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14. ABSTRACT

This report results from a contract tasking General Physics Institute (GPI) as follows: The contractor will investigate the process of ignition and propagation of combustion in gaseous fuel/air mixtures initiated by DC high current and microwave gliding surface discharges. These discharges are sources of intense UV radiation that are capable of rapidly changing and enhancing the state of combustible mixtures (chemical composition, degree of electron and vibrational molecular levels excitation, degree of dissociation and ionization of molecules) in a short time preceding the onset of combustion. In the first year the contractor will investigate the induction times, combustion propagation velocity, gas temperature and final composition in a quiescent mixture as a function of parameters associated with the electric discharge ignitor. The second year will focus on testing and characterizing the effectiveness of the discharges on ignition and combustion in a supersonic stream.

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ISTC Project No. 2681p

**Basic Research of Strong UV Radiating Pulse
Discharge as an Igniter of Gaseous Mixtures
Combustion**

Final Project Activity Report

on the work performed from May 1, 2005 to May 1, 2007

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Настоящая работа выполняется по договору с Международным научно-техническим центром (МНТЦ), Москва, при финансовой поддержке <Финансирующая Сторона>

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Key words: plasma-assisted combustion, microwave discharge, plasma chemistry, microwave torch, surface electromagnetic wave

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1. Introduction

Paraxial plasma streams, or plasma jets (torches), were used (or were proposed) to initiate the combustion in flows (including, supersonic) of gas mixtures in a number of works (see, e.g., [1-4]). Analyzing these papers, however, we come to the conclusion that the mechanisms of stimulating the combustion by plasma streams are not yet completely understood. For this reason, the purpose of the project proposal was to attempt to determine possible mechanisms.

The systems used for the torch formation are of different design. In modern physical laboratory and plasma technology, electrode plasmatrons are most generally employed. However, considerable interest has recently been expressed by researchers and engineers in so-called microwave torches (see [5-8]) in which the energy source is microwave radiation producing a plasma stream in the working gas flow.

By using microwave torches as plasma sources in our studies under the Project, we investigated the processes underlying the action of plasma jets on combustible gas mixtures.

Depending on the sort of the working gas used in the plasmatron, on the jet region that contacts with the combustible gas mixture, power released in the plasma, and other factors, different processes can come to play, affecting the chemically active gaseous medium. Among these processes are the following:

- Local heating of the combustible gas mixture coming into contact with the torch, up to temperatures above the ignition temperature;
- Enrichment of the combustible gas mixture with chemically active radicals, which decreases the mixture ignition temperature and gives rise to chain branching chemical reactions and promotes the heating process;
- Photochemical processes in the gaseous medium that are stimulated by ultraviolet radiation of the plasma stream;
- Ion--molecule reactions that occur in the torch region and contribute to the heating and oxidation processes, etc.

To determine main mechanisms of affecting combustible gas mixtures by plasma streams, it is necessary to know the plasma and gas parameters (electron density, electron temperature, gas temperature) inside the torch and also to know the composition of the gas flow after processing by microwave discharge. The plasma and gas parameters in the microwave torch were studied in sufficient detail (see, e.g., [7 - 9]). Note, however, that these experiments were usually carried out with argon torch in air. Of interest to us was to determine main characteristics of torches produced from various gases and gas mixtures passing to the atmosphere of chemically active (combustible) gases. Of course, the jet characteristics should depend on the kind of working gas passing through the central electrode of the microwave torch.

Experimental studies of the microwave torch employed as a plasmachemical reactor are practically absent. Nevertheless, there is good reason to believe that a molecular gas passed through the high-temperature regions with high electron densities will change its composition by producing a substantial amount of chemically active radicals.

By the time of initiation of the present studies, an adequate physical and mathematical model of the microwave torch was absent.

Reviewing the works of predecessors, we notice that theoretical studies of the problem of the interaction of plasma streams with flows of combustible gas mixtures are small in number. Here, we should first of all cite very interesting papers by A.M. Starik et al. [10 - 13] based on a mathematical model of the combustion of $H_2:O_2$ mixtures enriched initially with chemically active radicals, atoms, and excited molecules. Thus, in [10], the authors studied the role of the excitation of molecular vibrations in H_2 . An analysis of the character of the influence of the initial content of O, H, and OH on the ignition was performed in [11]. In [12], the problem of the combustion of a hydrogen--oxygen mixture is solved for conditions when some initial

number of molecules of singlet oxygen $O_2(a^1\Delta_p)$ is added to the mixture. At last, in [13], the authors considered the role of vibrationally excited H_2O molecules in an $H_2:O_2:H_2O$ combustible mixture.

It should be noted, however, that the papers [10 - 13] do not reflect all aspects of the processes of enhancement of the reactivity of combustible gas mixtures. It is necessary to extend the list of participating radicals (in particular, to consider the role of NO), to include the ion-molecule reactions into the model, and to refine the constants of a number of processes participating in the stimulation or suppression of chain branching reactions of combustion.

Little attention was given to the theoretical problem of the combustion of mixtures containing the hydrocarbon component (for example, $CH_4:O_2$) with an initial excess of radicals or vibrationally (or electronically) excited molecules.

Working under the Project, we planned to put into operation a special experimental setup for studying the microwave torch in air, when it serves for plasma generation, and in methane-containing gas mixtures, when it operates as a plasmachemical reactor in which a dense nonequilibrium plasma involves a substantial change in the composition of the working gas. As one of the most important directions of experiments, we planned to perform measurements of the composition of the working gas passed through the torch region.

It was planned that the theoretical research efforts would primarily be directed toward the construction of a theory for coaxial microwave torch.

We also planned to prepare a review of theoretical and experimental papers devoted to the initiation of combustion in hydrogen-oxygen mixtures acted upon by chemically active radicals and atoms present (either introduced or formed) in these mixtures.

At last, we planned to carry out experiments in testing chambers of the Central Institute of Aviation Motors (CIAM). In these experiments, the microwave torch, designed and tested previously at the GPI, was tested as an initiator of combustion in a kerosene-air mixture flow.

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[13] A.M. Starik, N.S. Titova, B.I. Lukhovitskiy.. Kinetics of low-temperature initiation of combustion of $H_2+O_2+H_2O$ mixtures at excitation of molecular vibrations of H_2O by laser radiation. Technical Physics, V. 74, N 1, pp. 77-83 (in Russian).

2. Problem of Activation of Hydrogen--Oxygen Mixtures (Analysis of the Literature and Testing of our Kinetic Model)

Over the period of report, we analyzed a great body of the literature concerning theoretical and experimental research on the Problem of activation of hydrogen--oxygen gas mixtures. The results were generalized in a review that will be submitted for publishing. The review includes three Chapters. Chapter I discusses theoretical and experimental studies of the special case that hydrogen and oxygen atoms whose concentration can exceed the critical value may well affect both the induction times and the ignition temperature range for $H_2:O_2$ mixtures. The question of how the induction times of $H_2:O_2$ mixtures are affected by electronically excited oxygen molecules in singlet states $O_2(a'\Delta_g)$, $O_2(b'\Sigma_g^+)$ is the concern of Chapter II of the review. At last, in Chapter III we discuss the influence of vibrational excitation of original reagents on the induction times of $H_2:O_2$ mixtures. According to our estimates the chemical activity of the medium can be enhanced by exposing the original hydrogen--oxygen mixture to ultraviolet radiation, electric discharge, or an electron beam.

Based on experimental data and theoretical calculations, we estimate the nonequilibrium concentration of atomic oxygen and hydrogen that involves a reduction of the induction period.

Analyzing the influence of vibrational excitation of H_2 and O_2 molecules, we come to the conclusion that, on account of high rates of VT-relaxation for main components of fuel mixtures, it is hardly possible to use the vibrational excitation for the purposes of reduction of the induction times.

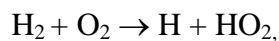
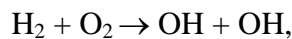
Preliminary excitation of oxygenic mixtures by electric discharges can provide a sufficiently high density of metastable electronically-excited molecules $O_2(a'\Delta_g)$. The effect that an addition of $O_2(a'\Delta_g)$ exerts on the combustion wave velocity and the induction times of $H_2:O_2$ mixtures was considered in a number of papers. It is shown that the induction period and the threshold temperature for ignition of hydrogen--oxygen mixtures can substantially be reduced if the concentration of the oxygen singlet exceeds $O_2(a'\Delta_g)/O_2 \geq 5\%$. However, lacking a reliable information about the rate constants and pathways of mains reactions with

the participation of $O_2(a'\Delta_g)$, the effect of singlet hydrogen additions cannot be examined in more detail.

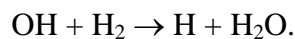
For a more comprehensive analysis of the experimental and theoretical results discussed in the review, we compared literature values with results of simulation using a kinetic model of the combustion of hydrogen--oxygen mixtures that were developed by us. Below, we present a brief description of this model along with results and a discussion of test calculations.

In the course of work on our mathematical model of the combustion of a $H_2:O_2$ mixture, we analyzed and used the data of [1-4]. The main reactions and rate constants are tabulated in Table 1.

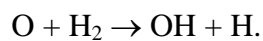
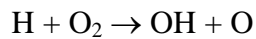
The combustion of hydrogen--oxygen mixtures is a typical example of the chain mechanism. The main source of radicals at low temperatures is associated with the reactions



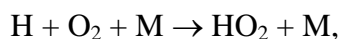
that are accompanied by a rather fast formation of H atoms and H_2O molecules in the process



The hydrogen atoms react with O_2 molecules to form O atoms:



This process is the main branched chain reaction proceeding at the combustion at high temperature. At high pressures and relatively low temperatures, the other (three-body) process comes into importance:



resulting in the production of HO_2 radicals which are chemically less active than O and OH radicals.

The distinctive feature of our model (Table 1) is the inclusion of the process



with the rate constant

$$k_1 = 10^{-9} \exp(-24200/T) \quad \text{cm}^3/\text{s}. \quad (2)$$

Many kinetic schemes [1-3, 5] ignore this reaction. In [6], it was pointed to the importance of reaction (1). In [4], the authors emphasized the importance of reaction (1) for the radical production at low temperatures. However, the constant k_1 used in [4] is too large to be consistent with available data. We used the rate constant k_1 that was deduced by analyzing the

NIST database and also by comparing the calculated and experimental dependences of the induction times on the initial gas temperature (see below).

We considered the results of experiments in which the gas heating occurs in a shock wave (SW). The characteristics of the mixture behind the SW front were determined from the measured values of the wave velocity and from initial gas parameters (pressure, temperature, composition). Note that the pressure is usually measured behind the shock wave. The induction time in experiments is counted from a sharp pressure peak (associated with the SW) to a sharp growth in the intensity of diagnostic radiation (associated with OH*, H₂O molecules, etc.).

To check whether our model adequately describes the kinetics of combustion of H₂:O₂ mixtures, we compared the calculated and experimental dependences of the induction period τ_{in} on the initial gas temperature (for various ratios of the mixture components). The induction time in the calculations, as in [4], was determined from a peak of the hydrogen atom density.

The induction period and gas parameters in the combustion region of an H₂: air mixture were calculated assuming that nitrogen is merely a buffer gas (i.e., N₂ molecules do not participate in chemical conversions). On this assumption we calculated the induction period for a stoichiometric mixture H₂:O₂:N₂ = 29.6:14.8:55.6 at a pressure of $p = 1$ atm in the temperature range $T = 900 - 2000$ K (Fig. 1). The results of simulation were compared with the available experimental data [7, 8] and the calculations [4, 5]. As is seen from Fig. 1, our model gives a good fit to the experimental data. The results of calculations using the GRI-Mech model [5] (curve 3) agree with the experiment at low temperatures $T \leq 1000$ K, but give somewhat longer induction times at $T > 1000$ K. The values calculated in [4] (curve 1) at $T < 950 - 960$ K differ by more than one order of magnitude from the experimental data and also from values calculated in [5] and in our model. The immediate cause for such apparent disagreement might be a too large constant for reaction (1): $k_1 = 2,8 \cdot 10^{-9} \exp(-24200/T)$ cm³/s [4].

Figure 2 shows the results of simulation for a stoichiometric mixture H₂: air at $p = 2$ atm and confirms all main conclusions that follow from Fig. 1 for $p = 1$ atm. For comparison, the results of calculations using the model [9] (borrowed from [6]) are shown in Fig. 2.

We also studied hydrogen--oxygen mixtures to which argon was added as a buffer gas. In [10], the combustion time was measured for a mixture H₂:O₂:N₂: Ar = 6:3:11:80 at a pressure of $p = 0,5 - 0,8$ atm in the temperature range $T = 900 - 1400$ K. These temperature values were measured in the gas behind the shock wave. We took, as the pushing gas, air (points in Fig. 3) and CO₂ (triangles). The induction time was determined from the time behavior of the radiation intensity of OH molecules at a wavelength $\lambda = 306.4$ nm. In [11], a

nonstoichiometric mixture $\text{H}_2:\text{O}_2:\text{Ar}=8:2:90$ was studied at $p=5$ atm and nearly the same initial temperatures. Figure 3 shows the results of calculations and measurements [10,11] of the induction time for these mixtures. The calculations for conditions of [10] were performed at $p = 0.6$ atm. The results of these calculations agree well with the experimental data over the entire temperature range under study.

