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APPLICATION OF FLUE GAS CLEANING TECHNOLOGIES FOR SOLID FUEL BOILERS OF COMBINE HEAT AND POWER PLANTS

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The ways to achieve the requirements of modern environmental legislation of Ukraine and the European Union regarding the limiting emissions of pollutants from large and medium-sized combustion plants concerning the solid fuel steam boilers of municipal and industrial combine heat and power plants (CHPP) in Ukraine is analyzed in the paper. The environmental requirements and technologies for cleaning flue gases of solid fuel boilers from the main pollutants, namely particulate matter, sulfur dioxide and nitrogen oxides, were considered, and the effectiveness, advantages and limitations for the implementation of these technologies on existing boilers of thermal power plants were analyzed. The existing state of gas cleaning equipment, put into operation more than fifty years ago, does not meet current environmental requirements, and the urgent issue is the reconstruction and modernization of existing and the construction of new gas cleaning plants. The use of fabric filters, electrostatic filters and wet scrubbers with a Venturi tube will allow to fulfill the requirements of European directives on dust emission limit values, and the most rational solution will be to use the existing wet scrubbers with a Venturi tube, which are equipped on the vast majority of CHPPs, by significantly increasing the specific flow rate of liquid for irrigation. For the capture of gaseous pollutants, a promising direction is the use of ammonium reagents for highly efficient desulfurization and obtaining ammonium sulfate as a desulfurization product, which is a mineral fertilizer, and the reduction of nitrogen oxides to molecular nitrogen. The use of an aqueous solution of ammonia in a wet scrubber with a Venturi tube will allow to simultaneously capture fly ash and sulfur dioxide in one device. To reduce nitrogen oxide emissions in CHPP boilers, it is advisable from the point of view of investment costs and spatial conditions to use the method of selective noncatalytic reduction.

Keywords: steam boiler, flue gas, emission, Venturi tube, particulate matter, sulfur dioxide, ammonia, nitrogen oxides.

Introduction

The relevance of the problem of cleaning flue gases from pollutants increases every year due to the strengthening of requirements for environmental protection and human health. The increase in air pollution caused by the activities of industrial enterprises, power plants and vehicles, requires effective technologies to reduce emissions of harmful components, such as sulfur dioxide, nitrogen oxides and particulate matter. One of the main sources of pollution is the activities of energy sector facilities that operate on fossil fuels, in particular coal. A diagram of global electricity production as of 2018 is shown on Fig. 1 [1]. It can be seen that 61% of the world's electricity was produced from fossil fuels, including coal [1].



Coal combustion produces a large number of emissions, with particular attention being paid to particulate matter, sulfur dioxide, and nitrogen oxides, which are the main pollutants.

The dynamics of the reduction of global emissions of sulfur dioxide and nitrogen oxides in electricity production between 1990 and 2019 is shown in Fig. 2 and Fig. 3 [1]. Emissions of these pollutants are decreasing every year. This is due to the development of new and improved flue gas cleaning technologies. The driving force for this is the strengthening of requirements for reducing pollutant emissions.

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In Ukraine, emissions of pollutants in the thermal power industry are regulated by the order of the Ministry of Environmental Protection and Natural Resources No. 309 of 27/06/2006 [2] on the standards of maximum permissible emissions of pollutants from stationary sources (general case) and the order of the Ministry of Ecology No. 62 of 16/02/2018 [3] on the establishment of technological standards of permissible emissions for thermal power (combustion) plants with a nominal thermal capacity of more than 50 MW. Tables 1 and 2 show emission standards for various emissions of pollutants.

The technological standards for permissible emissions for existing combustion plants, which for further long-term operation must undergo modernization of the main and gas cleaning equipment, are shown on Table 2.

Table 1. Maximu (m permissil order No.30	ble emission standards 99) [2]	Table 2. Technological standards for pollutant emissions for modernized thermal power plants (order No. 62) [3]			
Type of pollutants	Mass flow rate, g/h	Maximum permissible emission standards, mg/m ³	Type of pollutants	Nominal thermal power of the plant	Technological standard, mg/m ³	
Particulate matter	> 500	50		(P), MW 50 < P < 100		
by composition	\leq 500	150	Sulfur dioxide	$100 < P \le 300$	250	
Sulfur dioxide and				300 < P	200	
trioxide in terms	\geq 5000	500	N::	$50 \le P \le 100$	300	
of sulfur dioxide			Nurogen oxides	100 < P	200	
Nitrogen oxides			Particulate matter,	$50 \le P \le 100$	30	
in terms of nitrogen	\geq 5000	500	undifferentiated	$100 < P \le 300$	25	
dioxide			by composition	300 < P	20	

Table 3. Emission limit values for existing combustion plants under Directive 2010/75/EU [4]

Туре	Nominal thermal power	Emission limit values, mg/nm ³					
of pollutants	of the plant (P), MW	Solid fuel	Biomass				
	$50 \le P \le 100$	400	200				
Sulfur dioxide	$100 < P \le 300$	250	200				
	300 < P	200	200				
	$50 \le P \le 100$	300	300				
Nitrogen oxides	$100 < P \le 300$	200	250				
	300 < P	200	200				
Darticulata mottor	$50 \le P \le 100$	30	30				
differentiated	$100 < P \le 300$	25	20				
by composition	300 < P	20	20				

