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HEAT-TRANSFER PROCESSES IN A PLASMA REACTOR IN DESTRUCTION OF PESTICIDES

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The possibility of using a multijet electric-arc plasma-chemical reactor for decontamination and destruction of different powder pesticides with an expired effective date and storage life which have not been used in agriculture is studied. Studies of heat transfer are conducted on a setup operating without processing of raw material and with supply of pesticides (isophen and butyl ester of 2,4-dichlorophenoxyacetic acid) to the reactor. The composition of waste gases at the reactor outlet is studied.

At present, the best results on destruction of toxic industrial waste are obtained using plasmachemical reactors with the heat-transfer agent heated in electric-arc plasmatrons [1]. This makes it possible to increase the temperature in the reaction zone of a plasma reactor to 2500 K and virtually eliminate the formation of ultratoxic compounds (dioxins, furanes, etc.) in the gas phase. One technological scheme of an electric-arc setup for destruction of waste is presented in [2].

The attainment of a high temperature of the plasma ($T > 5000$ K) at the reactor inlet is an obvious advantage of electric-arc generators. This ensures the possibility of processing materials with complex structures and compositions when the decomposition of a substance and subsequent individual treatment (chemical, thermal, radiative, etc.) of the produced components is required. Among the indicated substances are different waste materials, including refuse pesticides [3].

Results of the investigations in [4–9] showed good prospects for plasma reactors with multijet mixing chambers for treatment of dispersed materials and dispersed solutions. In [8], it is found that the reactor with a conical mixing chamber is the most efficient for three-jet chambers and cylindrical channels coaxial to them. For an angle of taper of 60^{o} used in the present work (Fig. 1), a plasma flow, owing to its homogeneity at the outlet from the mixing chamber [10], produces optimum conditions for the necessary physicochemical reactions.

In order to choose operating conditions of plasmatrons for destruction of pesticides, researchers have recently conducted a number of experiments with substances which simulate the indicated agricultural chemicals. The present study is aimed at developing these works and is devoted to optimization of the modes of an electric-arc plasma reactor for destruction of pesticides with an expired effective date. We study the processes of heat transfer with particles of powder agricultural chemicals in a heterogeneous plasma flow and also the composition of waste gases. The latter is necessary for both investigation of the processes in the reactor and choice of the modes of the plasmachemical device which satisfy environmental requirements.

The electric-arc setup for treatment of agricultural chemicals includes, in addition to the plasma reactor with a three-jet mixing chamber, a system for supply of agricultural chemicals to the reactor (powder

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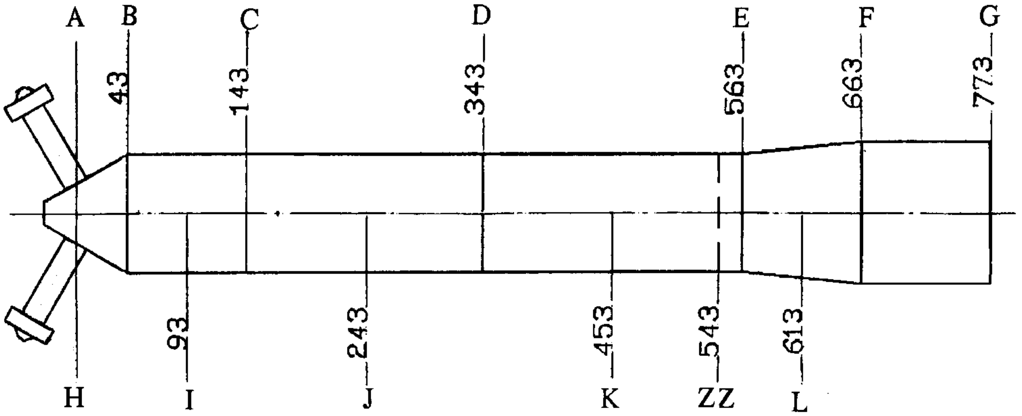