In [9], the temporal evolution of the relative content of H_2 , O_2 and H_2O molecules was measured during the combustion of hydrogen—oxygen mixtures with nitrogen additions. The studies were conducted in a mixing reactor at temperatures of $T_0 = 850 - 1021$ K, in the pressure range $p = 0.3 - 5$ atm.

Figures 4a and 4b show the experimental data [9] and the calculated dependences for a mixture $0.5\%\text{H}_2:0.5\%\text{O}_2:99\%\text{N}_2$ at a pressure of $p = 0.3$ atm and initial temperature of $T_0 = 880$ K. Under conditions of the experiment [9], the stirring effects play a part in determining the mixture ignition time. These effects are taken into account by properly changing the values of t . In Fig. 4, the calculated curves are shifted by $\Delta t = 34.75$ ms which is chosen such that the calculated and experimental results coincide at the time corresponding to a 50% decomposition of the initial hydrogen content in the mixture. In such a case, the calculated dependence agree well with the experiment.

Comparing the calculated and experimental data we arrive at the conclusion that the available experimental results are described adequately and our kinetic model can be used for predictions.

Based on the results of our analysis we can make the following preliminary conclusions.

Additions of oxygen and hydrogen atoms to combustible mixtures can reduce the induction times of these mixtures substantially. A considerable effect in this case is achieved when the relative concentration of such additions is more than 10^{-3} . The addition of atomic particles also makes it possible to lower the threshold temperature for ignition of mixtures. This effect, however, takes place only near the ignition threshold. For relatively low initial temperatures, a shift of the ignition temperature is due to a self-heating of the mixture through recombination of added atomic particles. In this case, the nonequilibrium character of the action is of little consequence.

In the case of spatially uniform excitation of a mixture by UV radiation or by electric discharge, the experimental data on the induction time can satisfactorily be fitted by using the available kinetic models. This fact testifies that we possess a relatively full database of the rate constants of chemical reactions proceeding in hydrogen--oxygen mixtures.

To increase the length of a chain of chemical reactions at temperatures below the ignition threshold, we propose to use the energy of nonequilibrium electronically excited vibration of molecules. The excited particles must be chemically active, and at the same time they must have a sufficiently long lifetime (with respect to their deexcitation at collisions). In this review we consider how the ignition of $H_2 : O_2$ mixtures is affected by oxygen singlet (OS) molecules $O_2(a^1\Delta_g)$ and by vibrationally excited molecules $H_2(v)$.

The oxygen singlet molecules can participate in the incipient chain reactions ($O_2(a^1\Delta_g) + H_2$) as well as in the branched chain reactions ($O_2(a^1\Delta_g) + H$). The rates of these processes grow substantially with temperature. However, the rate for quenching of the oxygen singlet $O_2(a^1\Delta_g)$ by hydrogen molecules also grows with temperature. All previous models ignored the fact that deexcitation of $O_2(a^1\Delta_g)$ accelerates as the mixture temperature increases, thereby sharply reducing the effect of OS on the induction times. At low temperatures, where the quenching is of minor importance, the presence of molecules $O_2(a^1\Delta_g)$ can substantially influence the ignition of $H_2 : O_2$ mixtures, if the level of relative OS concentrations will exceed 10^{-2} .

Vibrationally excited molecules $H_2(v)$ can also participate both in the incipient chain reactions ($H_2(v) + O_2$) and in the branched chain reactions ($H_2(v) + O$). The obtained results suggest that the threshold temperature for ignition of hydrogen--oxygen mixtures can be reduced considerably by exciting vibrational degrees of freedom of $H_2(v)$ molecules. However, when electric discharges are used for excitation of $H_2 : O_2$ mixtures and the discharge energy can be efficiently transferred to the vibrational subsystem $H_2(v)$, it will be difficult to attain high levels of vibrational excitation ($T_v^{H_2} \geq 2000$ K). For H_2 --air mixtures, when most of discharge energy goes into vibrational excitation of nitrogen molecules, this problem becomes even more complicated. The reaction rates for $H_2:O_2$ mixtures are given by the formula $k = k_0 T^n \cdot \exp(-E_a / T)$, where both E_a and T are expressed in K. Table 1 lists values of the factor k_0 expressed in cm^3/s for two-particle reactions and in cm^6/s for three-particle reactions.

Table 1

№	Reaction	k_0	n	E_a
1.	$H + O_2 + M \rightarrow HO_2 + M$	$5.8 \cdot 10^{-30}$	-0.8	-
R1.	$HO_2 + M \rightarrow H + O_2 + M$	$6.1 \cdot 10^{-6}$	-0.85	24800
2.	$O + HO_2 \rightarrow OH + O_2$	$3.0 \cdot 10^{-11}$	-	-200
R2.	$OH + O_2 \rightarrow O + HO_2$	$3.7 \cdot 10^{-11}$	-	26500
3.	$O + OH \rightarrow H + O_2$	$2.4 \cdot 10^{-11}$	-	353
R3.	$H + O_2 \rightarrow O + OH$	$1.62 \cdot 10^{-10}$	-	7470

4.	$\text{H} + \text{HO}_2 \rightarrow \text{OH} + \text{OH}$	$2.8 \cdot 10^{-10}$	-	440
R4.	$\text{OH} + \text{OH} \rightarrow \text{H} + \text{HO}_2$	$1.1 \cdot 10^{-13}$	0.678	18240
5.	$\text{H} + \text{HO}_2 \rightarrow \text{H}_2 + \text{O}_2$	$1.1 \cdot 10^{-10}$	-	1070
R5.	$\text{H}_2 + \text{O}_2 \rightarrow \text{H} + \text{HO}_2$	$3.0 \cdot 10^{-11}$	-	24080
6.	$\text{H} + \text{HO}_2 \rightarrow \text{H}_2\text{O} + \text{O}$	$5.0 \cdot 10^{-11}$	-	866
R6.	$\text{H}_2\text{O} + \text{O} \rightarrow \text{H} + \text{HO}_2$	$3.2 \cdot 10^{-13}$	0.621	27166
7.	$\text{H} + \text{OH} \rightarrow \text{O} + \text{H}_2$	$4.7 \cdot 10^{-20}$	2.643	2246
R7.	$\text{O} + \text{H}_2 \rightarrow \text{H} + \text{OH}$	$8.5 \cdot 10^{-20}$	2.67	3160
8.	$\text{OH} + \text{HO}_2 \rightarrow \text{H}_2\text{O} + \text{O}_2$	$4.8 \cdot 10^{-11}$	-	-250
R8.	$\text{H}_2\text{O} + \text{O}_2 \rightarrow \text{OH} + \text{HO}_2$	$7.71 \cdot 10^{-12}$	-	37280
9.	$\text{OH} + \text{OH} \rightarrow \text{H}_2\text{O} + \text{O}$	$2.5 \cdot 10^{-15}$	1.14	50
R9.	$\text{H}_2\text{O} + \text{O} \rightarrow \text{OH} + \text{OH}$	$4.0 \cdot 10^{-14}$	1.083	8650
10.	$\text{OH} + \text{H}_2 \rightarrow \text{H} + \text{H}_2\text{O}$	$1.7 \cdot 10^{-16}$	1.6	1660
R10.	$\text{H} + \text{H}_2\text{O} \rightarrow \text{OH} + \text{H}_2$	$7.5 \cdot 10^{-16}$	1.6	9270
11.	$\text{H} + \text{OH} + \text{M} \rightarrow \text{H}_2\text{O} + \text{M}$	$6.1 \cdot 10^{-26}$	-2.0	-
R11.	$\text{H}_2\text{O} + \text{M} \rightarrow \text{H} + \text{OH} + \text{M}$	$5.8 \cdot 10^{-9}$	-	52920
12.	$\text{H} + \text{H} + \text{M} \rightarrow \text{H}_2 + \text{M}$	$5.14 \cdot 10^{-30}$	-1.0	-
R12.	$\text{H}_2 + \text{M} \rightarrow \text{H} + \text{H} + \text{M}$	$1.65 \cdot 10^{-5}$	-0.86	52088
13.	$\text{O} + \text{O} + \text{M} \rightarrow \text{O}_2 + \text{M}$	$2.37 \cdot 10^{-31}$	-0.63	-
R13.	$\text{O}_2 + \text{M} \rightarrow \text{O} + \text{O} + \text{M}$	$3.66 \cdot 10^{-8}$	-	59380
14.	$\text{H} + \text{O} + \text{M} \rightarrow \text{OH} + \text{M}$	$1.3 \cdot 10^{-29}$	-1.0	-
R14.	$\text{OH} + \text{M} \rightarrow \text{H} + \text{O} + \text{M}$	$1.05 \cdot 10^{-5}$	-0.97	51420
15.	$\text{OH} + \text{OH} \rightarrow \text{H}_2 + \text{O}_2$	$1.0 \cdot 10^{-11}$	-	14800
R15.	$\text{H}_2 + \text{O}_2 \rightarrow \text{OH} + \text{OH}$	$1.0 \cdot 10^{-9}$	-	24200
16.	$\text{HO}_2 + \text{H}_2 \rightarrow \text{OH} + \text{H}_2\text{O}$	$1.08 \cdot 10^{-12}$	-	9400
R16.	$\text{OH} + \text{H}_2\text{O} \rightarrow \text{HO}_2 + \text{H}_2$	$1.2 \cdot 10^{-14}$	0.43	36100

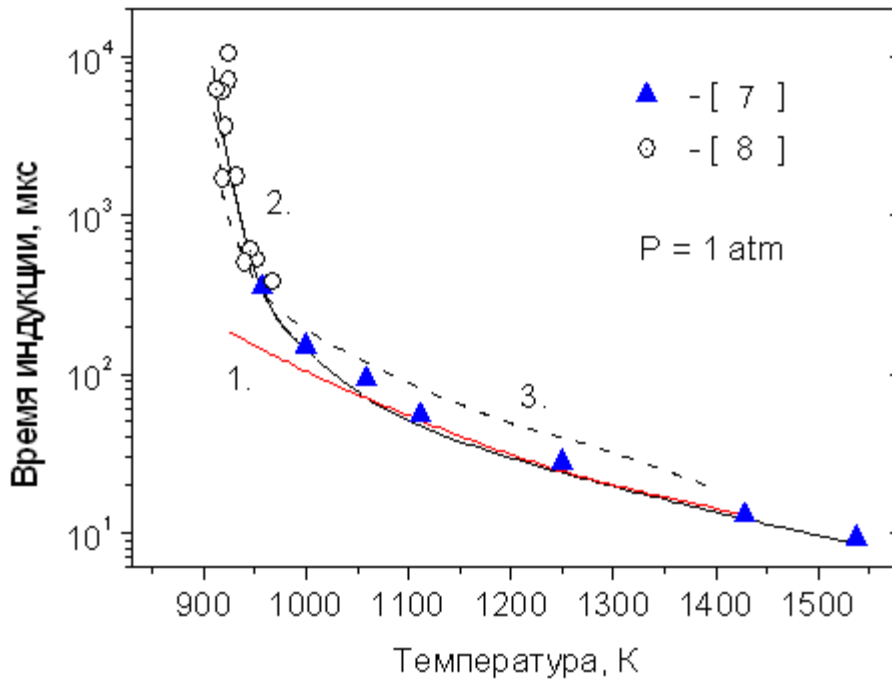


Fig.1.

Dependence of the induction period on the initial gas temperature for a mixture $H_2 : O_2 : N_2 = 29.6 : 14.8 : 55.6$ ($P = 1 \text{ atm}$).

Symbols show experimental points [7, 8]. Curves show results of calculations: (1) [4], (2) our model, and (3) (dashed line) [5].

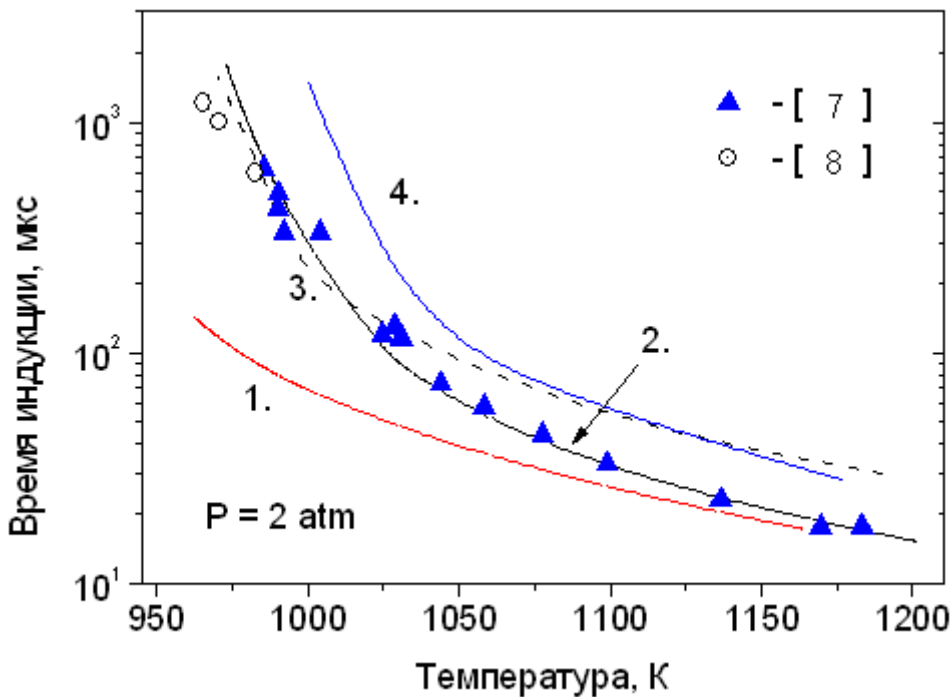


Fig. 2.