 Table 4. Emission limit values for existing medium

 combustion plants under Directive 2015/2193/EU [5]

Туре	Nominal thermal	Technological standard, mg/m ³			
of pollutants	(<i>P</i>), MW	Solid fuel	Biomass		
Sulfur dioxido	$1 \le P \le 5$	1100	200		
Sumur dioxide	5 < <i>P</i>	400	200		
NI:4	$1 \le P \le 5$	650	650		
Nitrogen oxides	5 < <i>P</i>	650	650		
Particulate matter	$1 \le P \le 5$	50	50		
differentiated by composition	5 < <i>P</i>	30	30		

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After signing the Association Agreement in 2014, Ukraine, in particular, undertook to comply with the requirements of Directive 2010/75 "On Industrial Emissions" [4] regarding the limitation of emissions from large combustion plants with a rated thermal input of at least 50 MW (Table 3) and Directive 2015/2193/EU "On the limitation of emissions of certain pollutants into the air from medium combustion plants" [5] with a rated thermal input of 1 to 50 MW by 2030 (Table 4).

In Ukraine, solid fuel boiler units are present in municipal and industrial CHPPs, for example, CHPPs of sugar factories, of which there are currently 29 in Ukraine. The volume of emissions from municipal CHPPs in Ukraine as of 2019 is shown in Table 5[6].

It is known that in the countries of the European Union the construction of new boilers and gas purification plants was financed by an additional tax and tariff on electricity for the population and industry. In our country, Article 243 of the Tax Code of Ukraine [7] provides for a tax rate for pollutant emissions. Thus, for 1 ton of pollutant emissions, in particular, for nitrogen oxides, 2574.43 UAH should be paid, for sulfur dioxide -2574.43 UAH, for particulate matter -96.99 UAH.

Table 5. Emissions from the largest municipal CHP plants in Ukraine (2019) [6]							
СНЪЪ	SOn tons	NO _x ,	PM,				
CIIII	50_2 , 10115	tons	tons				
Kalush	10599	612	2354				
Chernihiv	5782	1305	1869				
Darnytsia	4572	2131	2665				
Cherkasy	4488	7804	3626				
Kharkiv-2	3241	1258	2817				
Myronivskyi	2753	454	219				
Kramatorsk	1966	685	1891				
Sumy	598	345	829				
Total quantity	33999	14594	16270				

The amount of environmental tax paid by all municipal Ukrainian CHPPs operating on solid fuel, according to Table 5, was about 123 million UAH, or 5.2 million dollars (at the dollar exchange rate as of 12/30/2019). This does not cover the costs of constructing new gas purification plants at all. However, increasing the tariffs of the environmental tax will directly affect the cost of electricity and heat. However, the authors of the paper do not consider the issue of increasing the tariffs of the environmental tax, but instead focus on solving technological issues of reducing emissions of pollutants from thermal power plants.

Taking this into account, the question arises for heat power engineers: how to achieve the level of European limit values for pollutant emissions when modernizing existing boiler units. This is especially relevant for thermal power plants that must generate electricity and supply thermal energy for municipal and industrial needs. Steam boilers of municipal CHPPs have a steam capacity of 110 to 230 t/h, which meets the conditions for the application of Order No. 62, and in industrial CHPPs – a steam capacity of up to 40 t/h, which meets the conditions of Directive 2015/2193/EU. These boilers were manufactured in the 1950s and 1960s, when the main environmental aspect of the energy sector was the removal of particulate matter, so they are mainly equipped with wet scrubbers with a Venturi tube and had a design dust removal efficiency of about 97%, which at an input dust content of 30 g/nm³ corresponds to an output dust concentration of 900 mg/nm³. Battery cyclones were generally installed on the boilers with a cyclone fuel of the 2nd stage of the Cherkasy CHPP, which almost do not capture dust particles smaller than 10 microns. In recent years, 2 new electrostatic precipitators with an efficiency of more than 99.5% have been installed on two boilers at the Darnytsia CHPP. All other dust removal equipment requires modernization or replacement. At the CHPPs of sugar factories, battery cyclones or bag filters (new) are used to remove ash particles. Sulfur and nitrogen purification systems are not installed on the CHPP boilers.

The purpose of this paper is to review modern methods of cleaning flue gases from particulate matter, sulfur dioxide and nitrogen oxides, analyze their efficiency, advantages and limitations, as well as identify the main trends and the possibility of their implementation in the modernization of Ukrainian thermal power plants.

Particulate matter: impact and emission reduction methods

Particulate matter is formed by the thermal decomposition of the mineral component of the fuel and is the result of incomplete combustion of its organic part. Such particles are classified by size, which determines their ability to penetrate the body. In high concentrations, particulate matter can cause respiratory diseases, allergies, cardiovascular diseases [8], and sometimes can lead to lung cancer [9–11].

Industrial technologies for dust removal of flue gases are shown in the diagram shown in Fig. 4 [12].

The main dust removal devices in the thermal power industry are fabric (bag) filters, cyclones, electrostatic precipitators and wet scrubbers. Different technologies have different capture efficiency depending on the size of particulate matter, as shown in Fig. 5 [13].



Fabric filters are considered the most effective in terms of capturing particles smaller than 1 μ m. They are highly effective for both coarse and fine particles. Cleaning occurs by passing the dusty gas flow through the filter bag. The main mechanisms in this case are direct interception, inertial collision and diffusion [14].