Fig. 1. Schematic diagram of the plasma reactor: A, B, C, ..., L, ZZ, typical zones of the reactor – A, introduction of plasma jets into the reactor; B, C, D, E, and F, entry of the flow to the first, second, third, and fourth sections of the reactor and the hopper, respectively; G, outflow from the reactor; H, plane taken in the calculations as a zero cross section; I, J, K, and L, middle of the first, second, third, and fourth sections; ZZ, introduction of a quenching gas.

scrubber feeder) and a device for feeding powder waste materials to the reactor. The diameter of the reactor channel is 100 mm. The position of the characteristic functional zones of the reactor is shown in Fig. 1. The quenching device, where the end products of the plasmachemical process are subject to quenching via intense cooling by an air flow and then arrive at the expanding hopper for additional cooling before discharge to the ventilation system, is positioned in the cross section *ZZ*. To take a sample of waste gases for analysis, we used a device which consists of a water-cooled sampler immersed in a gas flow and a vacuum pump. The content of nitrogen and carbon oxides in the gas was determined using a Testo-350 analyzer (Germany) in the on-line mode with operation of the plasma-chemical reactor.

The operating parameters of the plasma reactor are: supplied electric power N = 105–185 kW; total flow rate of a plasma-forming gas 6–12 g/sec; total flow rate of a gas on the setup G_{Σ} = 21–42 g/sec; mass-mean temperature of a plasma jet at the outlet nozzle of the plasmatron T = 5500–6000 K; mass-mean temperature of a plasma flow in the mixing chamber $T_{\mathrm{m.c}}$ = 4300–4800 K; efficiency of treatment of pesticides G_{r} = 0.1–5.0 kg/h. Pesticides with an expired effective date [isophen ($C_{14}H_{18}O_7N_2$) and butyl ester of 2,4-dichlorophenoxyacetic acid ($Cl_{l2}C_6H_3OCH_2COOCH_2CH(CH_3)_2$)] were studied. The two agricultural chemicals refer to pesticides in the form of wettable powders and contain about 50% of the base material (butyl ester and isophen), and the rest is a filler whose composition includes kaolin, aerosil, and silica gel [11].

Heat transfer in the reactor was studied by the method of calorimetric measurements of the heat fluxes to the wall of the sectioned channel of the plasma-chemical reactor. As a result, we obtained the distributions of the specific heat flux to the wall along the reactor length $q(l)$ and the enthalpy of a heterogeneous plasma flow $H(l)$ for different operating conditions of the reactor. It was found that $q(l)$ has a complex dependence on specific parameters of the plasmatron operation and has an error of about 30%, which is made up of errors of each operating parameter of the plasmatron (arc-current strength, voltage, flow rate of the plasma-forming gas, etc.). In order to improve the reliability of the results of studying the effect of quenching and the concentration of pesticides on heat transfer in the reactor and the composition of waste gases, in the present work we drew all the conclusions on the basis of comparison of the results of measurements in one series of experiments for a pure plasma jet without and with quenching or for a pure jet and a jet with pesticide particles.

