OPTICAL CHARACTERISTICS AND PARAMETERS OF GAS-DISCHARGE PLASMA ON MIXTURES OF MERCURY DICHLORIDE VAPOR AND NEON

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The results of studies of the optical characteristics and parameters of DBD (dielectric barrier discharge) plasma on a mixture of mercury dichloride vapor, neon are presented. The following functions were established: EEDF, transport characteristics, specific discharge power losses on electronic processes, electrons' concentration and temperature, rate constants for elastic and inelastic electron scattering on components of the working mixture, depending on the E/N.

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INTRODUCTION

Gas-discharge plasma on a mixture of mercury dichloride vapor with gases is the working medium of exciplex sources of coherent and spontaneous radiation in the blue-green spectral region with a wavelength at maximum intensity (λ_{max}) 557 nm [1 - 12]. The creation of gas-discharge plasma and the excitation of the components of the working mixture was carried out at atmospheric pressure in volume, glow, barrier and surface discharges. The study of its optical characteristics was carried out in radiators with working volumes $\geq 200 \cdot 10^{-6}$ and $\sim 1 \cdot 10^{-6}$ m^3. Such volumes are necessary to create exciplex radiation sources in the blue-green spectral region with large and small values of the radiation power, and which are used to solve various scientific and applied problems [13]. In spontaneous radiation sources, helium was mainly used as a buffer gas. Their design was coaxial, and the radiating zone was the lateral surface. For a number of scientific and technological applications, it is necessary to provide greater radiation density and its uniformity over the cross section of the emitter and use in the working mixture of more "heavy" buffer gases than helium, which have less penetrating power through the walls of the emitter and thereby providing a longer service life [14].

The aim of the study was to identify regularities in the optical characteristics of a gas-discharge plasma on a mixture of mercury dichloride vapors with neon and to determine the partial pressure of neon at which the maximum radiation power in the blue-green spectral range in the emitter, which design was different from coaxial, is reached. In addition, the purpose of the study was to determine the plasma parameters: the electron energy distribution function, transport and energy characteristics, the fractions of the discharge power on the electronic processes, the concentration and temperature of electrons, and the rate constants of the processes of elastic and inelastic scattering of electrons on the components of the working mixture depending on the value of the reduced electric field (E/N is the ratio of the electric field intensity to the total concentration of the components of the working mixture), and also set the value of the parameter E/N, which ensures the maximum contribution of the discharge power into the radiation in the blue-green spectral region with a wavelength at the maximum intensity (λ_{max} = 557 nm).

1. EXPERIMENTAL INSTALLATION AND METHODS OF EXPERIMENT

The working mixture in the atmospheric pressure barrier discharge was excited by a power supply with a pulse-periodic output voltage with the possibility of frequency tuning in the range of 1…20 kHz and the amplitude of voltage pulses within 10…30 kV. The research methodology and the research itself were carried out on an experimental setup, which was described in detail in the article [15].

2. RESEARCH RESULTS AND DISCUSSION

The emission spectra of gas-discharge plasma were studied in the region of partial pressure of mercury dichloride 0.5…2 kPa, neon 5…140 kPa. The voltage, current, and repetition rate of the pump pulses were 20…30 kV, 300…325 A, and 10…20 kHz, respectively.

A characteristic radiation spectrum at a pulse repetition rate of f = 18 kHz for partial pressures of mercury dichloride and neon 1.5 and 140 kPa, respectively, is shown in Fig. 1. Only radiation of the system of electron-vibrational bands of the $B^2\Sigma^+_{1/2} \to X^2\Sigma^+_{1/2}$, υ'=0…5, υ''=9…19 transition of exciplex molecules of mercury monochlodide (HgCl*) [10, 11] is observed with a radiation maximum at a wavelength λ = 557 nm, a steep increase in intensity from the long-wavelength region and a slow decline in the short-wave region. The radiation spectra were interpreted using reference data of the work [18].