Dependence of the induction period on the initial gas temperature

for a mixture $H_2 : O_2 : N_2 = 29.6 : 14.8 : 55.6$ ($P = 2$ atm). Symbols show experimental points [7, 8]. Curves show results of calculations: (1) [4], (2) our model, (3) (dashed line) [5], and (4) [9].

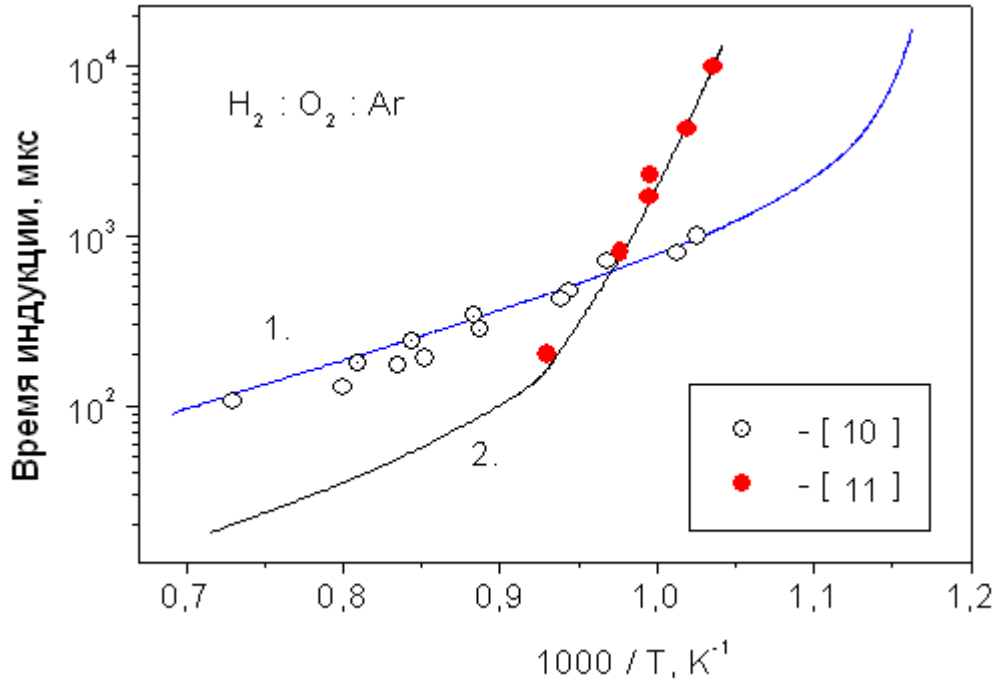


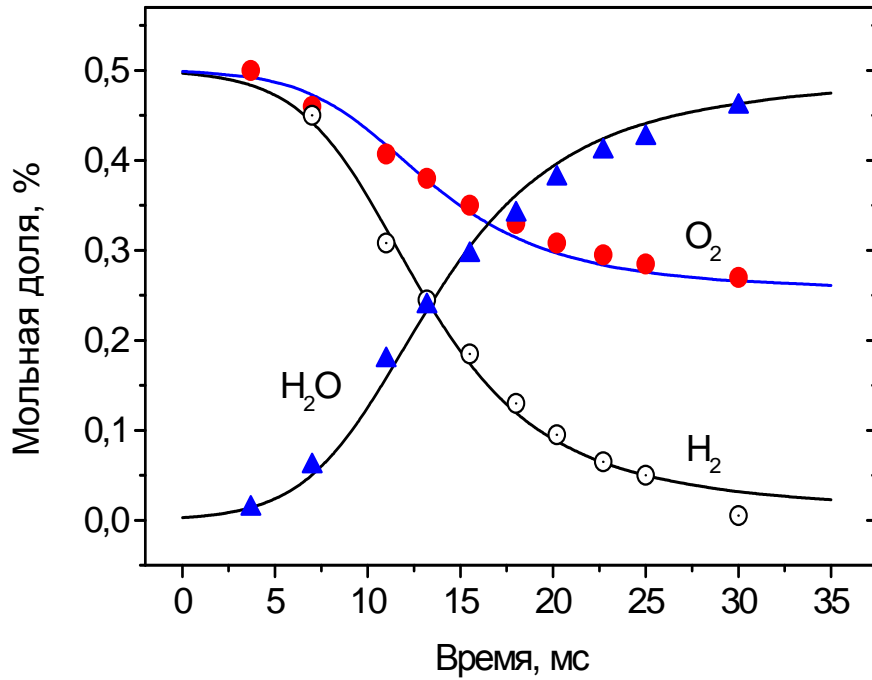
Fig. 3.

Dependence of the induction period on the initial gas temperature for mixtures

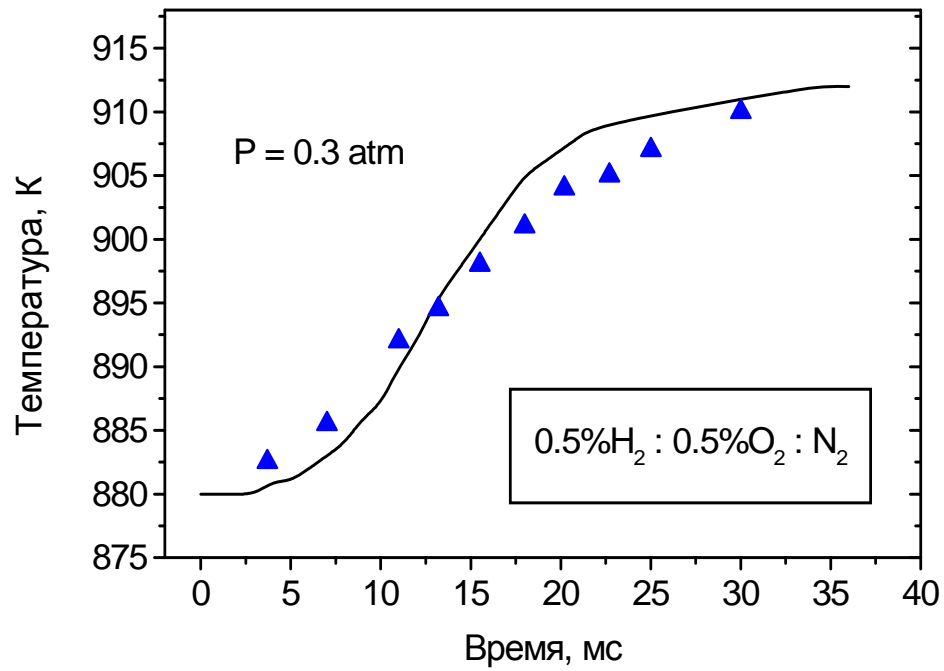
(1) 6% H_2 : 3% O_2 : 11% N_2 : 80% Ar ($P = 0.5-0.8$ atm)

and (2) 8% H_2 : 2% O_2 : 90% Ar ($P = 5$ atm).

Points show experimental data [10,11]. Curves show results of calculations.



a)



b)

Fig. 4.

Temporal evolution of (a) the relative content of H₂, O₂, and H₂O components and (b) gas temperature at the combustion of a mixture 0.5% H₂ : 0.5% O₂ : 99% N₂.

P = 0.3 atm, T₀ = 880 K. Symbols show experimental points.

Curves show results of calculations.

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3. Physical and mathematical model of a coaxial microwave torch

In the course of project implementation, we developed a two-dimensional magnetohydrodynamic (MHD) model of an equilibrium microwave discharge in a coaxial waveguide with a truncated inner electrode. Characteristics of a microwave discharge in atmospheric-pressure argon are calculated. The aim of this work is to determine the electromagnetic, thermal, and gasdynamic structures of the discharge and to estimate adequacy of the numerical model to the discharge features observed in experiments.

We consider a microwave discharge excited by a TEM wave in a coaxial waveguide with a truncated inner electrode - a cylindrical tube through which the working gas is supplied at a flow rate of G_0 (Fig. 1). It is assumed that the electrode material is perfectly conducting and the cylindrical wall of the outer electrode of radius R_e is permeable to the gas flow. In the axisymmetric equilibrium plasma flow that forms in the circular beyond-cutoff waveguide, there is an electromagnetic field with the components $\vec{E}(E_r; 0; E_z) \exp(i\omega t)$ and $\vec{B}(0; B_\phi = B; 0) \exp(i\omega t)$. The period of the electromagnetic field is much

shorter than the time during which steady-state distributions of the thermal and gas-dynamic parameters of the discharge are established.

Equations. The gas-dynamic, thermal, and electromagnetic parameters of the equilibrium plasma of a microwave discharge were determined by solving the set of time-independent axisymmetric equations of continuity, motion (Navier-Stokes equation), and energy balance and the wave equations:

$$\frac{1}{r} \frac{\partial}{\partial r} (r \rho v) + \frac{\partial}{\partial z} (\rho u) = 0 \quad (1)$$

$$\rho \left(v \frac{\partial v}{\partial r} + u \frac{\partial v}{\partial z} \right) = -\frac{\partial p}{\partial r} - \frac{1}{4} \sigma (E_z B_\phi^* + E_z^* B_\phi) + \frac{2}{r} \frac{\partial}{\partial r} \left(r \eta \frac{\partial v}{\partial r} \right) - \frac{2 \eta v}{r^2} + \frac{\partial}{\partial z} \left[\eta \left(\frac{\partial u}{\partial r} + \frac{\partial v}{\partial z} \right) \right] - \frac{\partial}{\partial r} \left[\frac{2}{3} \eta \left(\frac{1}{r} \frac{\partial r v}{\partial r} + \frac{\partial u}{\partial z} \right) \right], \quad (2)$$

$$\rho \left(v \frac{\partial u}{\partial r} + u \frac{\partial u}{\partial z} \right) = -\frac{\partial p}{\partial z} + \frac{1}{4} \sigma (E_r B_\phi^* + E_r^* B_\phi) + 2 \frac{\partial}{\partial z} \left(\eta \frac{\partial u}{\partial z} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left[r \eta \left(\frac{\partial u}{\partial r} + \frac{\partial v}{\partial z} \right) \right] - \frac{\partial}{\partial z} \left[\frac{2}{3} \eta \left(\frac{1}{r} \frac{\partial r v}{\partial r} + \frac{\partial u}{\partial z} \right) \right] \quad (3)$$

$$\rho C_p \left(v \frac{\partial T}{\partial r} + u \frac{\partial T}{\partial z} \right) = \frac{1}{2} \sigma (E_r E_r^* + E_z E_z^*) - \varphi_e + \frac{1}{r} \frac{\partial}{\partial r} \left(r \lambda \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left(\lambda \frac{\partial T}{\partial z} \right), \quad (4)$$

$$\frac{\partial}{\partial r} \left(\frac{1}{r \varepsilon_k} \frac{\partial r B}{\partial r} \right) + \frac{\partial}{\partial z} \left(\frac{1}{\varepsilon_k} \frac{\partial B}{\partial z} \right) + \frac{\omega^2}{c^2} B = 0 \quad (5)$$

The electric field components were determined from Maxwell's equations:

$$E_r = \frac{i}{\omega \mu_0 \varepsilon_0 \varepsilon_k} \frac{\partial B}{\partial z}, \quad E_z = \frac{i}{\omega \mu_0 \varepsilon_0 \varepsilon_k} \frac{1}{r} \frac{\partial r B}{\partial r}$$

Here, we have introduced the following notation: $\vec{V} (V_r = v; V_\phi = 0; V_z = u)$ is the velocity; T is the temperature; p is the excess in the pressure above atmospheric pressure; $\omega = 2\pi f$ is the circular frequency; ρ , λ , C_p , η , φ_e are the mass density, thermal conductivity, specific heat, viscosity, emissivity, respectively; $\varepsilon_k = \varepsilon - i\sigma/\omega\varepsilon_0$; $\varepsilon = 1 - \sigma/\varepsilon_0\nu_e$ is permittivity; $\sigma = K_\sigma e^2 n_e \nu_e / m_e (\omega^2 + \nu_e^2)$ is the electric conductivity with K_σ being a kinetic correction (a given function of the ratio between the collision frequencies of electrons with ions and atoms in a constant electric field [1]); ν_e , n_e , e , m_e are the effective collision frequency, electron density, the charge and mass of an electron, respectively; c is the speed of light; ε_0 and μ_0 are the permittivity and permeability of free space; i is the imaginary unit; and the asterisk stands for complex conjugate.

