-handling equipment with SNA deposits and (b) collecting SNA particles. It should be kept in mind that deposits of SNA salts on various elements of the gas-handling system can be controlled by wet methods, as the salts are water-soluble.
2. A droplet mechanism of radiation and chemical transformations can be readily realized in a wet variant to substantially reduce the energy consumption for the EB process.
3. Many countries use a wet technology of SNA granulation from a saturated solution to obtain the end product: granulated fertilizers. This technology fits snugly into the EB flowchart, and the whole process can be arranged with practically no wastewater.
4. In poorer countries with inadequate gas-purification technology (e.g., in countries of the former Soviet Union), wet methods of gas treatment are widely applied as they are more reliable and do not require sophisticated maintenance.

Therefore, a wet variant of EB technology may prove to be more acceptable for these countries as a more reliable one with a lower capital cost.

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