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On the Efficiency of Nitrogen-Containing Gaseous Waste Plasma Afterburning

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Abstract. The paper is devoted to the study of the plasma neutralization technology for toxic waste in the gaseous phase. It is noted that for effective implementation of plasma processes in waste processing technologies, it is necessary to search the optimal solutions taking into account the criteria of quality, productivity, cost and safety. In order to determine the feasibility of using plasma incineration technology, attention is paid to the definition of the appropriate nomenclature for gaseous toxic waste. The technology of plasma neutralization for thermal waste processing products – superoxicans (dioxins) and nitrogen-containing gas emissions (ammonia and nitrogen oxides) is studied. A modernized design of the plasma torch for the gaseous waste utilization is proposed. The known methods of thermal destruction for ammonia and nitrogen oxides are considered. Temperature approximations of their decomposition duration and efficiency in the range of plasma heating temperatures are found. The gas-dynamic parameters of the air-plasma flow in the process of nitrogen-containing gases thermal heating by plasma jet are determined using machine modelling methods. The effectiveness of the considered plasma neutralization technology is proved. The results of these studies should allow us to further formulate commonly used in engineering practice principles and methods for designing high-performance plasma heating technology.

1. Introduction

One of the most promising areas of the electroplasma technologies application [1–3] is their implementation in environmental projects related to the waste thermal processing [4]. The use of plasma torches in environmental projects is based on the effect of high-energy ($T_{pl} > 2000$ K) impact on materials. This effect allows to subject the compounds that make up this substance to deep decomposition – plasma incineration (“combustion”). However, for effective implementation of plasma processes in waste processing technologies, it is necessary to search the optimal solutions taking into account the criteria of the achieved result quality, productivity, cost and safety.

This article presents a number of results obtained in the framework of R&d under the RFBR grant “Development of fundamental scientific bases for the application of plasma incineration processes in waste recycling technologies”. In the course of this grant, the authors conduct research related to the elucidation of new physical laws about a plasma-arc discharge in a gas – dynamic flow, the relationship of these laws with the plasmatron properties, the physical concepts development about the processes of heat transfer accumulation, mass and charge flows in plasma-chemical systems and the features of polymorphic transformations in them. The results of these studies allow us to formulate commonly used in engineering



practice principles and methods for designing high-performance plasma heating technology [5]. The application of such developments, in turn, will be a new impetus for further fundamental research in the field of plasma and environmental technologies – the development of new plasma torches for heating materials, obtaining a new quality of waste disposal, switching to new algorithms of low-waste environmental technologies, solving numerous material science problems, etc.

One of the tasks solved in the study is to determine the nomenclature of gaseous toxic waste, for which it is advisable to use plasma incineration technology. It is known that the main costs for the implementation of environmental projects fall mainly on the gas treatment system [6], which includes several stages of passing gas emissions through cyclones, scrubbers, adsorbers, electromagnetic and mechanical filters. In this regard, the use of plasma technologies at the afterburning stage of waste gases can significantly improve the quality, simplify and reduce the cost of waste treatment and neutralization technologies, as well as solve the problem of neutralization of gaseous supertoxicants (dioxins, furans, etc.) formed during combustion.

2. Technique of researches

During the past period of project implementation (2019 – early 2020), studies were conducted on the processes of plasma incineration of supertoxicants (dioxins) in waste disposal and neutralization technologies [7]. The known methods of thermal neutralization of dioxins are considered and analyzed in detail. Based on a small amount of known information [8, 9] on their high-temperature neutralization (at temperatures of 15.000 and 50.000), the authors made approximations of their decomposition duration in the range of plasma heating temperatures based on the Arrhenius equation for the reaction rate constant. On the basis of the obtained approximating dependencies, the criteria for the effectiveness of dioxins high-temperature neutralization were obtained and the applicability of plasma incineration technology was justified.