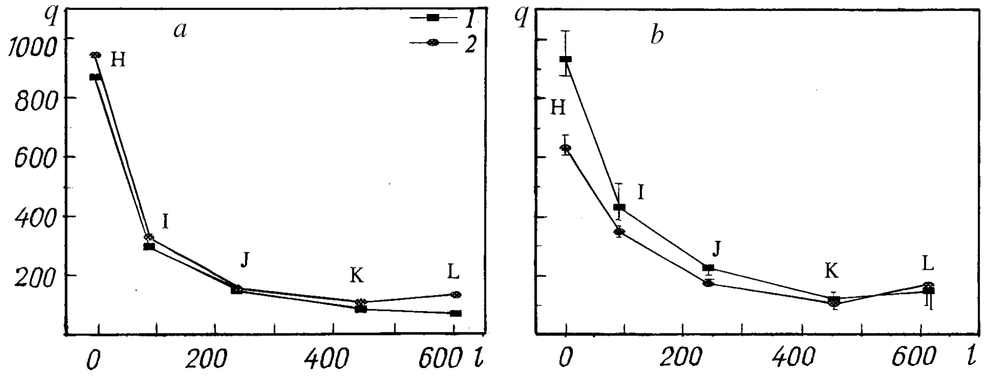

Fig. 2. Dependence of the distribution of the specific heat flux q along the reactor length l on the mode of reactor operation (N = 165, $N_{\mathrm{m.c}}$ = 105): a) G_{r} = 1 and G_{d} = 10: 1) G_{Σ} = 21.3 and G_{q} = 0; 2) G_{Σ} = 32.7 and G_{q} = 11.3; b) G_{q} = 11.3: 1) G_{Σ} = 48.4, G_{d} = 25.7, and G_{isophen} = 0.3; 2) G_{Σ} = 33, G_{d} = 10, and $G_{\mathrm{b.e}}$ = 0.76. l, mm.

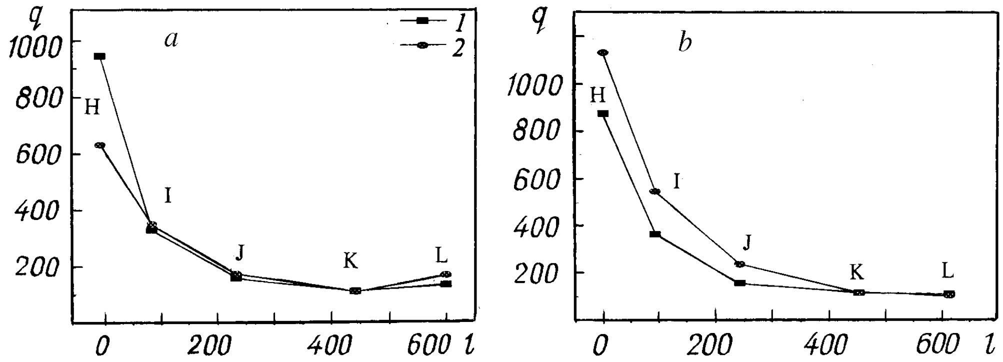

Fig. 3. Dependence of the distribution of the specific heat flux q along the reactor length l on the mode of reactor operation (N = 165, $N_{\mathrm{m.c}}$ = 105, G_{q} = 11): a) G_{d} = 10: 1) G_{r} = 0; 2) $G_{\mathrm{b.e}}$ = 0.76; b) 1) G_{r} = 0 and G_{d} = 17.2; 2) G_{isophen} = 1.36 and G_{d} = 12.9.

In the first stage, we studied heat transfer in a gas flow without its quenching and without supply of pesticides. It was found that the heat flux $q(l)$ to the wall of the reactor channel decreases with increase in the reactor length. This tendency is observed in different modes of the reactor and is in good agreement with the data obtained earlier [7, 8].

Studies of the effect of quenching on the character of heat transfer in the reactor were conducted for three values of the supplied power: N = 105, 165, and 185 kW. A smoother decrease in the heat flux $q(l)$ to the wall is observed for the intermediate value of power N = 165 kW, which correspondingly must lead to a gradual decrease in the temperature of the gas flow along the reactor length. This mode of reactor operation substantially simplifies calculation of heat transfer in the reactor owing to the absence of large gradients of the parameters of the working medium in supply of a quenching gas. It was taken as "optimum" and was studied in the work in detail.

The obtained experimental data of investigation of the heat fluxes to the reactor wall are shown in Figs. 2 and 3. Figure 2a presents a comparison of two distributions of $q(l)$ for operation of the plasmatron without (1) and with quenching (2) in the absence of pesticides. Distributions 1 and 2 virtually coincide within the experimental error. The differences exist only in the zones adjacent to the site of gas supply for quenching. In this case, a weak increase in the specific heat flux to the wall is also observed toward the reactor inlet. The main reason for this effect is, apparently, turbulization of the flow, including that in the layer bordering the wall. Simultaneously with decrease in the mean temperature of a plasma flow, equalization of the temperature over the flow cross section and, as a consequence, an increase in the specific heat flux to the reactor wall occur, which facilitates the efficiency of operation of the quenching device.