The advantages of fabric filters are recognized as high capture efficiency in a wide range; flexibility, which is ensured by the availability of various filter materials; wide range of productivity (160–8000000 nm³/h); acceptable operating pressure drops and power requirements; ability to work with various solid materials. The disadvantages of fabric filters include high hydraulic resistance (up to 1800 Pa), large area occupied by them, the ability to work only with dry flue gases, which involves their preliminary drying, fragility and operation in a certain temperature range [14]. The service life of fabric filters depends on the type of material from which they are made, the conditions and intensity of operation, and the cleaning method.

The use of high-efficiency air filters from a textile-organic metal base [15, 16] will ensure microporosity of such a material and strong adhesion. As a metal-organic framework, materials with metal centers of Al(III) [15] and Zr(IV) [15, 16] with functional groups [16] can be used. Such air filters can be reused, which is positive from the point of view of operating costs. The use of nanofibers with some additives for the manufacture of filter materials can be functional for the simultaneous removal of gaseous pollutants with simultaneous filtration of particulate matter [17].



The cyclone, the scheme of which is shown in Fig. 6, is one of the common methods of cleaning air from particulate matter (aspiration). In the heat power industry of Ukraine, battery cyclones are used in the second stage boilers of the Cherkasy CHPP. Due to the action of centrifugal forces in cyclones, particulate matter is cleaned from the air. The advantages also include high productivity and the possibility of using them to clean

high-temperature gas mixtures [18]. However, they are characterized by high hydraulic resistance (up to 1200 Pa), complete inefficiency in cleaning particles with a size of $<2.5 \mu m$ and a significant decrease in efficiency due to increased flue gas humidity [18]. Based on the above, we can conclude that cyclones can be used as devices for primary dust cleaning of flue gases.

Electrostatic precipitators are devices for dust removal from flue gases, the main principle of which is to charge particulate matter in the gas stream and attract it to the precipitation surface of the opposite polarity [14, 19].



Electrostatic deposition consists of three stages (Fig. 7):

- charging of the collected particles using a high-voltage corona electric discharge;
- collection of particles on the surface of an oppositely charged collection surface;
- cleaning of the deposition surface.

The advantages of using electrostatic precipitators are high efficiency of particulate matter removal with the ability to clean large volumes of flue gases in a short period of time, low pressure drop and low operating and maintenance costs [19]. The disadvantages of using electrostatic precipitators are high capital costs, the significant size of this type of equipment and the need to comply with strict operating conditions.

Dry electrostatic precipitators [20] are cleaned by shaking, which provides secondary removal, while wet electrostatic precipitators are cleaned by water. Particles larger than 1 μ m are negatively charged by capturing negatively charged gas ions generated by corona charging. This is called direct (impact) charging [19]. Most particles smaller than 0.3 μ m are charged with these ions as they pass by. This phenomenon is called "diffusion charging" and is associated with Brownian motion, which in turn is associated with thermal effects [19]. Particles with a size of 0.3–1 μ m combine these two charging mechanisms. However, particles with a diameter of 0.3–0.6 μ m are difficult to charge due to the transition between these two mechanisms [19], so in this diameter range there is a decrease in cleaning efficiency, as can be seen in Fig. 5.

The rate of deposition of particulate matter in an electrostatic precipitator is called the "drift rate", which depends on the electric force acting on the particulate matter and on the resistance force that increases as the charged particle moves towards the precipitating electrode, perpendicular to the main gas flow. The drift rate by the impact mechanism is determined by the following equation [14]:

$$w = \frac{2.95 \cdot 10^{-12} \cdot p \cdot E_c \cdot E_p \cdot d_p}{\mu_g} \cdot K_c$$

where p is the relative permittivity; E_c is the charging field voltage, V/m; E_p is the precipitating field voltage, V/m; d_p is the particle diameter, μ m; K_c is the Cunningham correction factor for particles $\leq 5 \mu$ m; μ_g is the dynamic viscosity of gas, Pa·s.

An estimate of the capture efficiency of dust of a certain size in an electrostatic precipitator can be obtained using the Matts-Öhnfeldt equation, which is a modification of the Deutsch-Anderson equation [19]

$$\eta = 1 - e^{-w_k \left(\frac{A}{Q}\right)^k}$$

where η is the efficiency of the fractional collection; w_k is the drift speed, m/s; k is a constant having a value of 0.4–0.6; A is the total collection area, m²; Q is the volumetric air flow rate, m³/s.

Compared to dry ones, wet electrostatic precipitators have the following advantages: due to the use of water to clean the precipitation electrodes, they do not have problems with discharge, in addition, they eliminate re-emission; there is the possibility of using a higher charge power, and the temperature of the flue gases is reduced due to high humidity, which leads to greater removal efficiency; they are easily integrated with other

desulfurization technologies to eliminate other pollutants [19]. However, there are disadvantages, namely, wet electrostatic precipitators complicate the particulate matter control system, increase water consumption; due to the possibility of H₂SO₄ formation, wet electrostatic precipitators must be made of more expensive corrosion-resistant materials, and they require wastewater desulfurization or disposal systems, which increases capital and operating costs, increases energy consumption, and may require flue gas heating [19].