Boundary conditions The following conditions were imposed at the boundary of the calculation region. It was assumed that the gas-dynamic velocity was zero at solid surfaces and that the gas penetrated freely through the wall of the outer electrode of radius R_e . The gas flow in the entrance cross section of the inner electrode was described by the Poiseuille law, and the pressure in the outlet cross section was set equal to atmospheric one. When calculating the temperature, we specified the temperature at the outer boundary, except for the outlet cross section, where the gas was assumed to be in thermal equilibrium. The magnetic induction was calculated assuming that the electrodes were perfectly conducting (the electric-field component tangential to the wall was set at zero). In the entrance cross section, through which the TEM

wave was supplied, and in the exit cross section, the emission conditions were imposed. The boundary conditions were formulated as follows:

$$Z = z_{\min}, R_{i2} < r < R_{e1}: \frac{\partial B}{\partial z} - ikB = -2ikB_l \frac{R_{i2}}{r} \exp(-ikz);$$

$$Z = z_{\max}, 0 < r < R_e: \frac{\partial B}{\partial z} + ikB = 0;$$

We assume that, for a given frequency, the radial dimensions R_{i2} and R_{e1} of the coaxial waveguide allow the propagation of only TEM waves, $B = B_l \frac{R_{i2}}{r} \exp(-ikz) + B_R \frac{R_{i2}}{r} \exp(+ikz)$:

the incident wave with the amplitude $B_l = \sqrt{\frac{\mu_0 \omega P_l}{\pi k c^2 R_{i2}^2 \ln(R_{e1} / R_{i2})}}$ and power P_l in the entrance

cross section and the reflected wave with the amplitude $B_R = \frac{\partial B / \partial z + ikB}{\partial B / \partial z - ikB} \Big|_{R_{i2} < r < R_{e1}}$ and power

$P_R = \frac{B_R B_R^*}{B_l B_l^*} P_l$; $k = \omega/c$. The conditions in the exit cross section correspond to a transmitted wave propagating in the positive direction along the z axis, k being the longitudinal wavenumber.

The integral electromagnetic energy balance was verified by using the equation

$$P_l = P_R + P_T + P_d,$$

where the power of the transmitted wave and the dissipated electromagnetic radiation power were described by the expressions

$$P_T = \frac{\pi}{\mu_0} \int_0^{R_e} E_R B^* r dr; \quad P_d = 2\pi \int_0^{R_e} \int_0^{z_{\max}} \frac{1}{2} \sigma (E_R E_R^* + E_z E_z^*) r dr dz.$$

To solve the above set of equations numerically, we discretized it by the control volume method on a rectangular nonuniform mesh. For the velocity components, we used shifted meshes, and the pressure field was calculated by using the SIMPLER correction method [2]. At every step of the iteration procedure, discrete analogs describing corrections to the dependent variables were found by the Gaussian elimination method. As a criterion for the termination of calculations, we used the limitation condition (no larger than 10^{-5}) on the Euclidean norms of the residual vectors (composed over all the control volumes) of discrete analogs for the sought-for dependent variables.

Results

Characteristics of a microwave discharge in were calculated for the following parameters, which corresponded to the experimental conditions of [4]: $f = 2,45$ GHz, $R_{i1} = 1$ mm; $R_{i2} = 2$ mm; $R_e = 10$ mm. Coefficients for an equilibrium plasma of atmospheric-pressure argon were taken from [1]. The particle densities were calculated from equilibrium; the electron collision

frequency was calculated by the formula $\nu_e = \sqrt{\frac{8k_B T}{\pi m_e}} (n_d Q_{ea} + n_i Q_{ei})$, where the collision cross

sections are defined as functions of the temperature and density [1]. The results of calculations at $l_i = 3\lambda/4$, ($\lambda = 2\pi/k$); $P_l = 800$ W; $G_0 = 10$ l/min are presented in Figs. 2 – 7.

Thermal and Electromagnetic Structures. A microwave discharge is excited near the end of the inner electrode (see Figs. 2, 3), where the electric field is maximum (Figs. 4, 5). In the gas jet emergent from the inner electrode, the head of the plasma torch forms. The torch head is separated from the nozzle exit by a cold gap (Figs. 2, 3), which provides thermal insulation of the inner electrode. (It follows from experiments with atomic gases that the nozzle heats but little.) Downstream of the torch head, the isotherms diverge due to heat conduction and convection and then converge as the surrounding gas is heated and the heat source power decreases due to a decrease in the dissipation of the electromagnetic field. The electromagnetic

field extends to the region occupied by a conducting plasma, which provides the propagation of electromagnetic waves along the torch and a gradual conversion of electromagnetic energy into heat. It can be seen from Figs. 4-6 that a rather complicated structure of the electromagnetic field forms downstream from the end of the inner electrode. We can distinguish the region of dissipation of the electromagnetic field energy (Figs. 3, 5) - the region where the axial component of the electric field and the radial component of the Poynting vector are dominant, and the region where the radial electric field and the axial electromagnetic energy flux are dominant. The electromagnetic field structure is periodic along the discharge axis. The shape of the contour lines of the electric field strength indicates that the electric field is concentrated near the boundary of the dissipation region.

This picture corresponds to the calculations in which the gas flow rate was gradually increased to a given value. When we start with the same "zero" approximation and a fixed flow rate of 10 l/min, the temperature field downstream from the nozzle acquires a W shape (Fig. 7). The formation of this picture may be explained by two factors: first, the intense convective cooling of the frontal region of the torch by an incident cold gas flow (less than 10% of the flow rate G_0). This fresh gas heats due to heat conduction (Fig. 7), penetrates into the discharge region ($\sim 1\%$ of G_0) and expands downstream; second, the character of distribution of heat sources for a radially converging flux of electromagnetic energy. In our case, the second effect is realized.

Gas-Dynamic Structure. It can be seen from Figs. 2 and 3 that the gas flow structure corresponds to a situation in which the gas flows around the discharge region. In this case, the major portion (90%) of the cold gas emergent from the inner electrode flows around the high-temperature region (the 4-kK isotherm). Near the end of the inner electrode, there is a pressure peak on the axis. At the end surface of the inner electrode, the pressure is lower, so the ambient gas flows into this region (Fig. 2). Evidently, such a gas-dynamic structure enveloping the dissipation region of the electromagnetic field favors the spatial stabilization of the discharge: the location of the torch head is fixed, and the transverse dimensions of the torch are constant along the axis.

As the input microwave power increases or the gas flow rate is reduced, the length of the energy dissipation region (from the head, along the torch axis) increases.

Influence of the Length of the Inner Electrode. It is found that, for an inner electrode length of $l_i = \lambda/2$ (Fig. 1), the numerical solution describes a cold gas flow emergent from the inner electrode into the surrounding space, while dissipation of the electromagnetic field is almost absent and the wave is reflected completely from the beyond-cutoff circular waveguide ($z > l_i$). For $l_i = \lambda/4$, the results of calculations are similar to those obtained for the case $l_i = 3\lambda/4$. These results show that the proper choice of the length of the inner electrode is of crucial importance for the existence of a discharge in the scheme under consideration (Fig. 1); this is consistent with what we observe in the experiment. If the length of the inner electrode is equal to an odd number of quarters of the wavelength, then its end is located in an antinode of the electric field of the standing wave that forms when the running TEM wave is reflected from the beyond-cutoff circular waveguide [1]. Such a length of the inner electrode can be considered as a necessary condition for the excitation of a discharge, though the plasma produced may introduce a certain detuning.

Conclusion

The proposed theoretical model is capable of describing the main electromagnetic and

gas-dynamic properties of a coaxial microwave torch. It is shown that the torch comprises a discharge core and a plasma jet along which a surface electromagnetic wave propagates. The gas flow structure and the spatial distributions of the electromagnetic energy, gas temperature, electric field strength, and magnetic induction have been determined. It is found that the nozzle of the inner electrode is separated from the hot plasma core by a cold gas gap, which provides thermal insulation of the inner electrode. As a result, the nozzle wall is heated only slightly.

However, the proposed simplified model fails to describe some of the experimentally observed effects. Thus, the maximum calculated plasma temperature (8000 K) is significantly higher than the measured one (4000 – 5000 K). Furthermore, the model is incapable of describing unsteady phenomena, such as the generation of bright dense (with high electron densities) plasma objects (plasmoids) that arise near the nozzle at a rate of about 1 kHz and propagate along the flow. To describe these and other features of the coaxial microwave torch, the model has to be modified.

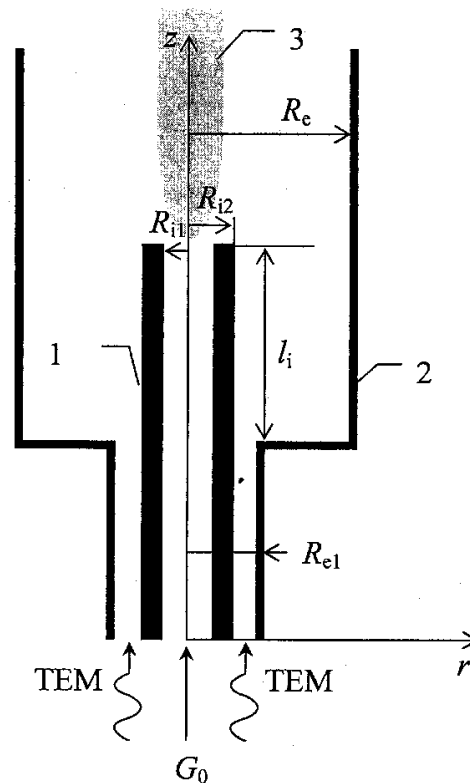


Fig. 1.

Schematic of the calculation region of a coaxial waveguide with a truncated inner electrode: (1) inner electrode, (2) outer electrode, and (3) microwave discharge plasma. The electromagnetic energy is supplied by a TEM wave with a power P , and G_0 is the working gas flow rate. Here, R_e , R_{e1} , R_{i1} , R_{i2} are radii of the outer and inner electrodes, and l_i is the length of the projecting part of the inner electrode.

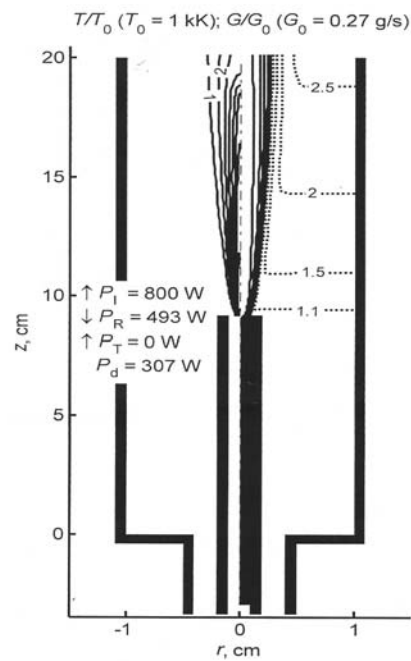


Fig. 2.

Isotherms (to the left of the z axis, with a step of 1.0) and streamlines of the working gas (solid lines to the right of the z axis, from 0.1 with a step of 0.2) and the surrounding gas (dotted lines).

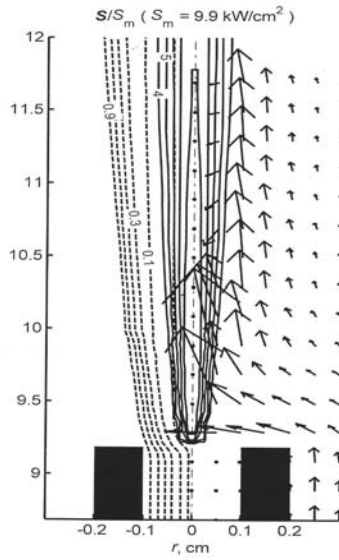


Fig. 3.

Fragment of the gas streamlines (dotted lines), isotherms (solid lines), and Poynting vectors (arrows).

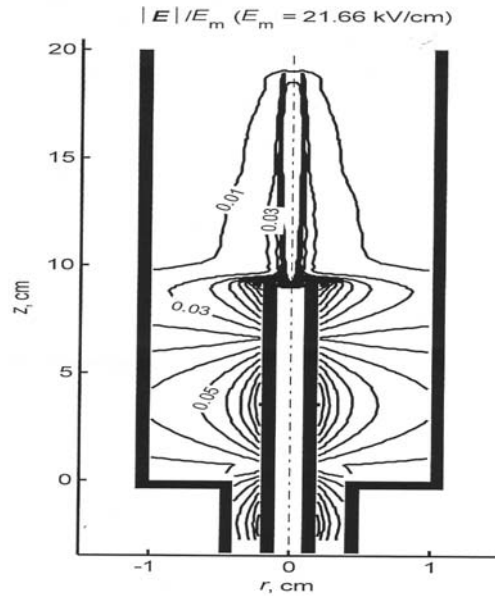


Fig. 4.

Contour lines of the reduced electric field strength (with a step of 0.2).