The band form and its width at half-height (15…16 nm) are similar to the bands corresponding to the B→X transition of mercury monohalides shown in works in which the creation of a barrier discharge on mixtures of mercury dihalides vapor, helium and other gases was carried out in large sized as well as in small-sized radiators. A sharp increase in the intensity of the radiation in the spectrum was observed from the side of the region with long wavelengths and its slow decrease in the region of shorter wavelengths. The spectra differ in the rate of intensity decrease in the ultraviolet part by the width of the emission band. The radiation spectrum for low partial pressures of neon (< 40 kPa) as compared with partial pressures above atmospheric is wider and extends further into the ultraviolet region.

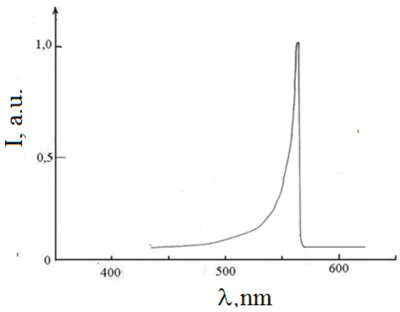

Fig. 1. The emission spectrum of gas-discharge plasma on a mixture of $HgCl_2$: Ne = 0.5…140 kPa. The pump pulse repetition rate is 18 kHz

The results of studies of the dependence of the average radiation power on the partial pressures of the neon buffer gas are presented in Fig. 2. An increase in the radiation power is observed with an increase in the neon partial pressure from 28 to 60 mW/cm^3 and with an increase in the neon partial pressure above 110 kPa, the power does not change until the neon partial pressure increases to 140 kPa.

The results of studies of temporal characteristics of gas-discharge plasma are presented in Fig. 3. Fig. 3,a represent the oscillograms of current pulses of dielectric barrier discharge and radiation, which is shown in Fig. 3,b for the ratio of the components of the mixture, at which the maximum radiation power was reached.

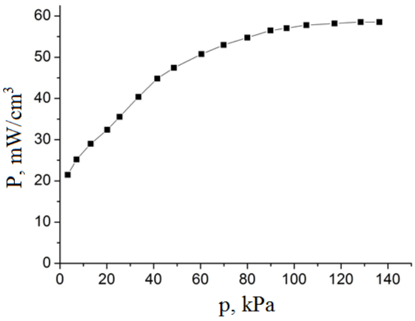

Fig. 2. The dependence of the specific radiation power of a gas-discharge plasma on the partial pressure of neon in the mixture of mercury dichloride vapor and neon. The partial pressure of mercuric dichloride vapor is 1.5 kPa. Amplitude of voltage – 30 kV. The pulse repetition rate is 18 kHz

The maximum values of the amplitude of the current pulse are 325 A. The current pulses are double, of different polarity, the delay between them in our experimental conditions is 150 ns. The leading edge was 10 ns, the duration was 50 ns. The radiation pulses are also double with a time shift relative to each other by 150 ns. Their amplitudes are different in magnitude, the amplitude of the second pulse exceeds the magnitude of the amplitude of the first. The error and reproducibility of the results of oscillographic measurements were 10 and 90%, respectively.

With an increase in the pulse repetition rate, the average radiation power of a gas-discharge plasma increases linearly to frequencies of 20 kHz. The nature of the discharge was similar to that typical of a barrier discharge [19]. With an increase in the pump pulse repetition rate, the radiation intensity of a uniform discharge increased, while the intensity of the filament channels fell. The thickness of the discharge region and the burning length of the discharge were 0.005 and 0.20 m, respectively.

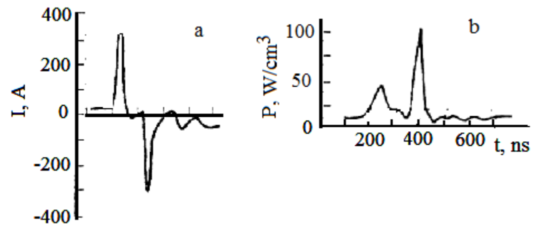

Fig. 3. Oscillograms of discharge current pulses (a) and radiation (b) for the spectral band λ_{max}=557 nm spectral band of HgCl molecules in a $HgCl_2$: Ne mixture. The partial pressure of saturated vapor $HgCl_2$ is 1.5 kPa, the partial pressure of neon is 140 kPa. Amplitude of voltage pulses 30 kV. Pulse repetition rate is 18 kHz*