Wet dust collection plants are the most common dust removal methods at CHPPs in Ukraine. Their advantages are low cost, ease



of operation and the possibility of combined flue gas cleaning using aqueous solutions with alkaline properties. However, the formation of a large amount of wastewater and corrosive effects are negative aspects when using wet dust cleaning methods [21]. The simplest wet cleaning method is a countercurrent irrigation scrubber, the scheme of which is shown in Fig. 9. Its principle of operation is to wet and capture particulate matter of flue gases with water droplets. The Venturi tube (Fig. 10) in wet dust cleaning methods is used to capture small particles (<2 μ m) and works according to the inertial particle collection mechanism. Due to a significant increase in the flow rate, the Venturi tube is a fairly productive device for flue gas cleaning.

The efficiency of trapping particulate matter by droplets in a Venturi tube is described by the following formula [22]:

$$\eta = 1 - Pt = 1 - \exp\left(-\frac{3}{2}\frac{q_w}{D_d \cdot \rho_l} \cdot \int_0^L \eta_{\Sigma}(x) \cdot \left|\frac{v_p(x) - v_d(x)}{v_d(x)}\right| dx\right),$$

where Pt is the fraction of untrapped particulate matter (penetration); q_w is the water spray density in a dusty gas stream, kg/m³; η_{Σ} is the deposition coefficient of particulate matter with diameter d_p on spherical drops with diameter D_d ; ρ_l is the density of water, kg/m³; v_p is the velocity of ash particles with diameter d_p , m/s; v_d is the droplet speed diameter D_d , m/s; L is the length of the Venturi tube from the nozzle to the tube outlet, m; x is the coordinate along the axis of the Venturi tube [22].

The effect of parameters on the capture of particulate matter using a wet scrubber has been studied in some papers [23, 24]. It was shown that increase in the flue gas velocity reduces the cleaning efficiency due to a decrease in contact time, while increase in the humidity improves the efficiency due to the effect on diffusion. It was also shown that salt solutions are more effective due to the diffusion-phoretic force created by salt solutions [23].

The capital costs and the costs of operation and maintenance of different types of dust cleaning plants [14] are shown in Table 6.

The capital costs for a boiler with a flue gas flow of 200 thousand nm^3/h when installing a dry plate electrostatic precipitator are about 2.7 million dollars on average; a bag filter with pulse cleaning – 2 million dollars; a wet scrubber with a Venturi tube – 1.46 million dollars.

From the point of view of capital and operating costs, the use of a wet scrubber with a Venturi tube is an inexpensive solution that can provide high efficiency in cleaning flue gases from particulate matter. However, if an aqueous solution of ammonia is used instead of water, this will simultaneously allow for the effective cleaning of flue gases from sulfur dioxide in one device.

Cleaning equipment	Capital costs, \$/(nm ³ /h)	Operating costs, \$/(nm ³ /h)
Cyclone	1.37 - 2.18	0.43 - 5,3
Dry tubular electrostatic precipitators	12.46 - 77.88	2.5 - 5.6
Dry plate electrostatic precipitators	6.23 - 20.56	1.87 - 21.8
Wet tubular electrostatic precipitators	24.92 - 124.61	3.74 - 6.23
Wet plate electrostatic precipitators	12.46 - 24.92	3.11 - 24.92
Fabric filter with pulse cleaning	3.74 - 16.2	3.11 - 14.95
Fabric filter with air reverse	5.6 - 52.96	3.74 - 16.82
Fabric filter with a successful mechanism	4.98-44.86	2.5 - 14.95
Venturi scrubber	1.56 - 13.08	2.74 - 74.77

Table 6. Capital and operating costs of dust cleaning plants [14]

Sulfur dioxide: impact and methods for reducing emissions

Sulfur dioxide is a highly toxic gas formed during the oxidation of sulfur. When released into the atmosphere, SO₂ can lead to the formation of acid rain, which negatively affects vegetation [25], soil [26] and water bodies. From the point of view of human health, SO₂ can cause respiratory tract irritation, coughing, bronchitis and exacerbation of asthma symptoms [11, 27]. A diagram of industrial flue gas desulfurization methods [14] is shown in Fig. 11. They can be conditionally divided into dry, semi-dry and wet ones.

Dry desulfurization methods

The dry desulfurization method consists in the adsorption of sulfur dioxide on the surface of a powdered sorbent. Natural reagents, such as limestone or dolomite, are some of the cheapest and most easily available reagents for this purpose [28]. Slaked lime $Ca(OH)_2$ can also be used for dry desulfurization.

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The powdered reagent is injected directly into the boiler fuel at high temperature (800– 1000°C) with its calcination for reaction with SO₂, as a result of which the sorbents decompose and become porous with a large surface area, with the formation of anhydrous calcium sulfate or anhydrite CaSO₄ [29]. The corresponding reaction equations are given below [14, 29, 30]:

 $\begin{array}{l} CaCO_{3(s)} \xleftarrow{\text{Heat}} CaO_{(s)} + CO_{2(g)};\\ Ca(OH)_{2(s)} \xleftarrow{\text{Heat}} CaO_{(s)} + H_2O_{(v)};\\ CaO_{(s)} + SO_{2(g)} + \frac{1}{2}O_{2(g)} = CaSO_{4(s)}. \end{array}$

However, this method is not sufficiently efficient – at a Ca/S molar ratio of about 4–5, a purification efficiency of 50% can be achieved [30]. By recycling the process, the removal efficiency



can be increased to 70–80% [30], which also does not meet current emission standards [14, 30]. Another significant disadvantage is the formation of anhydrous calcium sulfate, which has no commercial potential without the possibility of reusing it.

