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Characteristics and Parameters of Plasma of a High-Voltage Nanosecond Discharge in Argon at Atmospheric Pressure with an Ectonic Mechanism of Copper Vapor Introduction into Plasma

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Abstract—The characteristics and parameters of a high-voltage nanosecond discharge between copper electrodes in argon at atmospheric pressure are presented. In the process of microexplosions of inhomogeneities on the working surfaces of the electrodes in a strong electric field; copper vapors are introduced into the gap between the electrodes due to the formation of an ecton. This creates the prerequisites for the synthesis of thin nanostructured copper films which can be deposited on a dielectric substrate installed near the electrode system. The spatial and electrical characteristics of a nanosecond discharge were investigated at a distance $d = 2$ mm between the electrodes. A study of the emission spectra of the discharge and the oscillograms of the emission of the most intense spectral lines and bands was carried out, which made it possible to establish the main excited products that are formed in plasma. Optimization of the time-averaged UV radiation of a point emitter was carried out depending on the voltage supply of the high-voltage modulator and the repetition rate of the discharge pulses. By the method of numerical simulation of the discharge plasma parameters based on argon at atmospheric pressure and copper vapor by solving the Boltzmann kinetic equation for the electron energy distribution function: mobility, tempe-rature and density of electrons in plasma, specific discharge power losses for the main electronic processes and their rate constants depending on the value of the ratio of the electric field strength (E) to the total concentration of argon atoms and a small admixture of copper vapors (N) for plasma under conditions close to those realized in this experiment have been calculated.

Keywords: nanosecond discharge, argon, copper, plasma parameters

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INTRODUCTION

For the development of pulsed radiation sources, the channel stage of a spark discharge in inert gases of a high pressure is of considerable interest since it has a high radiation intensity in the far UV range of wavelengths: 200–250 nm [1, 2]. However, to study theoretical issues in the physics of overvoltage nanosecond discharges in inert gases with the ectonic mechanism [3] of introducing vapors of the electrode material, modeling the output charac-teristics of the corresponding radiation sources and optimizing their operating modes, it is important to know the plasma parameters of the channel stage of the spark discharge. Early studies of the physics and technology of a high-pressure gas discharge, which are summarized in the review [4], showed that the average gas density in the plasma channel of a spark discharge at its later stages was $\approx 5 \times 10^{-6}$ g cm^3, and this corresponded to the concentration of the plasma medium at $\approx 10^{17}$ cm^{-3}. The average temperature of the plasma in the channel reached 40–104 K. Since the temperature in the spark discharge channel remained constant within a wide range of changes in the rate of energy input into plasma, it was concluded that the saturation of the energy and radiation power of the spark discharge channel at its late stage of development is determined by the plasma temperature in a spark.

For a high-voltage nanosecond discharge in gases of atmospheric pressure and short discharge gaps, the formation of ectons from the working surfaces of the electrodes is accompanied by the release of vapors of the electrode material into the interelectrode space, which allows, when using electrodes made of transition metals (Cu, Zn, Fe, Al), to synthesize thin nanostructured films [5, 6] and to produce ultrafine metal powders in sufficient quantities [7, 8] for applications in various nanotechnologies.

The issues of accurately establishing the qualitative and quantitative composition of various samples by emission spectroscopy and the synthesis of thin nanostructured high-purity metal films are closely related to obtaining pure plasma in gas-discharge sources with an open cycle of operation. So, the authors of [9] proposed the use of xenon instead of argon in an arc discharge. Since water-cooled copper electrodes were used in the plasmatron, the lines of the copper atom were recorded in the plasma radiation spectra in the wavelength intervals 210–231; 224.7–327.3 nm and line group 510.5; 515.3; 521.8 nm. The concentration of copper vapors in plasma based on xenon increased significantly compared to that when argon was used as buffer gas, which leads to an increase in the emission intensity of spectral lines of the copper atom. The erosion of the central copper electrode increased in the xenon-based plasma and reached 6×10^{-11} kg/C, while for the argon plasma it did not prevail 1.8×10^{-11} kg/C. An increase in erosion in the xenon plasma at the same discharge currents is due to the predominance of the atomic mass of the xenon over the mass of argon by more than three-fold in the case of the main mechanism of the electron injection in plasma due to the ion bombardment of the cathode.

