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COMPREHENSIVE PURIFICATION OF GAS EMISSIONS FROM HEAT POWER PLANTS

The article presents a comprehensive scientific approach to solving a pressing environmental problem – cleaning gas emissions of thermal power plants from the most common pollutants: dust, sulfur dioxide, carbon monoxide and nitrogen oxides. The relevance of the study is due to the growth in the volume of emissions of harmful substances and their critical impact on human health and the state of the environment. The proposed technology for cleaning gas emissions consists of four consecutive stages. Dust is cleaned using a cyclone-rotary dust collector, which allows capturing fine dust with an efficiency of up to 99%. For the removal of sulfur dioxide, an absorption method of neutralization is proposed, in which acidic flue gases are cleaned using alkaline wastewater from industrial enterprises. For the removal of carbon monoxide, a catalytic method is proposed. A catalyst composition based on zinc, copper, chromium and aluminum oxides has been developed. Unlike platinum analogues, it can operate in dusty gases and has low hydrodynamic resistance due to the application of a paste-like mass to the inner surfaces of the reactor tubes. The final stage of the complex purification system is the thermal neutralization of nitrogen oxides using an arc plasma torch. The processes of high-temperature decomposition have been studied and it has been established that the introduction of reducing agents, such as methane or ammonia, into the reactor allows to shift the reaction equilibrium towards the formation of molecular nitrogen and oxygen. The proposed step-by-step purification system allows to reduce the concentration of harmful compounds in the purified gases to the maximum permissible values.

Key words: gas emissions, purification system, heat power, dust, sulfur dioxide, carbon monoxide, nitrogen oxides.

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КОМПЛЕКСНЕ ОЧИЩЕННЯ ГАЗОВИХ ВИКИДІВ ТЕПЛОЕНЕРГЕТИЧНИХ ПІДПРИЄМСТВ

У статті представлено комплексний науковий підхід до вирішення актуальної екологічної проблеми – очищення газових викидів теплоенергетичних підприємств від найбільш поширених забруднювачів: пилу, двооксиду сульфуру, монооксиду карбону та оксидів нітрогену. Актуальність дослідження зумовлена зростанням обсягів викидів шкідливих речовин та їхнім критичним впливом на здоров'я людини й стан навколишнього середовища. Запропонована технологія очищення газових викидів складається з чотирьох послідовних етапів. Очищення від пилу здійснюється циклонно-ротаційним пиловловлювачем, що дозволяє вловлювати дрібнодисперсний пил з ефективністю до 99%. Для видалення двооксиду сульфуру запропоновано абсорбційний метод нейтралізації, в якому кислі димові гази очищуються за допомогою лужних стічних вод промислових підприємств. Для видалення монооксиду карбону запропоновано каталітичний метод. Розроблено склад каталізатора на основі оксидів цинку, міді, хрому та алюмінію. На відміну від платинових аналогів, він може працювати у запиленних газах і має низький гідродинамічний опір завдяки нанесенню пастоподібної маси на внутрішні поверхні труб реактора. Завершальним етапом комплексної системи очищення є термічне знешкодження оксидів нітрогену за допомогою дугового плазмотрона. Досліджено процеси високотемпературного розкладання та встановлено, що введення в реактор відновників, таких, як метан або амоніак дозволяє змістити рівновагу реакції у бік утворення молекулярного азоту та кисню. Запропонована поетапна система очищення дозволяє зменшити концентрації шкідливих сполук в очищених газах до гранично допустимих значень.

Ключові слова: газові викиди, система очищення, теплоенергетика, пил, двооксид сульфуру, монооксид карбону, оксиди нітрогену.

Problem Statement

At the current stage of energy development, the database of maximum permissible concentrations (MPC) contains more than two thousand harmful substances that pollute the atmosphere. However, the overwhelming majority of emissions from heat power enterprises are formed by only four components: dust, sulfur dioxide (SO₂), carbon monoxide (CO), and nitrogen oxides (N_xO_y) [1, 2]. These compounds are formed during the combustion of any type of fuel – solid, liquid, or gaseous – and their ratio depends on the characteristics of the energy carrier [3]. In 2024, the volume of pollutants from stationary sources amounted to 8.5 thousand tons, which exceeds the indicators of 2023 (7.4 thousand tons) by 14.9% [4, 5]. A person passes about 20 m³ of air through the lungs daily. Even small doses of pollutants have a cumulative negative effect. In particular, carbon monoxide interacts with blood hemoglobin 300 times faster than oxygen, and its concentration of 0.32% is lethal. Sulfur dioxide and nitrogen oxides are the main factors in the formation of acid rain, which acidify soils, destroy building structures, and accelerate metal corrosion. Dust, in addition to direct harm, acts as an adsorbent that transports other toxic substances over long distances. Existing gas purification methods are often highly specialized or too expensive. For example, catalytic post-oxidation using platinum catalysts is economically

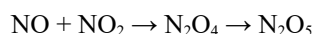
impractical for large gas volumes, and efficient electrostatic precipitators require significant electricity consumption. Thus, there arises an urgent scientific and practical task of developing a comprehensive purification system capable of simultaneously removing solid and gaseous impurities while ensuring compliance with MPC standards with minimal operating costs.