The addition of cold air to the gas flow at the reactor outlet owing to the large distance to the site of air supply for dilution of nitrogen oxides in order to improve the accuracy of an analysis of the composition of waste gases is likely to affect the thermal mode of the reactor.

Upon introduction of pesticides into the reactor (Figs. 2b and 3), its thermal mode changes greatly at a distance of 0–400 mm from the plane of introduction of plasma jets; at large distances, the specific heat flux is virtually constant. In supply of butyl-ester particles we observed a substantial decrease in the specific heat flux $q(l)$ compared to the jet without pesticides; when isophen particles are supplied, the heat flux increases. In the latter case, we observed a strong increase in $q(l)$ with increase in the pesticide supply to the reactor.

The main reason for the observed changes in $q(l)$ upon introduction of the pesticide into the reactor can be the effect of combustion of the organic component of agricultural chemicals. The presence of the dispersed material – agricultural chemical – in the flow facilitates the enhancement of heat transfer in the reactor channel. However, using the method of calorimetric measurements, we record only the total thermal effect and it is impossible to account for the effect of chemical reactions and the dustiness of the flow owing to the dispersed phase.

Thus, the experiments conducted indicate that the presence of organic and chlororganic compounds in the composition of pesticides can greatly affect heat-transfer processes in the plasma reactor. The determination of optimum modes of destruction of each existing type of pesticide (the total number of them exceeds at least one hundred) requires numerous measurements. This problem can be substantially be simplified by using the results of simulation of heat-transfer processes in the reactor after necessary measurements and determination of correlations that describe heat exchange between a heterogeneous flow and the walls of the reactor channel.

Processing of experimental data, based on which we calculated the results presented in Figs. 2 and 3, allows one to obtain initial information for a preliminary conclusion about the mechanism of heat transfer in the plasma-chemical reactor with pesticide materials. A decrease in the heat flux to the walls of the reactor channel along its length is observed in the experiments for both the dust-free flow and the dust-laden flow in supply of pesticide to it. In this case, according to [8], for the reactor with a conical mixing chamber and radial entry of plasma jets the results of the investigation of heat exchange between the plasma flow and the wall of the reactor channel are generalized well by the relation

$$\mathrm{St} = 1.012\,\mathrm{Re}_x^{-0.66}\,\mathrm{Pr}^{-0.67}\,.$$

Owing to the above, heat transfer in the reactor with a dispersed material – pesticide – is likely to be calculated using a similar relation supplemented with a special parameter [7], which allows for the effect of the dispersed material on transport processes.

Analysis of the composition of butyl ester and isophen and operating conditions of a plasma reactor with a high-temperature air flow used as the heat-transfer agent and the reactant shows that the main poisonous components in waste gases are, first of all, nitrogen and carbon oxides and also certain compounds of chlorine. The present work deals only with nitrogen and carbon oxides; the role of chlorine will be discussed later. Waste gases were monitored by an electrochemical method with the aid of a Testo-350 gas analyzer of the Testo Company (Germany) synchronous with measurement of the thermal characteristics of the plasma reactor. We studied the content, in a gas flow, of O_2 and NO, NO_2, CO, and CO_2 oxides, which are the main ingredients in formation of nitric and carbonyl acids, and also the content of carcinogenic nitrosodimethy-

TABLE 1. Volume Content of Nitrogen Oxides in a Gas Flow at the Outlet from the Plasma-Chemical Reactor in Different Modes of Its Operation