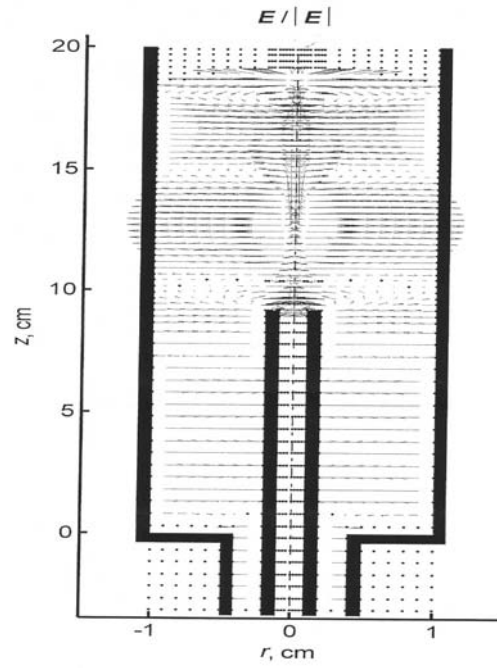


Fig. 5.
Directional field of the electric field strength ($|E| > 0.001 \cdot E_m$).
 $B_m = 1.09 \text{ mT}$

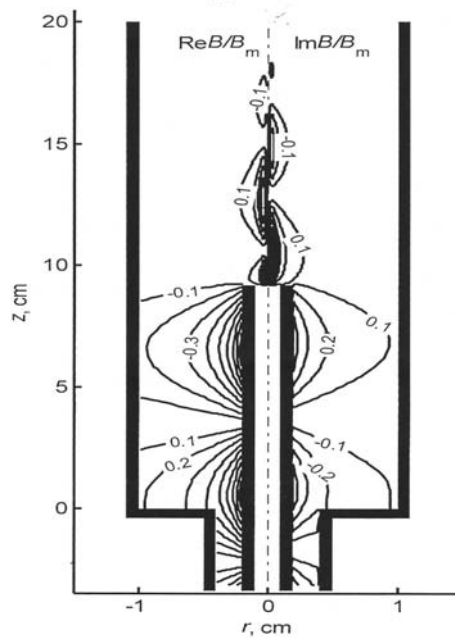


Fig. 6.
Contour lines of the reduced magnetic field induction (with a step of 0.1).

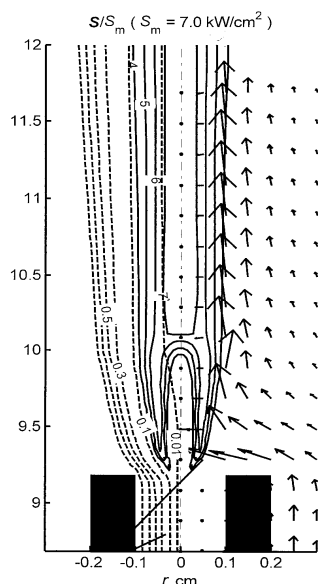


Fig. 7.

Fragment of the gas streamlines (dotted lines), isotherms (solid lines), and Poynting vectors (arrows).

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4. Microwave Torch as a Plasmachemical Generator of Nitrogen Oxides

Microwave coaxial plasma sources (microwave torches) are of interest for various plasma technologies and also may be useful tools in experimental physics [1, 2]. Among the research areas where the use of plasmatrons of this type may be expedient, we first mention plasma chemistry, since such sources offer the prospect of realization of reactions that are impossible or ineffective under equilibrium conditions.

One of the specific features of coaxial microwave plasma torches, apart from a substantial difference between the electron and gas temperatures, is a very efficient and fast heating of the gas flowing through the torch and its subsequent fast cooling (see, [3, 4]).

The operation of such a system in the absence of thermal equilibrium and the possibility of the fast gas heating with the subsequent quenching have motivated interest in microwave-torch plasma generators as plasmachemical reactors.

Analyzing the literature on microwave torches employed as plasmachemical reactors, two works in particular are worthy of special attention. One of them [5] describes the use of a microwave torch to decompose fluorocarbons (namely, CFC-11). In another work [6], a microwave torch was used to study the partial oxidation of liquid hydrocarbons (as a plasma converter of hydrocarbon fuel into a synthesis-gas). Both works confirm a high efficiency of employing microwave torches in plasmachemical processes.

In the present Project, our aim was to investigate a microwave torch as a means for solving the problem of initiating the combustion of high-speed flows of combustible gas mixtures. By introducing radicals, excited molecules, and atoms produced by the microwave plasmatron immediately into the flow, it would be possible to substantially decrease the induction times of the mixture ignition.

As working gaseous media, we intended to use natural combustible gases and their mixtures, and also air. Interest in operation with air flows arose in connection with results of recent studies that have shown the role of nitrogen oxides (NO) for promotion of the combustion process [7, 8]. This circumstance was taken into consideration in the choice of the experimental layout for our first experiment. A plasmachemical process under study was the nitrogen oxide production by using air flow as the working gas in the microwave torch plasmatron.

The design of the microwave torch used in our experiments has some important characteristic features and advantages over other known discharge systems (see, e.g., [1]). The construction is described in details in [3, 4]; here, we only outline its characteristic features. A microwave torch discharge is represented schematically in Fig. 1.

The torch designed at the GPI allows us to achieve a stable generation of the plasma jet near the nozzle by using a production magnetron with a power of $P < 1$ kW and a radiation frequency of 2.45 GHz. The increase in electric field at the nozzle up to the threshold intensity ensures that the torch can operate with almost all gases and gas mixtures (air, argon, nitrogen, methane, etc.). A nonabsorbed fraction of electromagnetic energy returns to the cavity after reflecting from the beyond-cutoff waveguide, which increases the energy absorption coefficient of the torch plasma. A very low level of microwave radiation in the surrounding space ensures the radiation safety of this device.

The microwave torch of this type was studied as a plasmachemical reactor for production of NO_x ($x = 1, 2$). The experimental setup is shown in Figs. 2a, 2b. Air at atmospheric pressure passes through the central hollow electrode, which is an inner electrode of the coaxial waveguide. We used a nozzle with a channel diameter of nearly 1.6 mm (the length was 10 mm); the gas flow rate was $Q = 10$ l/min and the flow velocity at the nozzle exit was 83.3 m/s. A maximal gas flow rate in our experiments did not exceed $Q \leq 40$ l/min.

Treated by discharge, the gas flowed into the chamber open at its end. After short-time operation of the torch, the concentration of produced oxides in the chamber came to a steady-state level, and their percentage was exactly defined by their production in the jet. Downstream from the end of the torch, we introduced a ceramic pipe to take gas samples. The gas-analyzing system consisted of a spectrometer unit for measuring the NO_2 concentration and a Spectra-1600 GL gas analyzer measuring the NO concentration to within 10% of a measured value. The gas analyzer, using the electrochemical effect for its operation, has a built-up pump drawing the sampled gas through the entire diagnostic system. The spectrometer unit consists of a deuterium lamp, having a continuous spectrum in a wide optical region, a tube with transparent ends for transportation of the sampled gas, and an S2000 (200 – 800 nm) or HR2048 (300 – 400 nm) spectrometers measuring the spectrum of the lamp radiation passed through the tube. The NO_2 concentration was measured by radiation absorption.

In the experiments we varied the gas flow rate determining the released energy density in the gas.

The results of these experiments are presented in Fig. 3.

It can be seen that that the concentrations of produced NO and NO_2 are comparable and amounts to several tenth percent. As the gas flow rate was increased to a maximum attainable value, the oxide production decreased. At the same time, the energy efficiency (ε) of NO_x production changed insignificantly: from 100 eV/molec at $Q = 40$ l/min to 75 eV/molec at $Q = 40$ l/min. Hence, as the gas flow rate (and, consequently, the released energy) increased by a factor of 2—2.5, the value of ε decreased by only 25%.

We carried out a series of theoretical studies directed toward the construction of a physical and plasmachemical model of a reactor using a microwave torch for its operation. We considered the NO_x production in the CMPT discharge in air at atmospheric pressure. The discharge channel is a axisymmetric plasma of radius $R \sim 1$ mm [3, 4]. At a distance of $L_d \leq$

1.5 – 2 cm from the electrode, the gas in the channel is heated to temperatures exceeding 4000–4500K [4]. Under these conditions, most of the discharge energy is expended for exciting vibrational degrees of freedom of nitrogen molecules. At a gas temperature $T \geq 4000$ K, the rate constant for VT relaxation of vibrational excitation of $N_2(v=1)$ by oxygen atoms exceeds $k_{VT} \geq 10^{-12}$ cm³/s [5], whereas the equilibrium density of $O(^3P)$ atoms in air at atmospheric pressure amounts to $\sim 5 \cdot 10^{17}$ cm⁻³ [6]. Hence, the characteristic time $\tau_{VT} \cong 2$ μ s of vibrational relaxation of $N_2(v)$ is much smaller than the discharge pulse duration, and the action of the discharge at times $t \gg 2$ μ s may be reduced to gas heating. For this reason, the discharge under study may be considered as a heat source with a given specific power.

As was mentioned above, the air is admitted into the discharge plasma through the central electrode. Intense ionization and heating of the gas occur in the region of strong field near the electrode. The formed hot plasma is then entrained by the gas flow; hence, the electromagnetic field energy is expended only for maintaining the conductivity and for an additional heating of the created channel region. Consequently, the discharge action (i.e. the existence of the gas unit volume in the region of microwave action) depends on the length of the microwave absorption region L_d and the gas flow velocity. The gas flow velocity was assumed to be equal to a value of the axial velocity of discharge propagation, which was measured experimentally [3]. The obtained velocity values depend linearly on the gas flow rate Q and amount to several ten meters per second. For $L_d = 2$ cm, the discharge action time is estimated as $\tau \cong 0.4 - 1$ ms.

In calculations of the nitrogen oxide production in the microwave torch discharge, we used a one-dimensional axisymmetric model. The radial profile of the specific power of a heat source was assumed to be Gaussian:

$$W_T(r) = W_T^0 \exp\left[-\left(\frac{r}{R_0}\right)^2\right], \quad (1)$$

and the time dependence $W_T^0(t)$ was known. The parameter of the problem was a value of the initial radius R_0 .

The kinetic block of the model included a system of processes describing the evolution of concentrations of dominant neutral components of a hot nitrogen – oxygen mixture: $N(^4S)$, $O(^3P)$, O_3 , NO , NO_2 , O_2 , N_2 . The main reactions and dependences of the rate constants on the gas temperature were taken from [7, 8].

For every component, we solved the equation:

$$\frac{\partial N_k}{\partial t} + \frac{1}{r} \frac{\partial (uN_k)}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} (rD_k \frac{\partial N_k}{\partial r}) + F_k^+ - F_k^-. \quad (2)$$

where N_k is the concentration of the k -th species, D_k is the diffusion coefficient, F_k^+ and F_k^- account for the production and loss of these particles, respectively.

In modeling the radial expansion of the hot gas channel, we used the set of one-dimensional (axisymmetric) time-dependent equations

$$\frac{\partial p}{\partial t} + \frac{1}{r} \frac{\partial (ur)}{\partial r} = 0 \quad (3)$$

$$\frac{\partial \rho u}{\partial t} + \frac{1}{r} \frac{\partial (\rho u^2 r)}{\partial r} + \frac{\partial P}{\partial r} = 0 \quad (4)$$

$$\frac{\partial \rho E}{\partial t} + \frac{1}{r} \frac{\partial (\rho u r (E + P))}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} (r\lambda \frac{\partial T}{\partial r}) + W_T \quad (5)$$

where ρ , u , P are the mass density, velocity, and pressure of the gas; $E = \varepsilon + u^2/2$, and ε is the internal energy in unit volume of the gas. Dependence $\varepsilon = \varepsilon(P, T)$ is determined by the equation of state of a given gas. For air in a wide range of parameters, we have $\varepsilon = P/\rho(\gamma-1)$, where $\gamma = C_p/C_v$ is the effective adiabatic exponent. The heat conductivity coefficient for air (the first term in the right-hand side of Eq. (5)) was taken from [13].

The set of Eqs. (3) – (5) was solved numerically on a mesh uniform on r (by using a modified Mc Cormack method [10] of second order of accuracy over space and time), simultaneously with equations of chemical kinetics (2).

To verify the kinetic block of the model, we performed calculations of the temperature dependence of the equilibrium concentration of nitrogen oxides in air at atmospheric pressure (Fig. 4). The results of simulation were compared with data [10]. Comparison was also performed for other components of the air mixture: $N(^4S)$, $O(^3P)$, O_2 , N_2 . The results of comparison show that the elaborated kinetic model adequately describes the equilibrium concentrations in hot air [10] in the temperature range $T = 2500$ — 7000 K.

Simulation of the NO_x production in the microwave torch discharge was performed at a given discharge power per unit length (400 W/cm), known from the experiment. In calculations, the microwave absorption in the plasma was varied from 70 to 100%. It was assumed that all absorbed power is converted into the gas heat.

The important result of simulation is a weak dependence of the energy efficiency of NO_x production on the discharge action time τ . The reason is that not only the released energy increases with τ , but also the radius of the heated region increases, resulting in an additional production of nitrogen oxides at the periphery of the discharge channel. Assuming that $\eta = 90$ % (η is the fraction of microwave power absorbed by the plasma), the calculated value of the energy cost of the nitrogen oxide production is estimated as $\varepsilon = 75 \pm 5$ eV/molec(NO_x) for action times $\tau = 0.5$ – 2 ms. These values agree with the experimental data (see the table) on the energy cost ε and its dependence on the discharge action time (which is determined by the gas flow rate).