Analysis of Recent Research and Publications

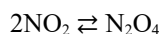
Modern methods of purification of industrial emissions are based on the use of gravitational, inertial, centrifugal, and electrical forces [6]. Depending on the nature of the forces used in dust-collecting apparatuses, dry and wet purification methods are applied [7]. Dry purification methods are carried out in dust-settling chambers, cyclones, porous and electric filters. Wet purification methods are implemented in apparatuses irrigated with liquid. The apparatuses may be used independently or in combination with each other. They may form a single-, two-, or three-stage purification system depending on the required gas purification quality. For any dust-cleaning equipment, the main characteristics are primarily: collection efficiency (%); velocity of the gas-dust flow (m/s); hydraulic resistance (Pa).

Analysis of traditional equipment indicates significant limitations: dust-settling chambers and inertial dust collectors have a low collection efficiency (40-50%) and are used only for preliminary purification. Cyclones effectively capture particles larger than 50 μm; however, fine dust (less than 10 μm) is almost not retained by them. The most advanced apparatuses (electrostatic precipitators and fabric filters) have high operating costs or cannot withstand high temperatures of flue gases from thermal power plants [8, 9]. This determines the necessity of developing combined cyclone-rotary devices (CRD), which combine the advantages of centrifugal separation and wet deposition [10, 11]. For sulfur dioxide removal, absorption methods are promising; however, the use of special reagents increases the process cost [12]. Carbon monoxide purification is traditionally performed by catalytic post-oxidation on platinum catalysts, which is economically inaccessible for large emission volumes [13–16]. In addition, existing packed catalysts have high hydraulic resistance and are quickly poisoned by dust.

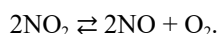
Oxygen compounds of nitrogen may exist in the form of the following oxides: nitrous oxide N₂O, nitric oxide (II) NO, nitrogen dioxide NO₂, dinitrogen trioxide N₂O₃, dinitrogen tetroxide N₂O₄, dinitrogen pentoxide N₂O₅ [17]. The oxidation state of nitrogen is largely determined by temperature. Depending on temperature, certain nitrogen oxides may exist. In the presence of an oxidizing agent (oxygen or ozone) and at low temperature, the oxidation reaction of nitric oxide (II) proceeds spontaneously and irreversibly in the following direction:



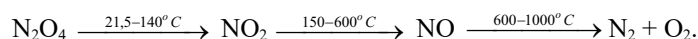
Nitrogen oxide (V) may exist in the solid state at low temperatures. Solid N₂O₅ in its stable form is nitronium nitrate NO₂⁺ NO₃⁻. When heated in the gaseous state, it dissociates into NO₂ and NO₃⁻. NO₃⁻ is easily oxidized by releasing an oxygen atom. Nitrogen oxide (IV) easily polymerizes into a dimer according to the reversible reaction:



The degree of polymerization also depends on temperature. Already at 21.15°C, liquid N₂O₄ dissociates into NO₂ molecules. At temperatures above 140°C, the reaction is completely shifted to the right, and only NO₂ may exist in the gas phase. Upon further heating of nitrogen oxide (IV), it decomposes:



Complete decomposition of nitrogen oxide (IV) into nitric oxide (II) and oxygen occurs at a temperature of 600°C. Nitric oxide (II) is quite stable; however, at temperatures above 1000°C it is in a dissociated state: 2NO ⇌ N₂ + O₂. Thus, the temperature region of existence of various nitrogen oxides in the gas phase may be represented by the following scheme:



Thermal decomposition of NO according to the equation: 2NO → N₂ + O₂ is an energy-intensive process requiring temperatures above 1000°C [18, 19]. The use of plasma technologies makes it possible to achieve the required temperatures; however, the issue of shifting the reaction equilibrium toward molecular nitrogen using available reducing agents remains relevant.