Volume content, %	Mode of reactor operation			
	without supply of raw material to the reactor ($G_{\text{isophen}} = 0$)		with supply of raw material to the reactor ($G_{\text{isophen}} = 0.3$ g/sec)	
	with quenching ($G_q = 11.3$ g/sec)	without quenching ($G_q = 0$)	with quenching ($G_q = 11.3$ g/sec)	without quenching ($G_q = 0$)
m_{NO}	0.98	0.3	1.4	0.3
m_{NO_2}	0.02	0.01	0.02	0.01

lamine [$(CH_3)_2HNO$]. The gas analyzer allows determination of the concentration of O_2 within the range 0–210,000 ppm (0–21% of the volume), the concentration of CO within the range 0–10,000 ppm (0–1% of the volume), the concentration of NO within the range 0–3000 ppm (0–0.3% of the volume), and the concentration of NO_2 within the range 0–500 ppm (0–0.05% of the volume). The maximum of the relative error of measurements of the concentration of the indicated gases was 5% of the measured value.

The estimates made earlier [12] by the method of evacuated flasks showed that the content of NO in waste gases after the reactor can reach 5000 ppm, which exceeds the upper limit of measurements by the Testo device. By virtue of the above, the flow coming from the reactor was diluted by a cocurrent flow of cold air. Mixing of turbulized flows began in the space zone at a distance of $X = 176$ cm from the quenching chamber. The coefficient of increase of the thickness of the mixing layer of cocurrent turbulent flows estimated based on the data of [13] is $b \approx 0.1$. Control samples were taken from the central part of the total flow at a distance of $X' = 300$ cm from the mixing zone. The thickness of the mixing layer in the sampling zone is $2bX' \approx 60$ cm $> D$, where $D = 40$ cm is the characteristic transverse dimension of the air flow. Owing to the above, we can assume that in cocurrent motion of waste and diluting flows rather effective mixing of them occurs on a portion of length $X' = 300$ cm. In this case, the true concentration of the oxides in the waste gas flow in the zone of its injection into the cold flow can be found by multiplication of the readings of the gas analyzer by the coefficient of dilution $K = 1 + G_g/G_\Sigma$, where G_g and G_Σ are the flow rate of the cold air and the gas from the plasma-chemical reactor, respectively. In our case, $K \approx 100$.

It is found from the measurements made that in the waste-gas flow, irrespective of the operating conditions of the reactor and the presence of pesticide, the concentration of monoxide CO is close to zero ($<10^{-4}$%) and the concentration of O_2 is at the level of 21%. The content of CO_2 was not monitored, since the algorithm of operation of the Testo-350 gas analyzer in determination of the concentration of CO_2 is based on calculation of the content of CO and O_2, rather than on direct measurements.

The values of the volume content of NO_x oxides at the reactor outlet, which are obtained by multiplication of the readings of the gas analyzer by the coefficient of dilution K, are given in Table 1.

It is seen from the tabulated data that in quenching and supply of isophen to the reactor the recorded concentrations of NO and NO_2 increase, whereas in operation without quenching their content is the same.

The obtained values of the concentration of NO are close to the data of [3, 12] and the results of our estimated measurements using the Testo-350 gas analyzer that are made without dilution of the waste flow.

Gulyaev et al. [14] noted the catalytic effect of excess oxygen on the reaction of decomposition of NO in nitrous gas mixtures at temperatures $T \sim 1300$ K. By virtue of this, in order to study the possibility of reducing the content of nitrogen oxides in the waste-gas mixture the scheme of dilution of the waste-gas flow by tangential injection of the cold-air flow into it in space zones with a gas temperature of $T \sim 1300$ K ($X = 56$ cm) for $K \approx 3$ was realized. The gas was taken for analysis from the central part of the flow in the cross section at a distance of 20 cm from the zone of dilution.