The analysis of experimental and theoretical results led us to the following conclusion. From the standpoint of nitrogen oxide production, the microwave torch may be considered as a system in which the absence of thermal equilibrium is of little consequence for the process, even though the electron temperature differs considerably from the gas temperature (see [4]). The decisive factor in the process of NO_x production is air heating to temperatures 3500 – 4000 K with the subsequent quenching of reaction products. The energy cost of the nitrogen oxide production achieved in this system is much lower than its values obtained experimentally (and predicted theoretically) for nonequilibrium-plasma of microwave discharges operating with relatively cold ($T \leq 800$ K) air (see [14]).

According to [15], high energy efficiency of the nitrogen oxide production ($\varepsilon = 3$ – 10 eV/molec (NO_x)) is best achieved by employing microwave discharges. However, such record levels may only be achieved when discharge parameters are varied within a relatively narrow range, at pressures of several ten Torr. Our reactor developed on the basis of a microwave torch discharge, even though it ranks below these systems in efficiency, is superior to them in performance because operates at atmospheric pressure. Compared to many other discharge systems, our torch is less expansive and ensures a high (up to 60%) efficiency in operation from the main.

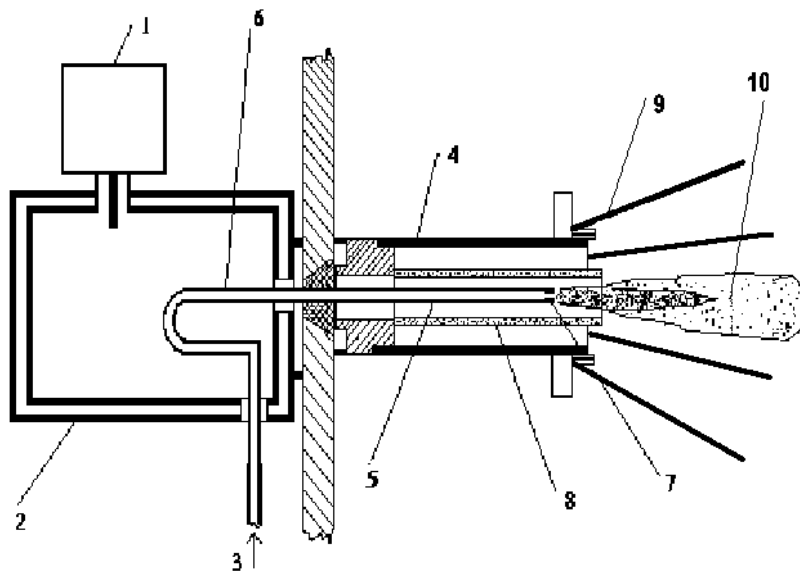


Fig. 1.

Schematic of the microwave torch:

- (1) magnetron, (2) rectangular cavity, (3) injection of the working gas, (4) outer electrode of the coaxial waveguide, (5) inner electrode, (6) magnetic loop, (7) nozzle, (8) quartz tube, (9) copper rods, and (10) plasma jet.

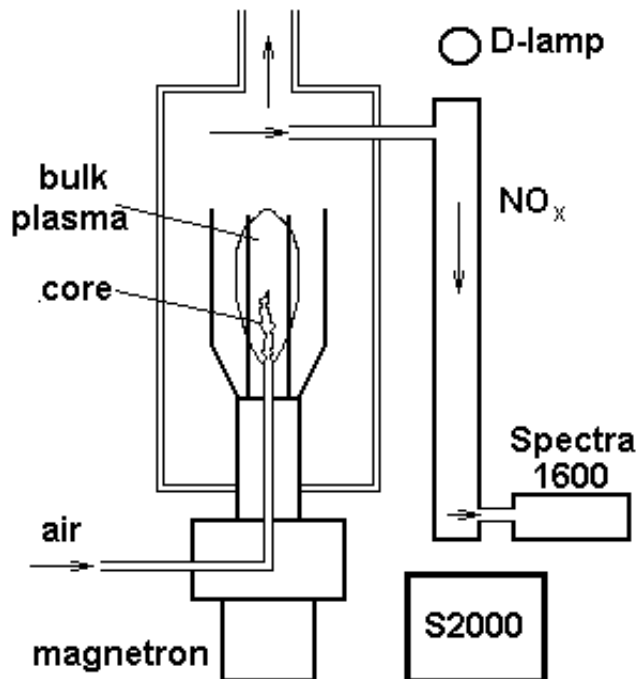


Fig. 2a.

Experimental layout.



Fig. 2b.

Photograph of the setup.

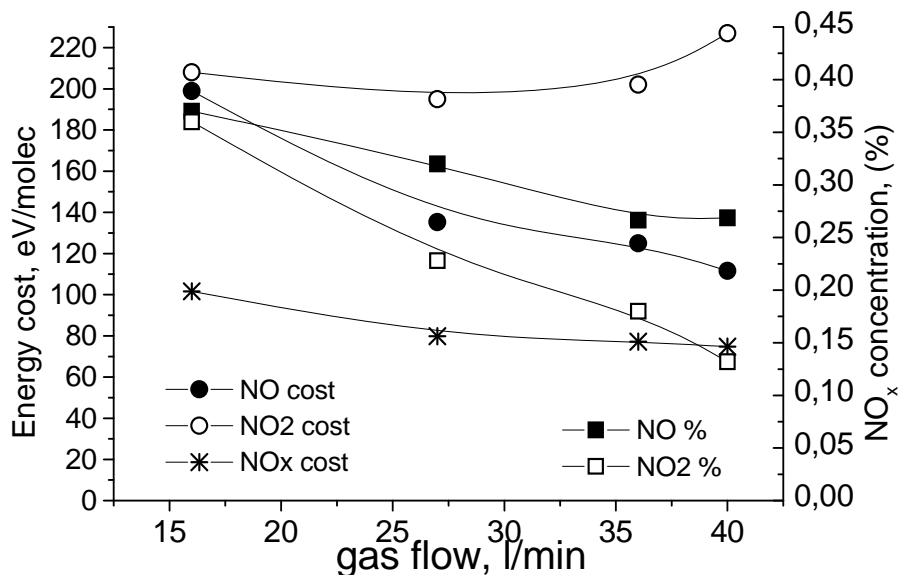


Fig. 3.

Concentration of NO_x and energy cost as functions of the gas flow rate.

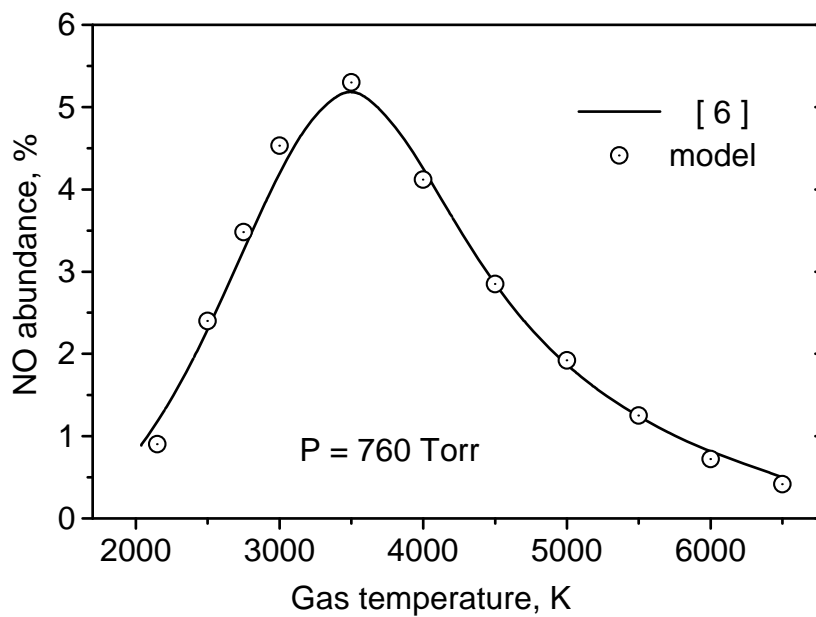


Fig. 4.

NO equilibrium content in air at 1 atm vs gas temperature. The curve corresponds to data [10], points show results of our calculations.

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5. Destruction of Methane in a Microwave Torch

Destruction of methane passing through a microwave torch was investigated using the experimental setup shown schematically in Fig. 1. As a working gas for the microwave torch, we used an Ar:CH₄ gas mixture in a variable proportion as 6:X (l/min), where X= 1, 2, ..., 6.

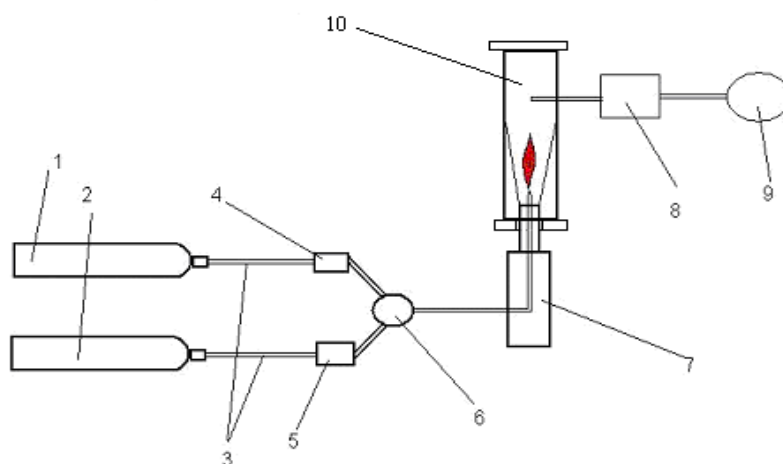


Fig. 1.

Schematic of the plasmachemical reactor using a microwave torch:
(1) argon cylinder; (2) methane cylinder; (3) gas pipeline for the components of the working gas mixture; (4), (5) flowmeters; (6) mixer; (7), (10) microwave torch and the reactor chamber; (8) SPECORD IR spectrograph; and (9) pump for gas sampling. The working gas mixture is Ar : CH₄.

The methane content in the mixture flowing into the torch and passed through the microwave torch was measured by optical absorption technique with the help of an SPECORD IR spectrograph. The hydrogen content was determined by chromatographic analysis. The experimental results are presented in Fig. 2., which shows the energy cost of decomposition of the CH₄ molecule as a function of X (the flow rate of the methane component).

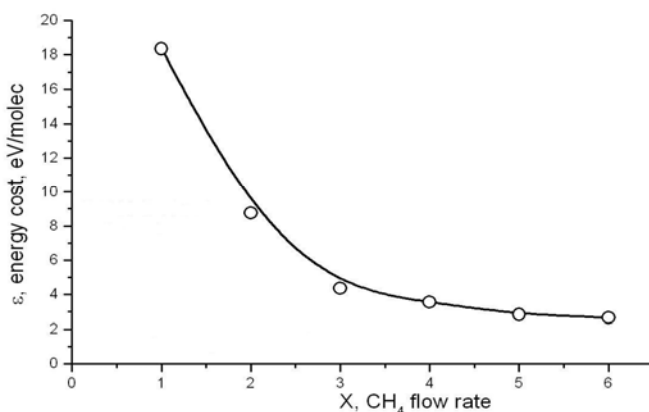
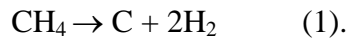


Fig. 2.

Energy cost of decomposition of the CH₄ molecule as a function the flow rate of the methane component.

At a relatively high content of methane, the energy cost of decomposition of the CH₄ molecule is estimated as $\varepsilon \approx 2$ eV/molec. This value is close to the enthalpy of activation of the destruction reaction for CH₄



6. Microwave Torch as an Initiator of Combustion in Premixed Kerosene-Air Streams

In Section 3 of the present report, we describe a mathematical model of a microwave torch developed in the course of project implementation. The results of calculations are presented in Figs. 2 - 7 of Section 3

It can be seen from the figures that an electromagnetic wave initially propagates through the coaxial waveguide formed by the inner and outer electrodes. In the absence of a flame, this wave is reflected from the beyond-cutoff coaxial waveguide which begins downstream from the nozzle (at the right of the left figure). In the presence of the flame, the electromagnetic wave can propagate along the axis, as the plasma acts as an inner electrode. However, the wave mode changes radically: the axial wave is converted into the surface electromagnetic wave. The Poynting vector acquires a radial component toward the axis, and the wave energy is concentrated near the system axis. The electric field strength drops substantially with distance from the plasma, in both the axial and radial directions. This structure is typical of a surface electromagnetic wave. The plasma sustained by the energy of this wave is, in turn, a guiding structure for propagation of this wave.

Note that the above structure differs somewhat from the often used and well-studied sufratron structure. For the latter, the interface where an electromagnetic wave is converted into a surface wave is a sharp boundary between the plasma and dielectric (dielectric wall, tube surface). In our case, the interface is a spatially (radially) extended boundary between the high- and low-density regions. The high-density plasma forms the body of the torch, whereas the low-density plasma appears as a photoionized "aureola". The excitation of the surface wave requires a sufficiently large gradient of the plasma density.