The use of sodium bicarbonate as a sorbent [31] is considered as an alternative to lime compounds, which has a fairly high purification efficiency from sulfur dioxide with the ability to simultaneously reduce nitrogen oxide emissions [31]. The reactions are given below [31]

$$2NaHCO_3 \rightarrow Na_2CO_3 + H_2O + CO_2;$$

 $Na_2CO_3 + SO_2 \rightarrow Na_2SO_3 + CO_2;$

$$Na_2CO_3 + SO_2 + \frac{1}{2}O_2 \rightarrow Na_2SO_4 + CO_2.$$

However, its use is limited by high operating costs due to the significant cost of this type of sorbent.

Semi-dry desulfurization methods

The semi-dry desulfurization method, the scheme of which is shown in Fig. 12 [29], is a combination of dry and wet desulfurization methods. The principle of operation of the semi-dry method is to spray finely dispersed drops of solutions or suspensions of alkaline compounds to absorb acid gases. Water moistens the flue gases, which improves the removal of both sulfur dioxide and particulate matter [31]. The water in which the reagent was dissolved evaporates, and as a result the product is removed from the technological process in a dry form in particulate matter capture devices.

This technology is characterized by low capital costs and the absence of wastewater generation, which makes it quite attractive for the modernization of boilers at existing thermal power plants [29].



The simplest method for semi-dry flue gas desulfurization is to add solutions or suspensions to the gas duct channel after the air heater or after the standard dust collector with a temperature in the range of 140-160 °C. However, there is also a drawback - the need to install an additional reactor-absorber due to insufficient reaction time in the gas duct.

The most common in the thermal power industry are semi-dry desulfurization plants using lime suspension in a molar ratio of Ca/S=1.5, which provide about 95% sulfur dioxide binding efficiency. The disadvantages of semi-dry lime technologies are the production of a non-commercial product (a mixture of calcium sulfite, calcium sulfate and unused lime), the need to recycle the reaction products to increase the efficiency of the sorbent for removing sulfur dioxide. An alternative is the use of ammonia NH_3 in semi-dry

desulfurization systems, which will help to get rid of some of the disadvantages of calcium technologies, while reducing capital costs by 23% and operating costs by 25% [32]. As a product of the interaction of ammonia solution droplets with flue gases containing SO₂, in the semi-dry desulfurization method, dry ammonium sulfate $(NH_4)_2SO_4$, which is recognized as a mineral fertilizer, can be obtained [33]. The high solubility of ammonia and its reactivity with the ability to react with humidified flue gases in the gas phase [34] will ensure high efficiency of sulfur removal (up to 98%) in smaller units, which can reduce capital costs.

The disadvantage of the method is the toxicity of ammonia [35], so the use of such a reagent involves strict emission control to prevent ammonia from "leaking" into the air, because the stoichiometric ratio of NH_3/SO_2 must always be less than 2.

Wet desulfurization methods

Wet methods are based on the phenomenon of absorption - the transfer of a substance from the gas phase to the liquid one – with subsequent neutralization of pollutants by alkaline sorbents. Various compounds with alkaline properties are used as sorbents, namely: calcium compounds – limestone or lime [36–38]; sea water [31, 39]; ammonium compounds [31, 40, 41]. The reactions occur mainly in the liquid phase, but, as in the case of ammonia, the reactions can also take place in a humidified gas stream. Due to its reliability and high cleaning efficiency, the wet flue gas desulfurization system is the most common at coal-fired power plants [29].

The industrial method of desulfurization using calcium compounds, the scheme of which is shown in Fig. 13 [42], follows the reactions described below [14]

 $SO_{2} + H_{2}O = H_{2}SO_{3};$ $H_{2}SO_{3} = H^{+} + HSO_{3}^{-};$ $Caco_{3} + H^{+} = Ca^{2+} + HCO_{3}^{-};$ $HCO_{3}^{-} + H^{+} = H_{2}O + CO_{2};$ $HSO_{3}^{-} + \frac{1}{2}O_{2} = SO_{4}^{2-} + H^{+};$

$$Ca^{2+} + SO_4^{2-} + 2 H_2O = Caso_4 \times 2H_2O.$$

The disadvantages of this technology are the high water consumption $(15-25 \text{ kg/m}^3 \text{ of flue}$ gases) [43], which is necessary for the recirculation of the limestone suspension, which has a very low solubility, high energy consumption, the desulfurization of a significant amount of wastewater, and the formation of large volumes of dihydrate gypsum.