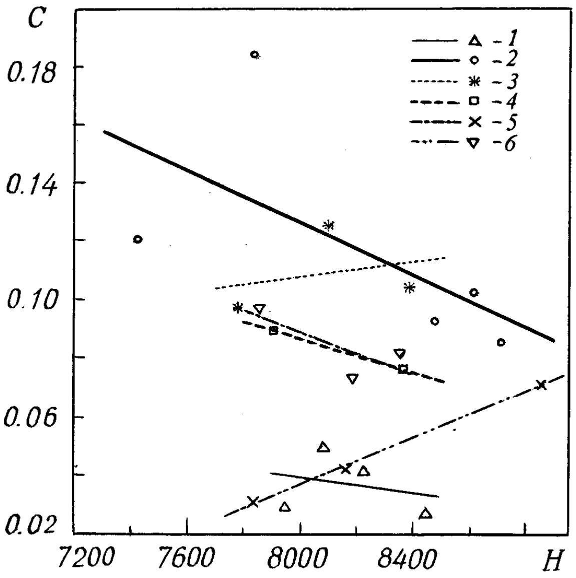

Fig. 4. Dependence of the volume content of NO on the specific enthalpy of the plasma *H* at the inlet to the plasma-chemical reactor for *N* = 165 and G_{Σ} = 6.5–13.5: 1) G_q = 0; 2) G_q = 11.35; 3) G_q = 0 and G_{isophen} = 0.3; 4) G_q = 11.35 and G_{isophen} = 0.3; 5) G_q = 0 and $G_{\mathrm{b.e}}$ = 0.9; 6) G_q = 11.35 and $G_{\mathrm{b.e}}$ = 0.9. *C*, vol.%; *H*, J/g.

The measured values of the concentrations of NO in the waste-gas flow in treatment of isophen and butyl ester and without them are presented in Fig. 4. The flow rate of isophen was ~ 0.3 g/sec, and the flow rate of butyl ester was ~0.9 g/sec.

The data presented in Fig. 4 indicate:

(1) Dilution of the waste flow by injection of cold air into it considerably decreases the content of nitrogen monoxide in the gas mixture. The maximum registered concentration of NO in the zone of monitoring was ~0.2%, and the minimum concentration was ~0.02%.

(2) Waste gases without quenching contain virtually the same amount of NO in operation of the plasma-chemical reactor in air and in treatment of butyl ester, but a smaller one than in treatment of isophen.

(3) The influence of quenching on the content of NO in waste gases greatly depends on the operating conditions of the plasma reactor. In operation of the plasmachemical reactor in air and in treatment of butyl ester, quenching increases the content of NO in the waste gases considerably. At the same time, in treatment of isophen, quenching decreases the concentration of NO for certain operating conditions of the plasma-chemical reactor. It should, however, be noted that the observed decrease is not very pronounced and virtually lies within the measurement error.

(4) In the mode with quenching, the content of NO in the waste-gas flow in treatment of isophen and butyl ester is virtually the same, but it is lower than in operation of the plasma-chemical reactor in air.

(5) With increase in the enthalpy of the plasma flow from 7200 to 8800 J/g, the content of NO in the waste-gas flow for the mode without quenching in operation of the plasma-chemical reactor in air and in treatment of isophen virtually does not change, whereas in treatment of butyl ester it increases.

(6) For modes with quenching we observe a decrease in the content of NO with increase in the enthalpy both in operation of the plasma-chemical reactor in air and in treatment of isophen and butyl ester. However, the falling character of the dependence of the NO content on the enthalpy is the most pronounced in the case of operation of the plasma-chemical reactor in air. The content of NO_2 in the waste-gas flow does not depend on the method of dilution and is at the level of hundredths of a percent.

The results obtained can be interpreted as follows.

In arc discharges in air at atmospheric pressure, the parameters of the generated plasma are close to equilibrium. Therefore, we can assume that the plasma produced in the plasma-chemical reactor is also equilibrium.