As part of the study, it was noted that the assessment of the effectiveness for neutralizing toxic gases using arc-type plasma torches is a multi-parametric task, since in addition to the design parameters, it is necessary to take into account the gas-dynamic and heat-energy parameters of the technology. For this purpose, the gas-dynamic parameters of the air-plasma flow in the mixing chamber (MC) developed by the authors for the environmental technologies plasmatron [10] were determined using mathematical modelling methods. In the MC interaction of two gas streams – forming a plasma arc (jet) and a secondary flow of toxic gas is organized by constructive and technological methods. Various configurations of the MC (cylindrical and confusor type), as well as different ways of the recycled gas supply into the plasma jet (2- and 4-channel feed at different angles to the axis of the plasma jet) were studied. Two ways of heating the volume of the disposed gas in the MC with the use of plasma jets of different configurations are considered. According to the research results, the optimal design features of a plasma torch for eco-technologies was determined. This technology use a plasma arc of at least 170 mm in length and a temperature on the axis of the cylindrical MC of at least 7000 K, which provides heating of the neutralized gas mixture in the range of 2–10 ms at average temperatures of 2000–4000 K. Supply of toxic gases – through 4 pipes with a diameter of 4 mm, located perpendicular to the MC axis at a distance from the nozzle of 11 mm.

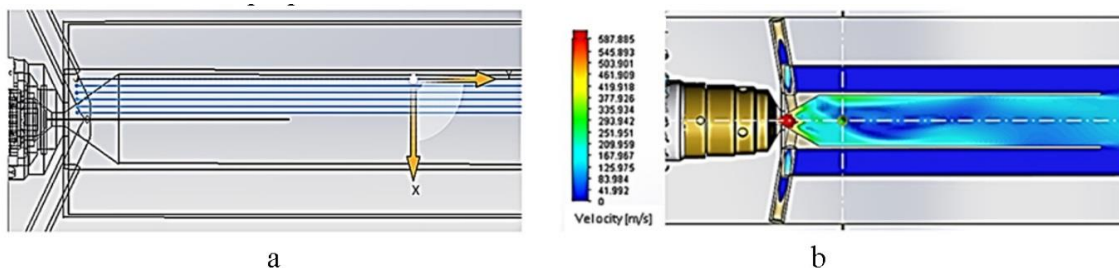


Figure 1. Calculation of gas dynamic parameters in the MC of plasma heating system: (a) linear trajectories for velocities and temperatures calculating; (b) velocity distribution.

In the course of the work, methods for calculating and analyzing the trajectory of gases (linear and spiral), their velocities, time and heating temperatures were proposed. Using these methods, the characteristic temperatures, rates and heating times of the disposed gas in various areas of the mixing chamber are calculated (Figure 1). The results correlate with the orders of the dioxins decomposition duration at such temperatures, thus indicating the effectiveness of the technology. This conclusion is also confirmed when evaluating the effectiveness of dioxins detoxification in accordance with the proposed criteria [11]. The obtained data allow us to evaluate the effectiveness of neutralization for dangerous types of nitrogen-containing gases formed in various technological processes (for example, metallurgical processes using nitric acid), as well as during thermal waste processing. These compounds should primarily include ammonia and a number of nitrogen oxides.

As you know, ammonia gas (NH₃) is a toxic compound, dangerous (up to fatal) if inhaled. A mixture of ammonia and air is explosive, and if there is a constant source of fire, it is flammable. Among the known classic nitrogen oxides – N₂O (dinitrogen oxide, also known as laughing gas), NO (nitric oxide), N₂O₃ (dinitrogen tetroxide), NO₂ (nitrogen dioxide), N₂O₅ (dinitrogen pentoxide) – hazardous chemicals include nitrogen oxide and dioxide. Nitric oxide irritates the eyes, skin and mucous membranes, and when inhaled causes serious poisoning, up to loss of consciousness and convulsions. Inhalation of nitric oxide, even at concentrations below the threshold, can lead to pulmonary edema. Nitrogen dioxide also irritates the skin and mucous membranes, and its inhalation can lead to chronic lung diseases and poisoning. In addition, with chronic exposure in low concentrations, nitrogen dioxide affects the immune system, reducing the body's resistance to diseases, causing genetic changes in people. The threshold of sensitivity to odors is 10 mg/m³, which is higher than the MPC (table.1). Symptoms of nitrogen dioxide poisoning differ from those of nitrogen oxide poisoning. However, it should be borne in mind that in some cases, people may be exposed to both of these substances simultaneously. In addition to production waste associated with the use of nitric acid, the formation of nitrogen oxides NO_x occurs during the combustion of all fuel types due to the content of nitrogen-containing organic substances in them. For example, the combustion products of 1 kg diesel fuel contain 20-40 g nitrogen oxides [12]. Nitrogen oxides, as sources of environmental problems such as acid precipitation, ozone depletion and photochemical smog, contribute to the degradation of ecosystems and human health. In this regard, an intensive search for ways to minimize the content of nitrogen oxides in emissions from various sources is quite natural.