The use of seawater for flue gas purification is due to the alkalinity of seawater, the pH of which varies between 7.6 and 8.4 due to the presence of carbonate and hydrocarbonate ions [31]. After the reaction, sulfate ions are formed, which are natural components of seawater, the discharge of which back into the sea does not significantly affect the environment. The main reactions are given below [39]

$$\begin{split} & SO_{2(g)} + H_2O \leftrightarrow SO_{2(aq)} \cdot H_2O; \\ & SO_{2(aq)} \cdot H_2O + H_2O \leftrightarrow HSO_3^- + H_3O^+; \\ & HSO_3^- + H_2O \leftrightarrow SO_3^{2-} + H_3O^+; \\ & HCO_3^- + H_3O^+ \leftrightarrow CO_{2(aq)} + 2H_2O; \\ & CO_{2(g)} + H_2O \leftrightarrow CO_{2(aq)} \cdot H_2O; \\ & HCO_3^- + H_2O \leftrightarrow CO_3^{2-} + H_3O^+. \end{split}$$



Fig. 13. Typical scheme of limestone wet desulfurization method with forced oxidation [42]



The main advantages of the wet desulfurization process using seawater include the absence of the need for chemical reagents due to their presence in seawater; the absence of by-products that need to be

treated or disposed of; the simplicity of the design and operation of the plant and the possibility of achieving high SO_2 removal efficiencies (up to 99%) [31]. The process is limited to coastal power plants and low sulfur content in the fuel; otherwise, additives (sodium hydroxide or magnesium hydroxide) must be used to neutralize the absorber wastewater before it is discharged into the sea [14].

The use of an aqueous ammonia solution in the wet desulfurization method, the scheme of which is shown in Fig. 14, is characterized by a high solubility of the alkaline reagent, which helps to reduce the amount of wastewater and the size of the apparatus. In addition, the reaction of ammonium with humidified flue gases in the gas phase [34] increases the efficiency of sulfur removal.

According to the scheme in Fig. 14 ammonia, flue gases, air for oxidation and process water are fed into the absorber. The aqueous ammonia solution is fed into the absorber in the form of finely dispersed droplets using several levels of spray nozzles, which will increase the contact area with the flue gases [44]. The main chemical reactions using aqueous ammonia solution are given below [31]:

$$NH_3 + H_2O = NH_4OH;$$

 $SO_2 + H_2O = H_2SO_3$.

$$H_2SO_3 + NH_4OH = NH_4HSO_3 + H_2O;$$

$NH_4HSO_3 + NH_4OH = (NH_4)_2SO_3 + H_2O.$

Oxygen blown into the lower part of the absorber oxidizes ammonium sulfite to sulfate according to the following equation:

$$2(NH_4)_2SO_3 + O_2 = 2(NH_4)_2SO_4.$$

A study of the influence of various factors on the efficiency of flue gas cleaning from sulfur dioxide with ammonium reagents is described in [45]. It was determined that increasing the pH value in the absorber to 5.5 leads to a rapid increase in efficiency. With a further increase in pH, the desulfurization efficiency increases, but more slowly [45]. An increase in the liquid/gas ratio has a positive effect on the cleaning efficiency and reaches a constant level at a value of 3 l/m^3 , which is explained by an increase in the contact surface [45].

The use of ammonia is limited by its toxicity [35], which craves increased requirements for the storage and transportation of the ammonia solution. The use of urea as an alternative source of ammonia solves the problem of storage and transportation, since urea is non-toxic and stable. Hydrolysis of an aqueous urea solution is an effective source of ammonia generation, which can provide high efficiency in cleaning flue gases from sulfur dioxide, but the use of this method is limited to the formation of ammonia as a result of thermal hydrolysis at a temperature of 140 °C and a pressure of 0.4 MPa for 4 hours [46].

Desulfurization method	Wet Ammonia	Seawater Desulfurization	Wet soda	Wet limestone	Semi-dry	
Features	High-value by- product; low operating costs	Low capital cost; simple operation; proximity to the sea	Low capital cost; easy operation	Highly efficient spray zone; inexpensive reagent	Low capital costs; dry by-product; no liquid waste	
Reagent	Ammonia	Seawater	Soda	Limestone	Lime	
By-product	Ammonium sulfate, fertilizer	Purified seawater	Sodium sulfate	Commercial gypsum	Landfill	
SO ₂ concentration at inlet	High	Low / medium	High	High	Low / medium	
SO ₂ removal, %	> 98	> 95	> 98	> 98	90 - 95	
Capital cost, \$/(nm ³ /h)	22.2-38	9.5 - 15.85	6.3 – 12.7	15.85 - 28.5	9.5 - 15.85	
Reagent cost, \$/(nm ³ /h)	50.7 - 66.5	0	63.4 - 82.4	9.5 - 15.85	38-47.55	
By-product cost, $\$/ton of SO_2$ 95.1 – 159.5 (sale) removed		0	_	7.6 – 12.7 (disposal) 9.5 (sale)	7.6 – 12.7	

Table 7. Characteristics of industrial flue gas desulfurization methods [14]

The main characteristics of modern desulfurization methods [14] are shown in Table 7. For a boiler with a flow rate of 200 thousand m^3/h , the capital costs when using the wet ammonium desulfurization meth-

od average \$6 million; wet soda desulfurization - \$1.9 million; wet limestone desulfurization - \$4.4 million. However, due to the sale of the by-product (ammonium sulfate) in the wet ammonium desulfurization technology, operating costs can be almost completely compensated, unlike other technologies. The technology of wet soda desulfurization has not received practical distribution due to the high cost of soda reagent.

The use of ammonium reagents is a promising direction in flue gas purification technologies. The use of a semi-dry method involves the installation of two dust cleaning systems: for removing solid ash particles and for collecting a useful dry product, which increases capital costs. Since all ammonium salts are water-soluble, ammonium desulfurization can be used in wet dust cleaning devices with subsequent separation of particulate matter from the solution. Also, the advantage of the wet method is the availability of appropriate cleaning equipment, which is mostly used in existing CHP plants with the possibility of its modernization to meet the necessary requirements. Another advantage is the possibility of complex flue gas purification using ammonium reagents in one device.