3. PLASMA PARAMETERS

Due to the fact that experimental physics does not have satisfactory methods for diagnosing dense gas-discharge plasma, the parameters of the barrier-discharge plasma at optimal for obtaining the maximum radiation power of the $HgCl_2$-Ne mixture (0.0107, 0.9893) with a total pressure 141.5 kPa were determined numerically and calculated as total integrals of the electron distribution function according to the Boltzmann equation in the two-term approximation [20]. Calculations of the EEDF were performed using the program [21]. Based on the resulting EEDF, mean electron energy, the specific power losses of the electric discharge on various elementary processes in the plasma, and the rate constants of elastic and inelastic scattering of electrons on mercury dichloride molecules and neon atoms are determined depending on the value of the reduced electric field (ratio of electric field (E) to the total concentration of mercury dichloride molecules and neon atoms (N)). The range of variation of the parameter E/N = 1…150 Td ($1\cdot10^{-17}…15\cdot10^{-16}$ V·cm^2) included the values of the parameter E/N, which were implemented in the experiment.

All calculations were carried out for a partial pressure of mercury dichloride equal to 1.5 kPa, neon 140 kPa, at which the maximum value of the radiation power was achieved in the experiment (see Fig. 2). In the electron collision integral with mercury dichloride molecules, nitrogen and neon in the Boltzmann kinetic equation the following processes were taken into account: elastic scattering and excitation of the energy states of the neon atom: elastic scattering, excitation of the energy states of Ne atom with a threshold energy of 16.62 eV, 16.67 eV (1s4), 16.84 eV (1s2), 18.72 eV (2p), 20.0 eV (2s + 3d), 20.65 eV (3p), 4.9 eV and ionization of neon atom, dissociative excitation of the electronic state of mercury monochloride ($B^2\Sigma^+_{1/2}$) and ionization of mercury dichloride molecules. The data on the absolute values of the effective cross sections for these processes, as well as their dependences on the electron energy, are taken from the database [21] and the works [22 - 24].

The concentration of electrons (N_e) was calculated by the known formula [25]:

$$N_e = j/e \cdot V_{dr.},$$

where j is the current density in the discharge, e is the electron charge, $V_{dr.}$ is the electron drift velocity.

The electron drift velocity was determined from the expression [25]:

$$V_{dr.} = \mu_e \cdot E,$$

where μ_e is the electron mobility, E is the electric field strength on the plasma.

The electric field strength on plasma E was calculated by the formula:

$$E = U_{pl.}/d,$$

$U_{pl.}$ – voltage on the plasma, d-discharge gap.

The voltage on the plasma was determined according to the second Kirchhoff rule using experimentally measured values of the voltage temporal progress applied to the electrodes of the gas-discharge cell U, as well as the voltage drop across the dielectric capacitance $U_{dl.}$ [26]:

$$U_{pl.} = U_{.} - U_{dl.},$$

voltage U was calculated by the displaced charge Q and the capacitance of the dielectric barrier C_d:

$$C_{d.} = Q/C_{d.}.$$

The charge transferred in the circuit was determined by integrating the current, taking into account the initial conditions:

$$Q(t) = \int_0^t I(t)dt + Q_0,$$

where $Q_0 = Q$ (t =0).

As a result, the electric field strength at the plasma gap for a mixture of mercury dichloride and neon was 2.2×10^6 V/m, and the reduced electric field (E/N) = 100 Td for the total concentration of the components of the mixture (N) = 2.2×10^{25} m^{-3}, at which the maximum radiation power in the spectral band (λ_{max} = 557 nm) of mercury monochloride molecules was observed in the experiment.

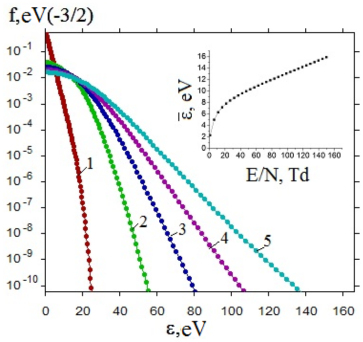

Fig. 4. The electron energy distribution functions in the discharge for the mixture: $HgCl_2$-Ne (0.0107, 0.9893) at a total mixture pressure 141.5 kPa for the values of the parameter E/N: 1 (1), 38.2 (2), 75.5 (3), 113.3 (4), 150 (5) Td; the inset shows the dependence of mean electron energy on the parameter E/N