Formulation of the Research Objective

The purpose of the research is the scientific substantiation and development of a comprehensive high-efficiency system for the sequential purification of gas emissions from heat power enterprises from dust, sulfur dioxide, carbon monoxide, and nitrogen oxides by combining mechanical, absorption, catalytic, and thermal methods. To achieve this purpose, the following tasks were set:

1. To investigate the operation of a two-stage cyclone-rotary dust collector (CRDC) capable of operating in both dry and wet modes;
2. To develop a technology for neutralization of acidic flue gases from boiler houses using alkaline industrial wastewater;

3. To obtain a new catalyst for the conversion of CO into CO₂ capable of operating in dusty flue gases and to develop a technology for its application;

4. To investigate the conditions of nitrogen oxides decomposition by the plasma-thermal method and using reducing agents.

Presentation of the Main Research Material

Any processes of purification of industrial gases become complicated if dust is present in them. At the first stage, the contaminated gas must be purified from solid suspended particles. An apparatus has been developed which, unlike known cyclones, can effectively capture fine dust particles up to 4 μm in size. The apparatus was named a cyclone-rotary dust collector (CRDC) because it implements the principle of two-stage separation of dust-gas systems in a centrifugal field.

First, in the first – lower stage, operating according to the cyclone principle; and then in the second – upper stage, which operates according to the principle of a rotary dust collector. The CRDC has a number of advantages compared to existing cyclones:

- acting as a gas blower, it creates a vacuum in the suction nozzle and pressure in the discharge nozzle; the dust collector can operate autonomously (without a fan) and purify gases that do not have initial excess pressure;

- due to the increase in centrifugal force acting on a particle in the rotary part of the dust collector, dust particles smaller than 10 μm are captured;

- by means of the rotor, fine dispersion of the liquid and uniform irrigation of the apparatus walls are achieved, which contributes to increased efficiency of capturing fine and agglomerating dust.

The efficiency of gas purification from dust in cyclone apparatuses depends on the centrifugal force acting on a particle, which in turn depends on the circumferential velocity of the gas flow (Figure 1). An increase in this velocity leads to dust carryover from the apparatus. This disadvantage was eliminated in the developed CRDC design.

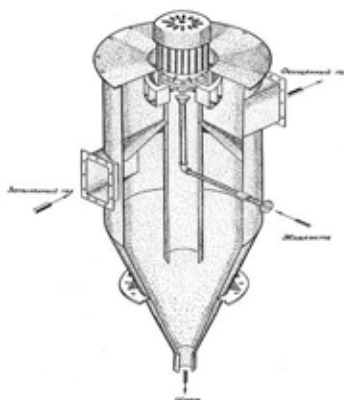


Figure 1. Cyclone-Rotary Dust Collector

The dust collection efficiency in wet mode reaches 99%, and in dry mode 98%. Therefore, irrigation is recommended only when working with dust that tends to agglomerate and may clog the apparatus. Thus, after purification of exhaust gases from solid contaminants, they may be purified from gaseous components.

At the second stage of purification, it is necessary to remove sulfur dioxide from the gas flow. The presence of sulfur dioxide in exhaust gases determines their acidic reaction. At the same time, a significant number of industrial enterprises discharge alkaline wastewater with a pH value of 12–14 into the sewer network. In the work, an absorption method and an apparatus (absorber) for neutralization of acidic flue gases containing sulfur dioxide by means of alkaline wastewater are proposed. The influence of various physicochemical factors on the gas neutralization process was investigated, and the optimal process parameters were determined.

The studies were carried out on a laboratory installation (Figure 2) using the method of mathematical planning of the experiment.

In the work, the method of a full four-factor experiment was applied, which makes it possible to obtain a mathematical description of the investigated process.

The results of laboratory studies were confirmed on an industrial installation (Figure 3), where production acidic gases of the thermal power plant and alkaline wastewater of textile production were used.

The industrial toroidal absorber with a diameter of 3 m and a height of 1.5 m has a gas capacity of 5 thousand m³/h. A closed irrigation cycle was used in the apparatus. Gas supply was carried out directly from the gas duct. Contact between liquid and gas in the absorber was carried out by spraying the liquid through five centrifugal sprayers with intersecting irrigation fields. The studies demonstrated the effectiveness of acidic gas purification by neutralization.

At the third stage, it is proposed to purify gases from carbon monoxide (CO) using the developed catalysts.