Table 1. MPC of dangerous nitrogen-containing gaseous compounds.

n/n	Substances	MPC, mg/m ³		
		Maximum one-time	Daily average	Hazard class
1	Nitrogen dioxide (NO ₂)	0.085	0.04	2
2	Nitric oxide (NO)	0.6	0.06	3
3	Ammonia (NH ₃)	0.2	0.04	4

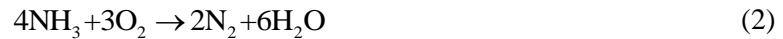
3. Results of research and their discussion

Almost all currently known works devoted to the neutralization of ammonia are studies of its decomposition using various catalytic systems made with transition metals (Fe, Pt, Ni, etc.) [13, 14]. Decomposition of ammonia begins at 270 °C, and at a temperature above 900 °C, ammonia is almost completely decomposed to nitrogen and hydrogen:



Decomposition of ammonia usually occurs with the formation of intermediate compounds. In this case, the rate of ammonia decomposition is largely determined by the rate of nitrogen

desorption. It is believed that the following variants of ammonia thermal conversion are possible – without a catalyst, at an elevated temperature (1200–1300 °C):



in the presence of a catalyst, at elevated temperature:



The authors [15] gave the time of decomposition of ammonia by reaction (1) in the temperature range of 700–1500K at a pressure of 3.92 MPa (table 2).

Table 2. Estimated time to establish chemical equilibrium t and characteristic completeness of completion of the decomposition reaction of ammonia X as a function of temperature T [15].

T, K	700	1000	1200	1500
t, c	$3.54 \cdot 10^{12}$	$1.11 \cdot 10^4$	6.31	$5.02 \cdot 10^{-3}$
X	0.822	0.9856	0.9951	0.9999

The Figures 2–4 show the results of numerical simulation of ammonia heating by an air-plasma jet in the mixing chamber of plasma torch for eco-technologies (the design and technological parameters correspond to the optimal values given below). Due to the fact that the efficiency of high-temperature decomposition of most substances is proportional to both temperature and heating duration, the product of these characteristics can be taken as an integral parameter. Analysis of the presented data shows that at the plasma heating temperatures in the MC, virtually complete decomposition of ammonia occurs. In this case, the most effective decomposition occurs at a distance of 3–4 mm from the MC axis, but the rest of the volume provides the necessary conditions for decomposition (Figure 3 a). Taking into account that heating occurs in a gas-dynamic non-uniform medium (in a rotating flow), it is advisable to pay attention to the results of calculation along the screw trajectories (with a pitch and diameter characteristic of the distribution of the gas flow in the MC). These results indicate a more effective (1.5–3 times) neutralization of the main mass of gas moving in the MC along a spiral trajectory. You should also note that under the conditions of heating the air plasma jet may be a partial oxidation of ammonia to nitrogen oxide NO (with excess oxygen according to reaction (3)), although given the flow rate of plasma (0.005 kg/s) and recyclable (0.004 kg/s) gases have to go mainly according to the scheme of reactions (1) and (2).