Mean energy of plasma electrons in a mixture of $HgCl_2$-Ne most depend on the parameter E/N in the range 1…17 Td (Fig. 4, insert). At the same time, it linearly increases from 2.3 to 7.2 eV. In the range of values of the parameter E/N = 17…150 Td, mean electron energy is also increased from 7.2 to 15.9 eV, but at a lower rate. The slower increase in mean electron energy in this range of the E/N parameter is associated with the losses of the energy of fast electrons on the excitation of the energetic states of mercury dichloride molecules and neon atoms. For the values of the reduced electric field at which the experimental studies were performed (100 Td), mean electron energy was 13.1 eV, the electron drift velocity 2.4×10^5 m/s, the electron concentration 9.8×10^{15} m^{-3}.

The distribution of the specific loss of discharge power on the dissociative excitation of $B^2\Sigma^+_{1/2}$ -state of mercury monohloride molecules by electrons in the change of the reduced electric field intensity in the range E/N = (1…150) Td is shown in Fig. 5.

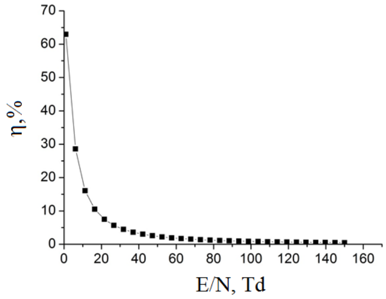

Fig. 5. The dependence of specific losses of discharge power on dissociative excitation by electrons of $B^2\Sigma^+_{1/2}$-state of mercury monochloride molecule in plasma on $HgCl_2$-Ne mixtures (0.0107, 0.9893) with total pressure of 141.5 kPa

For the dissociative excitation process of $B^2\Sigma^+_{1/2}$-state of mercury monohloride molecules by electrons, the specific loss of power of the discharge decreases with the increase of the parameter E/N. It reaches a maximum of 63% at E/N = 1 Td. The rate of reduction of the loss of specific power of discharge for this process and its magnitude are related to the nature of the dependence of the effective cross section of the energy state on the energy of the electrons and its absolute value, the dependence of the EEDF on the value of the reduced electric field and the energy of the dissociative excitation threshold of mercury monohloride molecules. For the dissociative excitation process of $B^2\Sigma^+_{1/2}$-state of mercury monochloride by electrons, the specific loss of discharge power is 0.9% for the reduced electric field E/N = 100 Td (at which experimental studies were conducted).

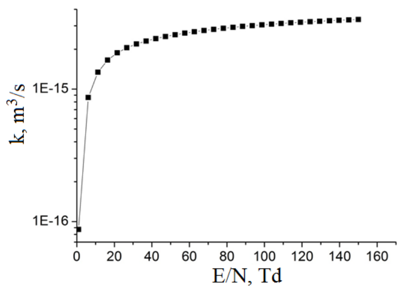

Fig. 6. The dependence of the rate constant of the dissociative excitation of the $B^2\Sigma^+_{1/2}$-state of a mercury monochloride molecule by electrons on the parameter E/N in a discharge in a mixture: $HgCl_2$-Ne (0.0107, 0.9893) at a total pressure of 141.5 kPa

Fig. 6 shows the results of a numerical calculation of the rate constant for the process of dissociative excitation by electrons of the $B^2\Sigma^+_{1/2}$-state of mercury monochloride molecules by electrons. It is observed the increase in it with an increase in the reduced electric field. The rate constant of dissociative excitation by electrons of the $B^2\Sigma^+_{1/2}$-state of mercury monochloride is equal to $3.1\cdot10^{-15}$ m^3/s for a reduced electric field E/N = 100 Td. The emission of spectral bands with a maximum at the wavelength λ = 557 nm of the electron-vibrational transition $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ of HgCl* molecules in a gas-discharge plasma on mixtures of mercury dichloride with neon occurs as a result of the processes leading to formation and destruction of the $B^2\Sigma^+_{1/2}$-state of mercury monochloride, the main of which are [22 - 24]:

$$HgCl_2+e \rightarrow HgCl_2\,(^{3,1}\Sigma^+_u) \rightarrow Cl + e \quad (1)$$

$$\rightarrow HgCl(B^2\Sigma^+_{1/2})+ Cl^- \quad (2)$$

$$HgCl\,(B^2\Sigma^+_{1/2}) \rightarrow HgCl\,(X^2\Sigma^+_{1/2}) + h\nu, \quad (3)$$

λ_{max} = 557 nm

$$HgCl(B^2\Sigma^+_{1/2}) + M \rightarrow HgCl\,(X^2\Sigma^+_{1/2}) + M + \Delta E, \quad (4)$$

where M – concentrations of molecules $HgCl_2$, Ne; ΔE – the difference of energy in the reaction.