Let us discuss another result of our calculations. Since the wave energy is concentrated about the axis and the electromagnetic field at the outer electrode drops substantially, the outer electrode can be truncated without substantially affecting the plasma--field structure. This means that a short section of the beyond-cutoff circular waveguide (with the length being on the order of the diameter) acts as a converter of a coaxial electromagnetic wave into a surface wave. The conversion does not take place if the condition that the outer electrode is longer than the inner electrode is not fulfilled. For example, this is especially the case with TIA-design torches [1], in which the generation of the surface wave does not take place and a plasma exists only in a small region near the end of the coaxial waveguide.

This essential difference is understood by reference to Fig. 1.

Figure 1a shows a TIA-design torch in which the inner and outer electrodes are almost equal in length. Figure 1b shows the structure of a microwave coaxial torch with a longer outer electrode. This modification has the evident advantage, in particular, the radiation safety. A nonabsorbed fraction of microwave energy transmitted through the torch is reflected from the beyond-cutoff waveguide, returns to the torch plasma and interacts with it. Figure 1c shows the structure of a microwave coaxial torch with a truncated outer electrode. Structurally, this version is similar to Fig. 1b, but the radiation safety is not so evident in this case. The plasma jet, when being outside the coaxial waveguide can emit electromagnetic waves. Furthermore, such emission reduces the fraction of absorbed electromagnetic energy. Nevertheless, the fraction of microwave energy emitted into the surrounding space is relatively small, and most of the energy is absorbed by the gas-discharge plasma.

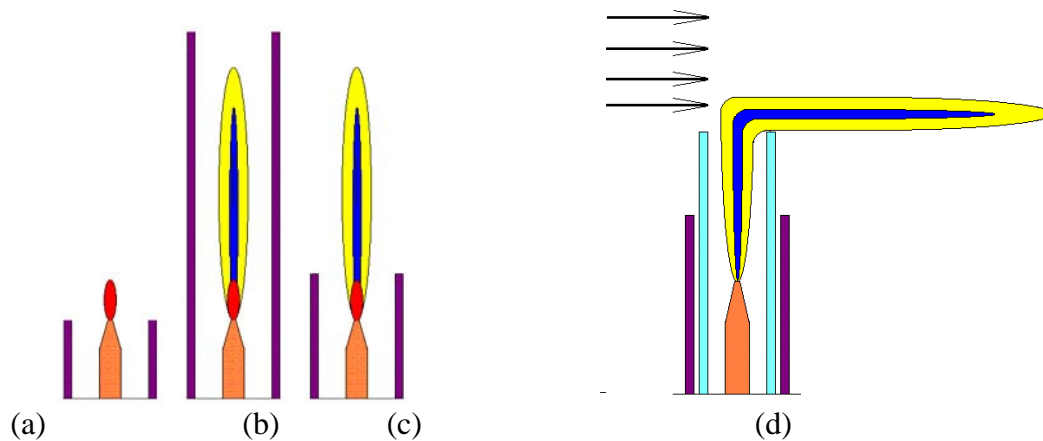


Fig. 1.

Schematic representation of versions of microwave torch: (a) TIA-design, (b) microwave coaxial torch with a longer outer electrode, (c) the same torch with a truncated outer electrode, and (d) microwave torch in a gas stream.

When the plasma jet of the torch is introduced into the combustible gas flow, this jet is entrained by the flow. However, the torch structure is retained: the torch only becomes bent by the flow (see Fig. 1d). A quartz tube is added with the aim to avoid the arcing of the torch plasma onto the outer electrode of the coaxial waveguide. A photograph of the microwave torch introduced into a supersonic air stream in the experiment carried out in the testing chamber of the Moscow State University (Fig. 2) agrees well with Fig. 1c.

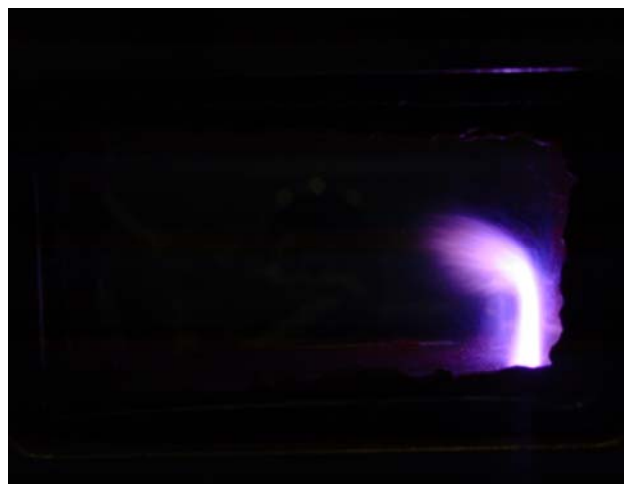


Fig. 2.

Photograph of the microwave torch introduced into supersonic air stream.

When the torch is introduced into a gas flow (Figs. 1a and 2), the body of the torch in its lower part (near the nozzle) has little effect on the state of flow, in particular, on the state of the fuel--air mixture when we attempt to ignite it with the help of the plasmatron positioned outside the flow. At the same time, the torch region far from the nozzle is invaded by the combustible gas and can affect the combustion process. The following factors may contribute to activation of the fuel-gas mixture: a high temperature of the torch, the production of chemically active atoms and molecules, and also UV radiation penetrating the flow. As a result the induction times may be reduced substantially, and that the interaction of the torch with the fuel--gas flow may provide ignition.

The possibility of using the microwave torch as an igniter was tested in experiments carried out in a ring flame stabilizer at the Central Institute of Aviation Motors (CIAM) [2, 3]. The experimental setup is shown in Fig. 3.

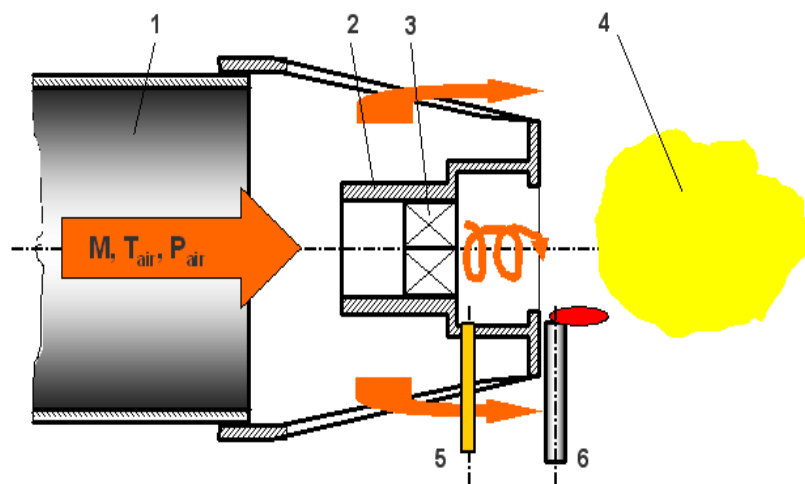


Fig. 3.

Experiment at the CIAM:

(1) air nozzle, (2) frontal nozzle, (3) air stream swirler, (4) flame, (9) tangential kerosene inlet, and (6) microwave torch.

An air flow heated to temperature T_{air} and having velocity M (the Mach number relative to normal conditions) passes through the frontal nozzle into an air stream swirler. At the swirler outlet an atomized kerosene is tangentially injected in the opposite direction to the jets of swirled air stream. This system ensures good mixing of the fuel with air. A microwave torch is placed near the exit of the nozzle. The plasmatron position relative to the axis is adjusted so that the plasma jet from the plasmatron should get to the kerosene--air mixture flow and should be entrained in the axial direction, as shown in Fig. 3.

The effect of ignition and burning of the mixture was assessed in visual observations .

In the experiment, the velocity and temperature of an air flow were initially regulated, and then a small mass of a fuel (kerosene) was injected through inlets situated around the stream. If the switching-on of the microwave torch did not result in the mixture ignition, we increased the fuel mass. When we managed in the ignition of combustion, even if unstable, a corresponding point (a red rhombus) was laid at the plot in Fig. 4. An increase of the fuel mass led to stabilization of the combustion. The corresponding event of an extra fuel was designated by a closed rhombus at the plot. The results are presented in Fig. 4.

Note that the attempts to ignite the mixture by using an aviation spark plug failed have not met with success.

The results of testing the microwave torch as an igniter of kerosene--air mixture in the CIAM apparatus may be summed up as follows:

1. Testing the microwave coaxial plasma torch placed downstream from the model frontal unit in the air flow with $M_{\text{air}} = 0.1 - 0.3$, $T_{\text{air}} = 430-490$ K и $\beta = 0.4 - 1.1$ has demonstrated the possibility of effective ignition of kerosene-air mixtures (β is the relative content of kerosene in air).
2. The range of the mixture parameters ensuring stable burning is much wider than that for a standard aviation spark plug. Thus, for a mixture with $T_{\text{air}} = 490$ K and $\beta = 0.5$, a dependable ignition was achieved at $M_{\text{air}} = 0.3$, whereas the standard plug is able to ignite flows if their velocities do not exceed $M_{\text{air}} = 0.1$

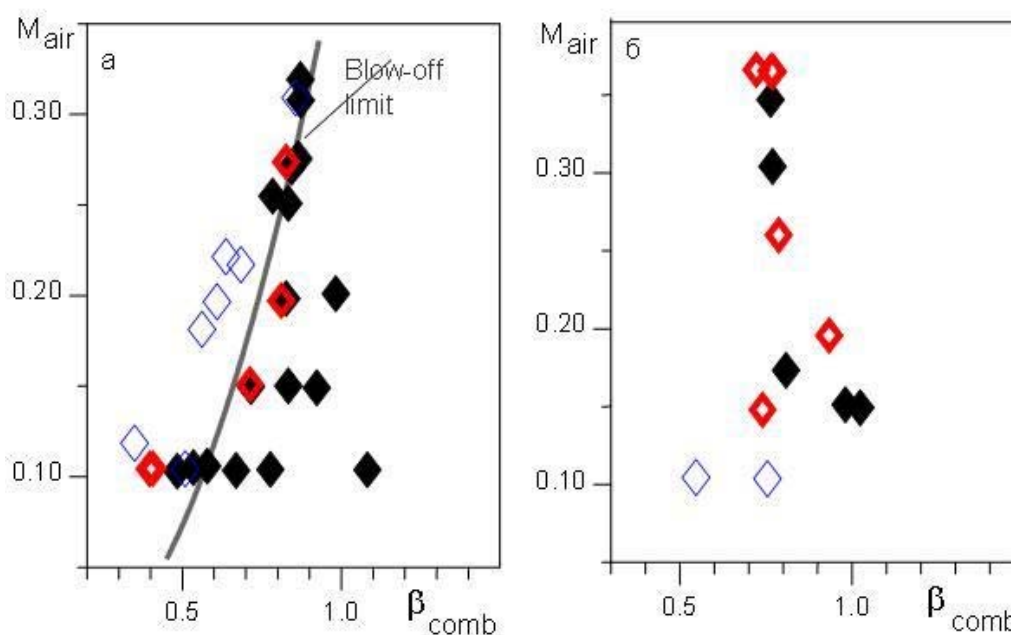


Fig. 4.

Regimes of the ignition and stabilized combustion of kerosene in the ring flame stabilizer with microwave plasma torch initiator: (a) $T_{\text{air}} = 470 - 500$ K, (b) $T_{\text{air}} = 400 - 440$ K; \diamond - without burning, \diamond - unstable burning, \blacklozenge - stable burning.

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6. Conclusion

In the course of project implementation, we have developed and tested the kinetic model, which makes it possible to analyze the role of nonequilibrium excitation in the activation of combustion of oxygen--hydrogen gas mixture. Comparing our calculations with experimental data, we can conclude that the model describes the available experimental results adequately and can be used as a prediction model in our calculations.

A detailed analysis of data available in the literature (in both the field of theory and experiments), which was performed by the participants of the Project and was published in form of a review (see [8] in Appendix 1), as well as calculations in our kinetic model allowed us to distinguish main processes of nonequilibrium action and to estimate the role of the excess density of chemically active particles in accelerating the combustion processes in gas mixtures. The role of oxygen and hydrogen atoms, electronically excited $O_2(a^1\Delta_g)$ molecules and vibrationally excited H_2 and O_2 molecules with densities above the equilibrium density was examined.

A major part of research work over the course of reporting years was devoted to the initiation of combustion of gas mixtures by using the microwave coaxial discharge known as the "microwave torch". The construction of the torch developed and tested at the GPI has a number of special features and the advantages over some versions previously described in the literature. This ensures that the problem of plasma-assisted combustion of gas flows can be included into a wide range of applications.

A theory has been developed for a microwave coaxial torch discharge. Using a self-consistent two-dimensional magnetogasdynamic model, we have calculated characteristics of an equilibrium microwave discharge in an argon atmospheric-pressure jet in a coaxial waveguide with a truncated inner electrode. One important conclusion was reached: in the plasmatron of this kind, the plasma jet act so as to extend the truncated central electrode, and the fundamental coaxial mode is converted into a surface wave that propagates along the torch and, in so doing, interacts weakly with the outer electrode of the coaxial waveguide. Based on the concept developed by our theoretical model, we have constructed a microwave torch intended for use as a stimulator of ignition of kerosene--air jets. The construction is optimized so that the coaxial system is located in the cold-gas region, outside the fuel--air stream.