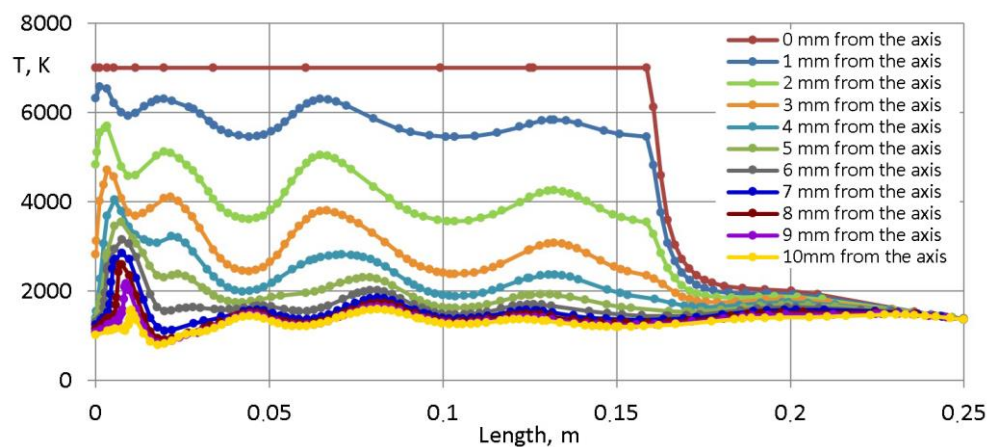


Figure 2. Temperature distribution in the plasma torch MC (calculation by linear trajectories).

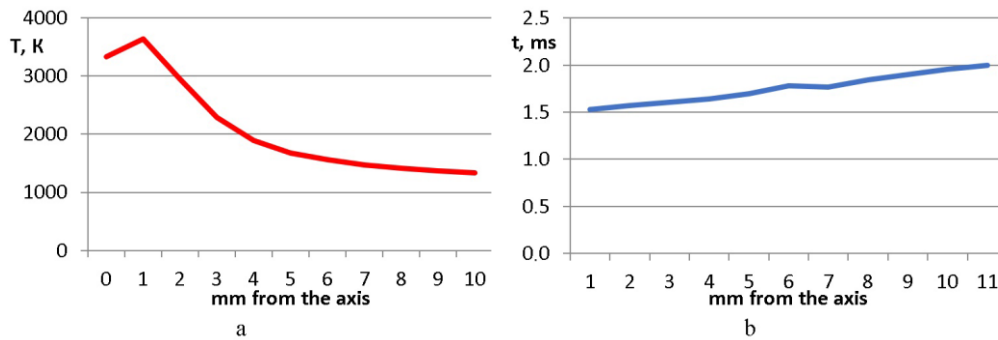


Figure 3. Average value of temperatures (a) and heating duration (b) of ammonia in the cross section of the plasma torch MC.

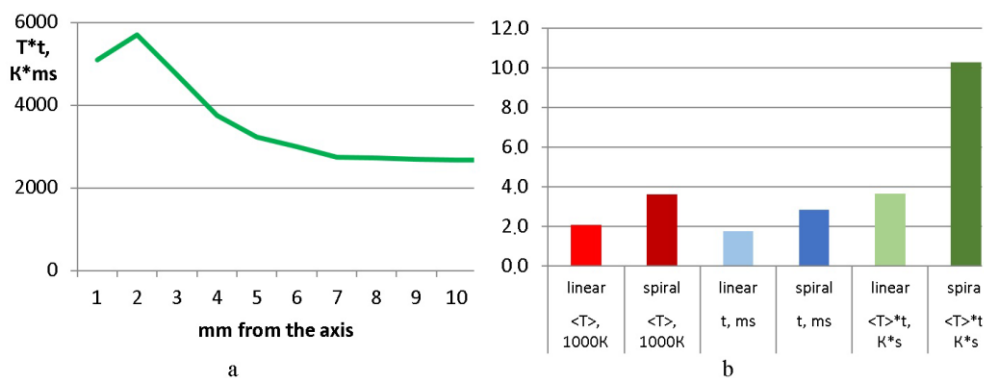


Figure 4. Ammonia heating efficiency (a – integral parameter, b – average parameters by trajectories) in the cross section of the plasma torch MC.