The impact of nitrogen oxides and methods of their control

Nitrogen oxides NOx (a mixture of nitrogen monoxide NO and nitrogen dioxide NO₂) are formed during high-temperature fuel combustion [12]. Nitrogen oxides are components of acid rain, which negatively affect ecosystems. Within the boiler, almost 95% of nitrogen oxides are nitric oxide. In atmospheric air, the content of nitrogen dioxide is about four times higher than that of nitric oxide. Nitrogen dioxide is a toxic gas that can cause various diseases, such as respiratory and cardiovascular ones [8, 47]. It is also a component for the formation of ozone, which can reduce air quality, lead to climate change and the deterioration of natural ecosystems [11, 48].

NOx is formed by three main mechanisms [49–51]:

- thermal NOx is formed as a result of the reaction between nitrogen and oxygen in the air;

- fuel NOx is formed as a result of the oxidation of nitrogen contained in the fuel;

- fast NOx is formed as a result of the combustion of molecular nitrogen in a burner flame in the presence of hydrocarbons.

Methods for controlling nitrogen oxide emissions are divided into primary and secondary ones. The methods are summarized in the diagram shown in Fig. 15 [52].



Primary (regime-technological) methods for reducing NOx emissions are focused on reducing the formation of nitrogen oxides, namely by reducing the temperature in the combustion zone, controlling the amount of excess air, reducing the oxygen concentration in the ignition zone, reducing the time the fuel stays in the zone with the maximum temperature, using low-emission burners, multi-stage fuel and air supply, etc. [49, 50]. Primary methods are an important tool in the fight against nitrogen oxides, but they have low efficiency, their use is possible only in combination with secondary denitrification methods. In existing boilers, the use of primary methods often leads to a deterioration in fuel combustion conditions, an increase in chemical and mechanical unburnt fuel.

The most effective methods for cleaning flue gases from nitrogen oxides are secondary methods, i.e. methods for reducing the content of formed nitrogen oxides. Among them, attention is paid to two main methods – selective non-catalytic reduction (SNCR) and selective catalytic reduction (SCR). The main reagent for the reduction is ammonium compounds – ammonia NH_3 and urea $(NH_2)_2CO$.

Selective non-catalytic reduction involves the injection of nitrogen-containing chemicals into the upper part of the fuel within a certain temperature window without the use of a catalyst [14]. The temperature range of this reaction is 800–1150 °C [53]. Although in some sources [54, 55] the value of the temperature range is slightly different. SNCR does not require catalysts, which frees the plant from all technical problems associated with the introduction of additional catalytic desulfurization of flue gases. The main reactions using ammonia and urea, respectively [14, 56]

$4NO + 4 NH_3 + O_2 = 4N_2 + 6H_2O;$

$4NO + 2(NH_2)_2CO + O_2 = 4N_2 + 2CO_2 + 6H_2O.$

Maintaining the temperature range is extremely important in the SNCR method. If the temperature is lower, the reduction will not occur, which will lead to ammonia breakthrough. At the same time, too high temperature will lead to oxidation of the reducing agent with the threat of increased NOx emissions. The dependence of the formation of reagents in the SNCR process on temperature is shown in Fig. 16 [57]. Optimization of SNCR raises at least 4 main questions [55]:

- control of gas enthalpy - the need for optimized cooling of flue gases before introducing the reducing agent;

- maintaining the optimal stoichiometric ratio to prevent ammonia from being released into the atmosphere;

– control of the mixing process;

- ensuring sufficient stay time.

Selective catalytic reduction has an efficiency of up to 90% and uses catalysts to accelerate the reaction between ammonia and nitrogen oxides at lower temperatures [14]. Ammonia is chemisorbed on the active sites of the catalyst, NOx reacts with the adsorbed ammonia to form molecular nitrogen and water vapor according to the following reactions: [14, 58, 59]:

$$4NO + 4NH_3 + O_2 = 4N_2 + 6H_2O;$$

 $2NO_2 + 4NH_3 + O_2 = 3N_2 + 6H_2O.$

The catalyst can be precious metals, such as platinum [59], base metals – compounds of vanadium, tungsten, titanium [59, 60], zeolites [14, 61]. SCR systems are usually used in large boilers. There are three main configurations of selective catalytic reduction for coal-fired boilers, the schemes of which are shown in Fig. 17 [14]:

- high dust, where the SCR reactor is placed before the particulate matter removal device between the economizer and the air preheater. In this configuration, the catalyst is exposed to fly ash and chemical compounds present in the flue gases, which can potentially abrasively destroy the catalyst [14];

- low-dust, where the SCR reactor is located after the particulate matter removal device. This configuration reduces catalyst degradation due to fly ash erosion. However, this configuration requires an expensive high-temperature electrostatic precipitator or a flue gas heating system to maintain the optimum operating temperature [14];

- tailings, when the SCR reactor is installed downstream of a flue gas desulfurization (FGD) unit. It can be used primarily in liquid slag removal boilers, as well as in retrofitted plants with limited space and insufficient downtime for retrofitting. However, this configuration is generally more expensive to operate than the high-dust



Fig. 16. Reduction of NOx emissions as a function of temperature [57]: A – optimum temperature for the SNCR process (low ammonia slippage); B – optimum temperature for the SN+CR+SCR process (high ammonia slippage)



configuration due to the flue gas preheating requirements. The advantages of this configuration are a longer catalyst life and the use of more active catalyst formulations to reduce the overall cost of the catalyst, especially when burning fuels containing components that quickly deactivate the catalyst [14].