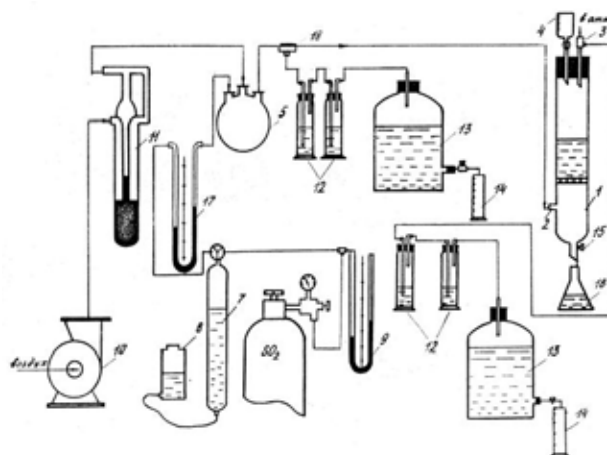


Figure 2. Diagram of the Laboratory Installation

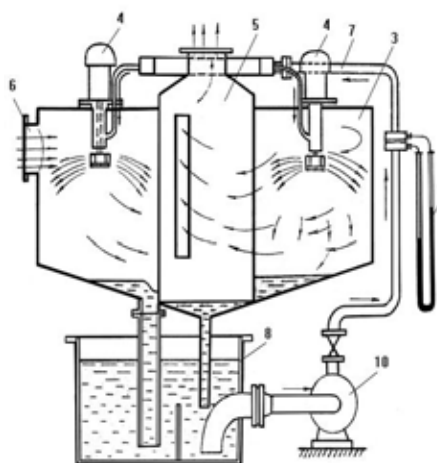


Figure 3. Diagram of the Toroidal Absorber

Despite the high toxicity of carbon monoxide and the large volumes of emissions into the atmosphere, until recently neither in our country nor abroad have effective methods of sanitary purification of gases from this component been implemented. The only exception is automobile exhaust gases, purification of which is carried out by catalytic post-oxidation of carbon monoxide using platinum-vanadium catalysts. However, platinum is a very expensive metal, and its use for purification of large volumes of gases is impossible. Research in this direction led to the development of new types of catalysts that meet the specified requirements. The main task was to determine the optimal catalyst composition that has maximum activity. Catalytic activity – the main characteristic of catalysts – was investigated on a flow installation, the diagram of which is shown in Figure 4. The investigated catalysts were produced from a mixture of catalytically active substances – zinc, copper, chromium oxides, aluminum powder, and cement – which were thoroughly mixed until a homogeneous mass was obtained. Ammonia water was added; at this stage hydrogen was actively released from the obtained mixture, and a foamed porous pumice-like structure was formed.

This catalytic mass in a paste-like state is applied to the inner surface of tubes using a punch. After hardening of the contact mass, it is dried and calcined at a temperature of 350°C. The post-oxidation process is carried out in tubular reactors (Figure 5). In existing apparatuses, gas is forced through a layer of packed granular catalyst. Due to this, such reactors have high hydraulic resistance.

The proposed reactor design consists of tubes mounted on tube sheets and combined into a common casing [20]. A catalyst layer of 3–5 mm is applied to the inner surface of the tubes. Unlike existing apparatuses in which gas is forced through a packed granular catalyst layer, the proposed tubular reactor has very low hydraulic resistance. The advantages of using a paste-like foamed catalyst consist in the fact that gas passing through the tubes coated with catalyst does not encounter significant resistance. The reactor has high throughput capacity and provides a high degree of purification without requiring large operating costs. In addition, the tubular reactor can operate with dusty gases.

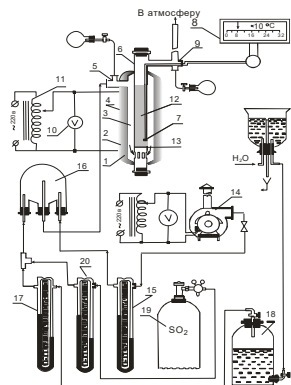


Figure 4. Diagram of the Flow Installation for Studying the Catalytic Activity of Catalysts

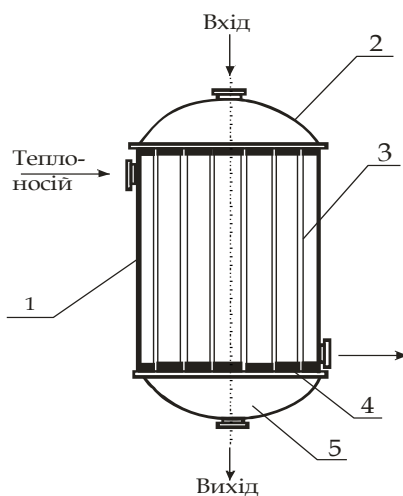


Figure 5. Tubular Reactor: 1 – shell; 2 – cover; 3 – tubes with catalyst; 4 – tube sheets; 5 – bottom.