Special attention should be paid to the possibility of plasma neutralization for toxic nitrogen oxides. To date, gas purification from nitrogen oxides is based on the use of a variety of methods: catalytic, biochemical, non-catalytic, thermal, etc. Given the fact that catalytic methods have a number of known disadvantages (high cost of materials, the need for regular regeneration, etc.), you should pay attention to other methods of neutralization. Non-catalytic methods are based on the use of certain reducing agents, such as the above-mentioned ammonia, which is quite effective (up to 90%) in reducing nitrogen oxides [16]. The ammonia is stored in the container as ammonia water, which is injected into the gas stream at a high temperature, thus causing the reduction of nitrogen oxides according to the following schemes:



As can be seen from diagrams (4) and (5), this conversion of nitrogen oxides produces completely non-toxic compounds.

Thermal methods are practically not suitable for utilization of nitrogen oxides, since at typical temperatures (700–800 °C), the probability of nitrogen oxidation entering the combustion zone together with air and gas mixture is high [17]. In this regard, it is advisable to use step-by-step cleaning of gas mixtures, using methods aimed at neutralizing nitrogen oxides or step-by-step combustion of fuel. In recent years, plasma-chemical methods based on the destruction of recycled gases in a plasma jet with a temperature of several thousand degrees have become increasingly popular. The most effective is two-stage thermal decomposition of gaseous waste: the first stage is incomplete incineration of waste, and the second stage is the afterburning of incomplete combustion products, such as nitrogen oxides, carbon, and sulfur. In [18], it was shown that at temperatures of

2000–4000 °C, the equilibrium concentration of nitrogen oxide at the outlet remains quite high, about 2.4 % (at the initial concentration of nitrogen oxide 5 %), which corresponded to the degree of decomposition equal to 52 %. The process of high temperature method of nitric oxide decomposition can be described by the scheme:



Due to the fact that the amount of nitrogen oxide produced in the system (4) depends directly on the oxygen concentration, the authors [18] proposed using reducing agents (solid, liquid, and gaseous) for oxygen binding. It was found that the introduction of reducing agents into the system shifts the equilibrium of the system (4) towards products, i.e. towards the formation of nitrogen and oxygen, and this shift does not depend on the aggregate state of the reducing agent. Therefore, for the complete decomposition of nitrogen oxides, conducting reducing agents is preferable when using the plasma-chemical method.

To assess the effectiveness of neutralization, the authors calculated the equilibrium concentrations of nitrogen monoxide during its decomposition by plasma-chemical method. The experimental equations obtained in [19] on the plasma-chemical synthesis of nitric oxide were used as a basis. The rate of nitric oxide (II) formation at temperatures from 3500 K and above is determined according to the equation:

$$\frac{dC_{NO}}{d\tau} = 1,5 \cdot 10^{13} \cdot e^{-\frac{360340}{RT}} \cdot \frac{(C'_{NO})^2 - (C_{NO})^2}{\sqrt{C_{O_2}}}, \quad (7)$$

where C_{NO} and C_{O_2} are the initial concentrations of nitrogen oxide and oxygen; C'_{NO} is the equilibrium concentration of nitrogen oxide. Since the process of nitric oxide (II) obtaining is reversible, the time τ_p to establish its equilibrium concentration can be found by the formula:

$$\tau_p = \frac{5 \cdot 10^{11} \cdot e^{-\frac{360340}{RT}}}{\sqrt{C_{O_2}}} \cdot C'_{NO}. \quad (8)$$

Consequently, the equilibrium concentration of nitric oxide (II) of the reversible process of its decomposition (6) can be calculated by the equation:

$$C_{NO} = \frac{\tau \cdot \sqrt{C_{O_2}}}{5 \cdot 10^{11} \cdot e^{-\frac{360340}{RT}}}. \quad (9)$$

The choice of the initial oxygen concentration is based on the fact that the content of atomic oxygen in the fuel combustion products varies from 0.4 to 8.0 wt.%. The dependence of the equilibrium time in the system (6) on the temperature is presented in [19]. The calculated concentrations of nitric oxide (II) are shown in table 3. This table shows that as the temperature increases, the equilibrium concentration of nitrogen oxide in the system decreases. In all cases, the content of nitric oxide (II) in the system is less than the MPC value, which indicates a high degree of decomposition.