Reactions (1) and (2) are the main sources of formation of exciplex molecules HgCl* [22, 23]. Electron-vibrational transitions $B^2\Sigma^+_{1/2} \rightarrow X^2\Sigma^+_{1/2}$ of HgCl* molecules lead to the emission of spectral bands with maximum intensity at wavelength $\lambda_{max.}$ = 557 nm (reaction (3)). In the quenching reaction (4) there is an electron-vibrational transition of mercury monohloride molecules to the ground state without radiation [24].

In addition to processes (1)-(4), which lead to the formation and destruction of the $B^2\Sigma^+_{1/2}$-state of mercury monochloride, next processes are also possible:

$$HgCl_2+e \rightarrow HgCl_2\,(D) \rightarrow HgCl(C\,^2\Pi_{1/2}, D^2\Pi_{3/2}) + Cl + e, \quad (5)$$

$$HgCl\,(C\,^2\Pi_{1/2}, D^2\Pi_{3/2}) + HgCl_2\,(Ne) \rightarrow HgCl(B^2\Sigma^+_{1/2}) + M + \Delta E_{1,2}, \quad (6)$$

in addition to processes (1) and (2), a non-radiation emitting process is possible:

$$HgCl_2+e \rightarrow HgCl_2\,(^{3,1}\Sigma^+_u) \rightarrow HgCl(X^2\Sigma^+_{1/2}) + Cl + e. \quad (7)$$

Process (5) is the process of excitation of $HgCl_2$ molecules by electrons to the state D [27, 28]. This state of mercury dichloride molecules is the sum of all states that are located between the threshold energy (7 eV) and the ionization energy (11.4 eV) [27]. It can be expected that the effective cross section for the excitation of this state by electrons for mercury dichloride molecules is close to the effective excitation cross section D of the state of mercury dibromide molecules, the value of which is 10^{-15} cm^2 [28].

Emission from the D states of the $HgCl_2$ molecules is not observed, due to the fact that this state dissociates with the formation of mercury monochloride molecules in (C, D) states. The emission of HgCl* molecules with C and D states under our experimental conditions is not observed due to the high efficiency of the quenching process (6). The population of these states is transferred to $B^2\Sigma^+_{1/2}$-state of HgCl molecules or to other non-optical channels [6, 28]. The reaction of the collision of mercury dichloride molecules with electrons (7) is the channel for the formation of mercury monochloride molecules in the ground state, the rate constant of which is 8×10^{-15} m^3/s [29].

The sharp increase in intensity from the part of the spectrum with long wavelengths and its slow decrease in the region of short wavelengths (see Fig. 1) is explained by the course of potential curves (excited $B^2\Sigma^+_{1/2}$-state is shifted towards large internuclear distances of relative $X^2\Sigma^+_{1/2}$-state) and processes of relaxation of the population of the upper vibrational levels of the excited electronic state, which occur faster than the electronic-vibrational transition to the ground $X^2\Sigma^+_{1/2}$-state [30, 31].

The oscillatory structure of the current pulse (see Fig. 3,a) is caused by charging and discharging the dielectric capacitance during a voltage pulse with an amplitude sufficient to break the discharge gap [19]. The difference in the shape of current pulses at the front and back fronts is associated with opposite directions of current flow through the gas-discharge gap (1.4×10^{-2} m) and, as a result, unequal charge resorption conditions on the inner surface of the dielectric under conditions equal to the barrier discharge used in our experiment.