The capital and operating costs of the main denitrification methods are shown in Table 8 [14]. As can be seen from the table, the capital costs of the SCR method are 6 times higher than the corresponding costs of the SNCR, and the operating costs are 3 times higher. A significant disadvantage of SCR is the use of expensive catalysts, which increases capital investment. Catalysts are also vulnerable to other pollutants, which can reduce their efficiency. Therefore, in municipal CHPPs, it is more appropriate to use the SNCR method, which is also effective.

	Variable			Power, MW							
Tashua	opera-	100		300		500		700		1000	
logy	ting	Capital	Operating	Capital	Operating	Capital	Operating	Capital	Operating	Capital	Operating
logy	costs,	costs,	costs,	costs,	costs,	costs,	costs,	costs,	costs,	costs,	costs,
	\$/MWh	\$/kW	\$/kW·year	\$/kW	\$/kW·year	\$/kW	\$/kW·year	\$/kW	\$/kW·year	\$/kW	\$/kW·year
SCR	1.23	321	1.76	263	0.76	243	0.64	232	0.58	222	0.53
	1.32	349	1.86	287	0.81	266	0.69	255	0.63	244	0.57
	1.41	377	1.96	311	0.87	289	0.73	277	0.67	265	0.62
SNCR	1.04	55	0.48	30	0.26	22	0.20	18	0.16	15	0.13
	1.15	56	0.50	30	0.27	23	0.20	19	0.17	15	0.14
	1.27	57	0.51	31	0.27	23	0.21	19	0.17	15	0.14

Table 8. Capital and operating costs of denitrification methods

Conclusions

1. Existing technologies for cleaning flue gases from pollutants, in particular, particulate matter, sulfur dioxide and nitrogen oxides, for the modernization of existing boiler units of CHPPs operating on solid fuel in Ukraine are considered in the paper. The target group of solid fuel boiler units of CHPPs must undergo environmental modernization to comply with European environmental directives.

2. For the removal of particulate matter, the best equipment in terms of compliance with modern environmental requirements is a bag filter, an electrostatic precipitator and a wet scrubber with a Venturi tube with significant irrigation. The choice of a wet scrubber with a Venturi tube using an aqueous ammonia solution is a rational solution due to the possibility of simultaneous removal of sulfur dioxide.

3. The use of ammonium reagents for wet removal of sulfur dioxide is a highly effective method with the production of a useful product - ammonium sulfate. The semi-dry ammonium method of sulfur removal is also effective, but a significant drawback is the need to install additional purification equipment to particulate matter from the product. The best method for complying with modern environmental directives with the possibility of upgrading the existing purification equipment of CHPP should be recognized as the wet method of sulfur removal.

4. The selective non-catalytic reduction method can be easily integrated into existing CHPP boilers without significant changes to the boiler design, therefore it is a more rational solution, especially for small combustion plants.

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Застосування технологій очищення димових газів для твердопаливних котлів теплоелектроцентралей

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У статті проаналізовано шляхи задоволення вимог сучасного екологічного законодавства України й Европейського Союзу щодо обмеження викидів забруднюючих речовин від великих і середніх спалювальних установок стосовно твердопаливних парових котлів комунальних і промислових теплоелектроцентралей (ТЕЦ) України. Розглянуто екологічні вимоги й технології очищення димових газів твердопаливних котлів від основних забруднюючих речовин, а саме твердих частинок, діоксиду сірки й оксидів азоту, проведено аналіз ефективності, переваг і обмежень для впровадження иих технологій на теплоелектроиентралі. Акиентовано на тому, що існуючий стан газоочисного обладнання, введеного в експлуатацію більше п'ятдесяти років тому, не відповідає чинним екологічним вимогам, а отже, нагальним є питання реконструкції та модернізації наявних і спорудження нових газоочисних установок. Доведено, що застосування тканинних, електростатичних фільтрів і мокрих скруберів із трубою Вентурі дозволить задовольнити вимоги європейських директив щодо граничних значень викиду пилу, а найбільш раціональним рішенням буде використання наявних мокрих скруберів із трубою Вентурі, якими оснащена переважна більшість ТЕЦ, шляхом істотного збільшення питомої витрати рідини на зрошення. Для уловлення газоподібних забруднюючих речовин перспективним напрямком є використання амонійних реагентів для високоефективної десульфуризації і отримання як продукту сіркоочищення сульфату амонію, що виступає мінеральним добривом, та відновлення оксидів азоту до молекулярного азоту. Використання водного розчину амоніаку в мокрому скрубері з трубою Вентурі дасть змогу одночасно уловлювати в одному пристрої летку золу і діоксид сірки. Для зниження викидів оксидів азоту на котлах ТЕЦ доцільним із точки зору інвестиційних затрат і просторових умов визнається застосування методу селективного некаталітичного відновлення.

Ключові слова: паровий котел, димові гази, труба Вентурі, тверді частинки, діоксид сірки, амоніак, оксиди азоту.

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