Figure 5 show the results of numerical modelling of gas-dynamic and thermo-physical processes of nitrogen heating in the plasma torch MC, which can be approximated with a small error to the processes of dangerous nitrogen oxides heating. The results of the analysis show that heating occurs at temperatures above 2000 K (the average temperature is 2700 K when calculated using a linear and 4000 K for a spiral trajectory) within a time of 2–3 ms. Taking into account the data presented in the table 3 it is possible to conclude that the NO content at characteristic temperatures and times of plasma heating in MC is significantly lower than the MPC, which indicates the effectiveness of the proposed method for nitrogen oxides neutralization.

Table 3. Typical decomposition time and NO concentration depending on heating temperature and oxygen concentration.

Oxygen concentration C_{O_2} , mol/dm ³	Heating temperature T , K	Time of complete decomposition t , ms	NO concentration	
			C_{NO} , mol/dm ³	C_{NO} , mg/m ³
0.0625	3000	1	$9.6 \cdot 10^{-10}$	0.029
0.0937	3000	1	$1.2 \cdot 10^{-9}$	0.035
0.125	3000	1	$1.4 \cdot 10^{-9}$	0.041
0.0625	4000	0.001	$2.6 \cdot 10^{-14}$	$0.78 \cdot 10^{-6}$
0.0937	4000	0.001	$3.2 \cdot 10^{-14}$	$0.95 \cdot 10^{-6}$
0.125	4000	0.001	$3.7 \cdot 10^{-14}$	$1.09 \cdot 10^{-6}$

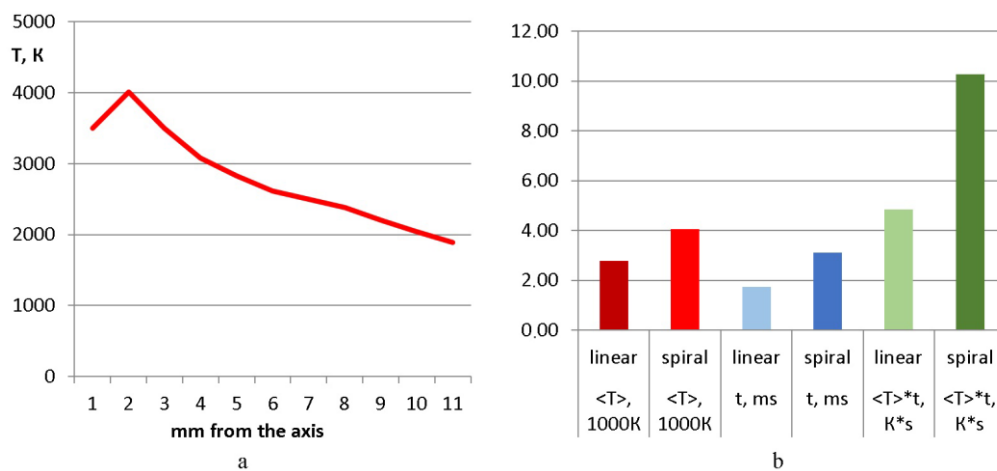
**Figure 5.** Average value of nitrogen heating temperature (a) and efficiency parameters (b) in the cross section of the plasma torch MC.

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4. Conclusions

Based on the results of this study, we can conclude that plasma neutralization method for waste containing toxic nitrogen of gaseous phase composition is justified by the example of ammonia, nitrogen monoxide and dioxide. The advantages of this method in comparison with the known technologies of high-temperature incineration and waste disposal are the speed and efficiency of the process. However, we should continue to develop and analyze this eco-technology in order to find optimal parameters for its application, as well as the development of new principles and methods of their design [20].

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