The regularity of the difference in the amplitudes of the first and second radiation pulses (see Fig. 3,b) is explained as follows. The first and second pump pulses (current) form mercury monochloride molecules in $B^2\Sigma^+_{1/2}$- and $X^2\Sigma^+_{1/2}$-states due to dissociation of mercury dichloride molecules when colliding with electrons. The second pump pulse (current), in addition, leads to an additional increase in the population of $B^2\Sigma^+_{1/2}$ state of mercury monochloride molecules due to the process:

$$e+HgCl(X^2\Sigma^+_{1/2}) \rightarrow e+HgCl(B^2\Sigma^+_{1/2}), \quad (8)$$

where $HgCl(X\,^2\Sigma^+_{1/2})$ mercury monochloride molecules in the ground state, and which did not have time to recover in a triatomic molecule (mercury dichloride) in the interpulse period (150 ns) in the process [29, 32]:

$$HgCl(X^2\Sigma^+_{1/2}) + Cl + N_2 \rightarrow HgCl_2 + N_2. \quad (9)$$

CONCLUSIONS

The design of the gas-discharge radiation source provided a diffuse and uniform discharge pattern, the cross section of which was $(5\times14)\cdot10^{-6}\cdot m^2$, and its length was 0.2 m.

The gas-discharge plasma of a barrier discharge on a mixture of mercury dichloride and neon vapor produces the emission of the spectral band of an exciplex molecule mercury monochloride in the blue-green spectral region (λ_{max} = 557 nm).

The following functions were established: electron energy distribution functions, transport characteristics, specific discharge power losses on electronic processes, as well as processes rate constants: elastic and inelastic scattering of electrons on the working mixture components depending on the magnitude of the reduced electric field.

For values of the reduced electric field at which experimental studies were carried out (100 Td), mean electron energy was 13.1 eV, which corresponded to the electron temperature of 151960 K, the electron drift velocity was 2.4×10^5 m/s, and the electron concentration was 9.8×10^{15} m^{-3}. The portion of the discharge power going into the process of dissociative excitation by electrons of molecules of mercury monochloride reached a maximum of 0.9%, with the values of the E/N parameter equal to 100 Td for the electronic state $B^2\Sigma^+_{1/2}$. The rate constant of the process leading to the formation of mercury monochloride molecules is 3.1×10^{-15} m^3/s for a reduced electric field (E/N) = 100 Td, at which experimental studies were carried out.

To achieve high values of the radiation power of gas-discharge plasma on a mixture of mercury dichloride vapor and neon in the blue-green spectral region (λ_{max} = 557 nm), it is necessary to reduce the value of the reduced electric field strength to 1 Td, at which the portion of power input is maximal.

The gas-discharge source of radiation of the blue-green spectral range, the working medium of which was the plasma of a barrier discharge on a mixture of mercury dichloride vapor and neon, can be used in scientific research in the field of biotechnology, photonics, medicine, as well as to create indicator gas-discharge panels.

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ОПТИЧЕСКИЕ ХАРАКТЕРИСТИКИ И ПАРАМЕТРЫ ГАЗОРАЗРЯДНОЙ ПЛАЗМЫ НА СМЕСЯХ ПАРОВ ДИХЛОРИДА РТУТИ И НЕОНА

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Представлены результаты исследований оптических характеристик и параметров газоразрядной плазмы барьерного разряда на смеси паров дихлорида ртути и неона. Установлены: ФРЭЭ, транспортные характеристики, удельные потери мощности разряда на электронные процессы, концентрация и температура электронов, константы скоростей процессов упругого и неупругого рассеяния электронов на компонентах рабочей смеси в зависимости от величины приведенной напряженности электрического поля.

ОПТИЧНІ ХАРАКТЕРИСТИКИ І ПАРАМЕТРИ ГАЗОРОЗРЯДНОЇ ПЛАЗМИ НА СУМІШАХ ПАРІВ ДИХЛОРИДУ РТУТІ І НЕОНУ

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Представлено результати досліджень оптичних характеристик і параметрів газорозрядної плазми бар'єрного розряду на суміші парів дихлориду ртуті і неону. Встановлено: ФРЕЕ, транспортні характеристики, питомі втрати потужності розряду на електронні процеси, концентрація і температура електронів, константи швидкостей процесів пружного і непружного розсіювання електронів на компонентах робочої суміші в залежності від величини приведеної напруженості електричного поля.