According to the data of spectroscopic and calorimetric measurements, the mass-mean temperature of a high-temperature flow in front of the quenching chamber of the plasma-chemical reactor is $T \sim 3500$ K. According to [15], the content of NO in this plasma is ~ 4–5 vol.%. In the absence of quenching, the plasma flow toward the discharge zone is accompanied by a decrease in the gas temperature to $T \sim 1200$ K and, owing to thermal decomposition of NO, by a decrease of up to 0.3% in its concentration (see Table 1). Fast cooling of the plasma flow ($\Delta T/\Delta t \sim 10^6$– 10^8 K/sec) in the quenching chamber to temperatures of $T \sim 2300$ K leads to holding of the volume content of NO in the waste-gas mixture at a level of ~1.5% (see Table 1).

In the case of dilution of the waste flow by injection of cold air into it, additional cooling of the waste flow occurs and, moreover, the effects of catalysis of the reactions of NO decomposition by excess oxygen from the injected air, which were noted in [14], manifest themselves. Because of this, the concentration of NO in the waste mixture decreases, and in the mode without quenching in operation of the plasma-chemical reactor in air and in treatment of butyl ester without nitrogen, it approaches an equilibrium value of ~0.04% in the air heated to $T \sim 1000$ K.

The falling character of the dependence of the volume concentration of NO on the energy contribution (specific enthalpy of the plasma H) at the inlet to the plasma-chemical reactor can be taken as a consequence of the reaction [16]

$$NO + NO \Rightarrow N_2 + O_2 \,.$$

The effect of this reaction is substantial when $T > 1500$ K, which determines the maximum permissible heating of the system of synthesized molecules of nitrogen monoxide. The gas temperature increases with increase in the energy contribution, and when $T > 1500$ K the content of NO begins to decrease (Fig. 4).

On the basis of the results obtained we can draw the following conclusions:

(1) A three-jet electric-arc module allows one to realize long-time modes of operation of the plasma-chemical reactor with introduced pesticides with an expired effective date. This makes further investigations of application of these reactors to industrial destruction of pesticides promising.

(2) The established possibility of investigating heat transfer in the plasma reactor with isophen and butyl ester by simulation methods makes the development of these methods urgent for investigation of processes in the plasma reactor with pesticides in order to develop a unique thermophysical model of destruction of such a wide class of agricultural chemicals.

(3) Butyl ester does not affect the content of nitrogen monoxide in the waste-gas flow, whereas isophen increases its amount about twofold. The used quenching of waste gases at a rate of their cooling of $<10^6$ deg/sec does not retain the values of the concentration of NO reached in the reactor plasma. The content of nitrogen monoxide in the waste gases does not exceed 0.1% of the volume and corresponds mainly to an equilibrium heated air. The technology of treatment of these amounts of NO in gas mixtures is well developed and presents no special technological difficulties.

(4) Reliable monitoring of the content of nitrogen monoxide at the outlet of technological plasma units requires thorough choice of the monitoring device, its verification, and preparation of a sample of the analyzed gas mixture.

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NOTATION

q, specific heat flux; kW/m^2; l, length of the reactor, mm; N, total supplied power, kW; $N_{\text{m.c}}$, power at the inlet to the mixing chamber, kW; G_{r}, specific flow rate of raw material, g/sec; G_{d}, gas flow rate for dilution of nitrogen oxides, g/sec; G_{g}, gas flow rate for quenching, g/sec; G_{isophen}, flow rate of isophen, g/sec; $G_{\text{b.e}}$, flow rate of the concentrate of butyl ester of 2,4-dichlorophenoxyacetic acid, g/sec; m, content of NO in the equilibrium plasma at atmospheric pressure; H, enthalpy, J/g; C_{NO}, content of NO, vol.%; St, Stanton number; Pr, Prandtl number; Re, Reynolds number; K, coefficient; X and X', distance, cm; b, coefficient of increase of the thickness of the mixing layer of cocurrent turbulent flows; $\Delta T/\Delta t$, temperature gradient, K/sec; ppM, one million fraction of the concentration of the substance in mol.%. Subscripts: x, along the reactor-channel length; q, quenching; d, dilution; r, raw material; m.c, mixing chamber; isophen, isophen particles; b.e, particles of butyl ester; Σ, total.

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