At the fourth stage of purification, neutralization of nitrogen oxides is carried out. To achieve this purpose, it is advisable to use thermal methods of decomposition of nitrogen oxides (N_xO_y). Under heating conditions, higher nitrogen oxides undergo thermal decomposition with the formation of lower oxides, as well as elemental nitrogen and oxygen. This regularity forms the basis of the developed method of thermal decomposition of nitrogen oxides, which was investigated in the temperature range from 500°C to 5000°C. To achieve high temperatures, a 20 kW arc plasma torch with a tungsten cathode and a copper anode cooled by flowing water was used (Figure 6). Arc stabilization was carried out by a magnetic-vortex method. The plasma torch was positioned vertically with the lower arrangement of the cathode.

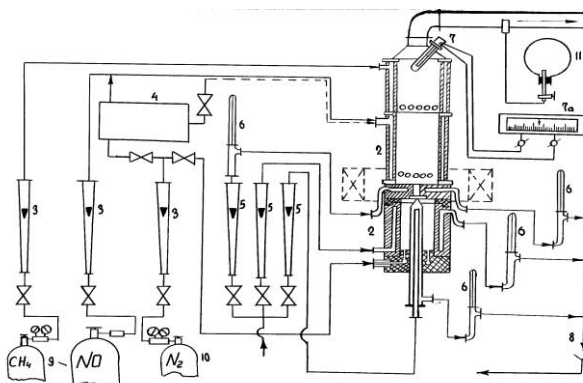


Figure 6. Diagram of the Plasma Torch for Decomposition of Nitrogen Oxides

A reactor was connected to the exhaust nozzle of the plasma torch, which simultaneously performed the function of a heat exchanger for preliminary gas heating. Nitrogen was used as the plasma-forming gas, and liquid dimer of nitrogen oxide (IV) served as the source of nitrogen oxides. The plasma-forming gas with a temperature of 10,000–15,000°C entered the reactor, where it mixed with nitrous gas preheated in the heat exchanger to 600°C. As a result of mixing, the mass-average temperature of the gas mixture in the reactor reached 1000–3500°C. The installation was equipped with a set of measuring instruments for determining gas flow rate, temperature, and other process parameters.

The resulting gas mixture was heated to a temperature of 2000–4000°C, at which decomposition of nitrogen oxides occurred with the formation of molecular nitrogen and oxygen. The degree of decomposition was determined by measuring the concentration of nitrogen oxides at the inlet and outlet of the reactor. The initial concentration of NO was 0.001–10%, and the mass-average temperature in the reactor was 2000–2100°C. With an increase in nitrogen oxide concentration, the degree of decomposition increased. At the same time, the concentration of nitrogen oxides after thermal decomposition remained sufficiently high and reached up to 2%, which is unsatisfactory from the standpoint of sanitary gas purification.

For more complete purification of the gas mixture, the influence of reducing agents on the decomposition process of nitrogen oxides was investigated: gaseous (hydrogen, ammonia, methane); liquid (kerosene, gasoline, fuel oil); solid (coke, coal, graphite). The results showed that in the presence of gaseous and liquid reducing agents, the reaction equilibrium shifts toward the formation of molecular nitrogen and oxygen. The concentration of NO after treatment with reducing agents decreased to 0.1–0.3% at an initial concentration of 2–3%.

Conclusions

1. A holistic four-stage system of comprehensive purification of gas emissions from heat power enterprises has been developed, which allows sequential removal of dust, sulfur dioxide, carbon monoxide, and nitrogen oxides to the level of maximum permissible concentrations.

2. The efficiency of the cyclone-rotary dust collector has been proven. Due to two-stage separation in a centrifugal field, it ensures the capture of fine dust (less than 10 μm) with an efficiency of up to 99% in wet mode and 98% in dry mode.

3. The method of neutralization of acidic flue gases by their absorption with alkaline wastewater of industrial (in particular textile) enterprises with a pH of 12–14 has been substantiated. This makes it possible not only to purify gas from sulfur dioxide but also to rationally utilize aggressive industrial wastewater.

4. A low-temperature catalyst based on zinc, copper, chromium oxides, and cement has been synthesized. Due to its paste-like structure and application to the inner surfaces of reactor tubes, the system has minimal hydraulic resistance and is capable of operating in dusty flows, which is a significant advantage over platinum analogues.

5. The parameters of thermal neutralization of nitrogen oxides using an arc plasma torch in the temperature range of 2000–4000°C have been established.

6. It has been proven that the introduction of reducing agents (methane, ammonia) shifts the reaction equilibrium toward the formation of molecular nitrogen, reducing nitrogen oxide concentration from critical levels to sanitary standards.

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