The construction of the initiator of combustion in a kerosene-air mixture flow, which was developed and assembled at the GPI, was tested in a ring flame stabilizer at the Central Institute of Aviation Motors (CIAM). The initiator is placed behind a model frontal device of a turbojet combustor. The air-flow parameters were $M = 0.1\text{--}0.3$ and $T_{\text{air}} = 430\text{--}490$ K. The equivalent ratio of components of preliminarily mixed kerosene--air mixture was $\beta = 0.4\text{--}1.1$. The ignition by a standard aviation spark plug was compared with ignition by the microwave torch. It was shown that the stable combustion by using the microwave torch is achievable over a more extended range of the flow parameters and mixture-component ratios as compared to the ignition by the plug.

What is the key factor (temperature, plasma chemistry, or photoinduced processes) determining the advantages of the coaxial microwave plasmatron? The question remains an open question and calls for further investigation. In the course of project implementation, we have accomplished a series of studies for determining plasmachemical characteristics of the microwave plasmatron. We have carried out experimental studies of the microwave torch as a plasmachemical reactor of nitric oxides in an air gas jet. A plasmachemical model of the

generator has been constructed. The results of calculations in this model are consistent with the experimental data. Comparison of the experimental results with calculations allowed us to identify mechanisms responsible for high efficiency of the NO_x production in the torch. The role of nitric oxides in stimulating the processes of ignition of fuel-air mixtures was considered.

Experiments with an Ar:CH₄ mixture used as a working gas have demonstrated a very high efficiency of plasmachemical decomposition of methane in the coaxial torch. The microwave torch, in which hydrogen produced by destruction of methane becomes added to a fuel-air mixture, holds promise as an alternative to a standard plug.

Worthy of note is the peculiar feature of the torch as a plasmachemical reactor: the gas heats very rapidly to 5000--7000 K and then cools also rapidly to 2000--3000 K, with the corresponding quenching of the decomposition and oxidation products.

Appendix 1: List of published papers and reports (with abstracts)

[1] I.A.Kosyĭ, V.P.Silakov

Step Photoionization as a Mechanism Responsible for the Raether Paradox

Plasma Sources Sci. Technol., vol. 14, 2005, pp. 594-598.

An attempt is made to explain the phenomenon known as the Raether paradox, which consists in an abnormally deep penetration of the ionizing radiation of the localized electric discharge into a surrounding high-pressure gas. A theory developed for interaction of thermal-equilibrium radiation with molecular nitrogen is invoked to explain this effect. It is shown that the deep penetration effect is satisfactorily explained by the model taking into account the step ionization of metastables. The model assumes that gas molecules (atoms) can exist in excited electronic states that are stable against different deexcitation processes. Long-lived states can be populated predominantly through upper levels effectively excited by radiation emitted from the electric discharge.

[2] N. K. Berezhetskaya, S. I. Gritsinin, V. A. Kop'ev, I. A. Kosyĭ, D. van Wie

Long-Lived Plasmoids Generated by Surface Microwave Discharges in Chemically Active Gases

Plasma Physics Reports, 2005, vol. 31, No. 10, pp.

The generation of long-lived microplasmoids is observed during the irradiation of a metal-dielectric surface with a high-power microwave beam in a chemically active gas mixture (H₂ + O₂; CH₄ + O₂). The lifetime of these plasmoids substantially exceeds the characteristic recombination and cooling times of plasmoids arising at the target surface in a chemically inactive medium.

[3] I.A. Kosyĭ, V.P. Silakov, N.M. Tarasova, David Van Wie.

Long-Lived Plasmoids Generated by Surface Laser Sparks in Combustible

Gas mixtures.

Plasma Physics Reports, 2006, V. 32, No. 4, pp. 1-3.

The paper presents results of experimental studies of laser sparks generated at the surface of a metal target contacted with a chemically active (combustible) gas. The microscopic plasmoids, which are generated at the target surface and are ejected into the medium, initiate its combustion. It is shown that such plasmoids have an abnormally long lifetime (to 1--1.5 ms), which substantially exceeds the lifetime in a chemically inactive medium.

[4] S. I. Gritsinin, I. A. Kossyi, E. B. Kulumbaev, and V. M. Lelevkin
Calculation of a Coaxial Microwave Torch

Plasma Physics Reports, 2006, V. 32, No. 10, pp.

Parameters of an equilibrium microwave discharge in an atmospheric-pressure argon flow in a coaxial waveguide with a truncated inner electrode are calculated numerically by using a self-consistent two-dimensional MHD model. The results obtained agree satisfactorily with the experimental data.

[5] S. I. Gritsinin, V. Yu. Knyazev, I. A. Kossyi, N.A. Popov
Microwave Torch as a Plasmachemical Generator of Nitric Oxides
Plasma Physics Reports, 2006, vol. 32, No. 6, pp.

The possibility of using a microwave coaxial plasmatron (a microwave torch) as an efficient plasmachemical generator of nitric oxides in an air jet has been studied experimentally. A plasmachemical model of the generator is developed. Results of calculations by this model do not contradict experimental results. A conclusion about the mechanisms governing NO_x production in a plasma torch is drawn by comparing the experimental and calculated results.

[6] I.A.Kossyi, S.I.Gritsinin, V.Yu.Knyazev, N.A.Popov
Microwave Coaxial Torch as Plasmachemical Reactor
In "Strong Microwaves in Plasmas", Ed. by A.G.Litvak, 2006, vol. 2, pp. 773-778, Inst. Appl. Phys., Nizhny Novgorod.

A microwave coaxial plasmatron (a microwave torch) is studied experimentally for use as a plasmachemical generator of nitric oxides in an air jet. A plasmachemical model of the generator is developed. Results of calculations in this model do not contradict experimental results. Comparison of the experiment with calculations affords a basis for a conclusion about mechanisms governing the NO_x production.

[7] N.K.Berezhetskaya, S.I.Gritsinin, V.A.Kop'ev, I.A.Kossyi, David Van Wie
Long-Lived Plasmoids Generated by Microwave Discharges in Combustible Gases

In "Strong Microwaves in Plasmas", Ed. by A.G.Litvak, 2006, vol. 2, pp. 681-685, Inst. Appl. Phys., Nizhny Novgorod.

The phenomenon of generation of long-lived plasmoids in a chemically active gas (H_2+O_2 ; CH_4+O_2) was observed when a metal-dielectric surface was irradiated with a high-power microwave beam.

[8] N.A. Popov

Influence of nonequilibrium excitation on ignition of hydrogen--oxygen mixtures. Review.

High Temperatures, 2007, vol. 45, No. 2, pp.

The paper reviews recent works existent in the literature devoted to the effect of reduction in the induction time of an $H_2:O_2$ mixture under the action of various radicals and excited particles. Analysis is made of experimental and theoretical studies of the role of oxygen and hydrogen atoms, electronically excited $O_2(a^1\Delta_g)$ molecules and vibrationally excited H_2 and O_2 molecules.

Appendix 2: List of presentations at conferences and meetings (with abstracts)

[1] S.Gritsinin, V.Knyazev, I.Kossyi, Yu.Shckhman, V.Vinogradov
Microwave Torch Application for Kerosene-Air Mixture Ignition
VI Int. Workshop on Microwave Discharges, Abstracts, Zvenigorod, Russia, 2006, p. 48.

A microwave initiator of combustion in a kerosene—air mixture (a microwave torch) is placed behind a model frontal device of a turbojet combustor. The air-flow parameters are $M = 0.1—0.3$ and $T_{air} = 430—490$ K. The equivalent ratio of components of preliminarily mixed kerosene—air mixture is $\beta = 0.4 - 1.1$. The combustion by using a standard aviation spark plug is compared with the combustion by using the microwave torch. It is shown that the stable combustion by using the microwave torch is achievable over a more extended range of the flow parameters and the mixture-component ratios as compared to the ignition by the plug.

[2] I.A.Kossyi, N.K.Berezhetskaya, S.I.Gritsinin, V.A.Kop'ev, V.P.Silakov, N.M.Tarasova and D. M. Van Wie
Abnormal Long-Lived Plasmoids Produced by Surface Microwave Discharge or Surface Laser Spark in a Combustible Gas Mixture
44th AIAA Aerospace Sciences Meeting and Exhibit, Reno, Nevada, 2006, Report AIAA-2006-1213.

The irradiation of metal--dielectric surfaces with high-power microwave beams as well as the laser irradiation of metal targets contacted with chemically active gas mixtures ($H_2:O_2$; $CH_4:O_2$) is accompanied by the generation of long-lived plasmoids. The lifetime of such plasmoids substantially exceeds the characteristic recombination times and the cooling times of plasmoids arising at the target surface in chemically inactive gases.

[3] S.Gritsinin, V.Knyazev, I.Kossyi, Yu.Shikhman, V.Vinogradov
Microwave Torch Application for Kerosene-Air Mixture Ignition
In "Microwave Discharges: Fundamentals and Applications", VI Int. Workshop. Proceedings, Ed. By Yu. Lebedev, Yanus-K, Moscow, 2006, pp. 297-302.

A microwave initiator of combustion in a kerosene—air mixture (a microwave torch) is placed behind a model frontal device of a turbojet combustor. The air-flow parameters are $M = 0.1—0.3$ and $T_{air} = 430—490$ K. The equivalent ratio of components of preliminarily mixed kerosene—air mixture is $\beta = 0.4 - 1.1$. The combustion by using a standard aviation spark plug is compared with the combustion by using the microwave torch. It is

shown that the stable combustion by using the microwave torch is achievable over a more extended range of the flow parameters and the mixture-component ratios as compared to the ignition by the plug.

[4] N.K.Berezhetskaya, S.I.Gritsinin, V.A.Kop'ev, I.A.Kossyi, N.A.Popov
Surface Microwave Discharge as Initiator of Combustion in Gas Mixtures
Book of Abstracts of the XXXIII International (Zvenigorod) Conference on
Plasma Physics and Controlled Fusion, Zvenigorod, 2006, p. 259

The paper presents results of experiments in which the combustion in a methane--oxygen gas mixture is initiated by surface microwave discharge. A distinctive feature of this process is the propagation of a primary glow wave which can be characterized as an «incomplete-combustion wave» (the gas temperature is on the order of 1500 K). The propagation of this wave is accompanied by the volume combustion of the mixture reaching the temperature typical of the complete combustion of a CH₄:O₂ mixture (on the order of 3000 K in this final stage)

[5] N.A.Popov, I.A.Kossyi
Effect of Nonequilibrium Excitation of Hydrogen-Oxygen Mixture on
Ignition

45th AIAA Aerospace Sciences Meeting and Exhibit, Reno, Nevada, 2007,
Report AIAA-2007-1031.

The paper reviews the literature devoted to the effect of reduction in the induction time of an H₂:O₂ mixture under the action of various radicals and excited particles. Experimental and theoretical studies of the role of oxygen and hydrogen atoms, electronically excited molecules O₂(a'¹Δ_g) and vibrationally excited H₂ and O₂ molecules are analyzed.

[6] V.A.Vinogradov, Yu.M.Shikhman, S.I.Gritsinin, A.M.Davydov,
A.Yu.Knyazev, I.A.Kossyi

Application of MW Plasma Generator for Ignition of Kerosene/Air Mixture
45th AIAA Aerospace Sciences Meeting and Exhibit, Reno, Nevada, 2007,
Report AIAA-2007-1384.

A microwave initiator of combustion in a kerosene—air mixture (a microwave torch) is placed behind a model frontal device of a turbojet combustor. The air-flow parameters are $M = 0.1—0.3$ and $T_{\text{air}} = 430—490$ K. The equivalent ratio of components of preliminarily mixed kerosene—air mixture is $\beta = 0.4 - 1.1$. The combustion initiation by a standard aviation spark plug is compared with the initiation by the microwave torch. It is shown that the stable combustion by using the microwave torch is achievable over a more extended range of the flow parameters and the mixture-component ratios as compared to the ignition by the plug.

[7] I.A.Kossyi, S.I.Gritsinin, P.A.Guschin, V.Yu.Knyazev, N.A.Popov
Microwave Torch as Tool for an Airflow Chemical Transformation

45th AIAA Aerospace Sciences Meeting and Exhibit, Reno, Nevada, 2007,
Report AIAA-2007-0429.

The construction of a microwave torch developed at the A.M. Prokhorov General Physics Institute is described, and some peculiarities are discussed from the standpoint of its application as an initiator of combustion of combustible gas mixtures. The microwave torch was studied both

theoretically and experimentally as a plasmachemical reactor of nitric oxides in an air jet. The goal of the paper is to determine the main processes of formation of NO and NO₂ molecules that can reduce the induction times for gas mixtures substantially. The first results are presented from studies of the methane decomposition by using an Ar+CH₄ mixture for plasma production. The preliminary results of the use of a microwave torch as an initiator of a kerosene—air mixture in the model frontal device are discussed.

Appendix 3: Information on patents and copy rights (List and describe patents and copyrights which were obtained or may be obtained as a result of the project)
It seems expedient to prepare a patent application for the method of initiating the combustion of kerosene--air flows in aviation engines by using the original design of a microwave torch developed and investigated at the GPI.