The results of experimental studies of the emitting and spectral characteristics of a spark channel in argon at atmospheric pressure between aluminum electrodes, both in a magnetic field and without it, are given in [10, 11].

In [12], the results of thorough studies of the effect of a longitudinal magnetic field on the main characteristics of a spark discharge in argon at atmospheric pressure with an admixture of aluminum vapor, which were introduced into plasma during the formation of ectons on the surface of electrodes, are presented. It is shown that the magnetic field shifts the maximum of the spectral radiation density to the UV range of the spectrum, reduces the rate of expansion of the spark channel and losses due to transverse radiation, and increases the specific electric power in the spark, the conductivity and temperature of plasma at the arc stage of the discharge. This creates the prerequisites for producing "hot" plasma and developing new gas-discharge sources of UV and X-ray radiation on its basis.

In [13], a method is given for the synthesis of ultrafine particles with their simultaneous deposition on a substrate. To melt and disperse a metal, a pulsed arc discharge was used, which moved over the surface of non-magnetic steel electrodes in its own magnetic field and which was ignited at atmospheric pressures of air or inert gases. At the cathode, a pulsed electric power density of 10^8–10^{10} W/cm^2 was achieved. This method is used to synthesize ultradispersed particles with a diameter of several nanometers on the surfaces of polymer membranes of hydrogen elements. However, the optimization of the output characteristics of such a discharge and their relationship with the number and charac-teristics of the synthesized particles was not carried out, and the physical parameters of the plasma remained undetermined.

The results of modeling the plasma parameters of a nanosecond discharge in argon at atmospheric pressure are presented in [14]. On the base of a two-dimensional axisymmetric diffuse-drift model, the calculations of the influence of the initial conditions on the formation and development of a cathode-directed ionization wave between two plane electrodes were carried out. The experiments were performed with the preliminary UV overionization of a 1 cm discharge gap between stainless steel electrodes. Calculations have shown that the formation of a discharge begins with a cathode-directed wave that moves at a speed of $\approx 2.5 \times 10^7$ cm/s, and the concentration of electrons in the plasma after reaching the time to 40 ns, excluding the cathode layer, was maintained in the range 10^{13}–10^{14} cm^{-3} at an electric intensity fields $\approx 10^5$ V/cm. However, in that paper, there is no information on a possibility of synthesizing nano-particles from the electrode material, and the discharge ignition mode was not optimal for the intensive formation of ectons and the introduction of iron vapor into plasma.

To the best of the authors knowledge, the results of studying the conditions for the synthesis of thin films based on copper nanostructures from the destruction products of copper electrodes in a high-voltage nanosecond discharge are practically absent in the respective literature. This is why, a study of the synthesis conditions and the main characteristics of such films in inert gases is urgent, which is impossible without the use of an expensive laser or high-vacuum equipment.

The current article presents the results of a study of the spatial, electrical, and optical characteristics of a high-voltage nanosecond discharge of atmospheric pressure in argon with an admixture of copper vapors, which was introduced into plasma due to the formation of ectons on the working surfaces of copper electrodes in a strong electric field, as well as the results of calculating the parameters of a nanosecond discharge plasma in mixtures of argon with copper vapor obtained by numerical simulation using the Boltzmann equation for the electron energy distribution function (EEDF); the simulation was carried out under conditions as close as possible to those realized in the experiment.

TECHNIQUES AND EXPERIMENTAL CONDITIONS

The study of the characteristics of overstressed nanosecond discharge was carried out on a discharge module, the diagram of which is shown in Fig. 1.

An overstressed nanosecond discharge between copper electrodes was ignited in a sealed plexiglas

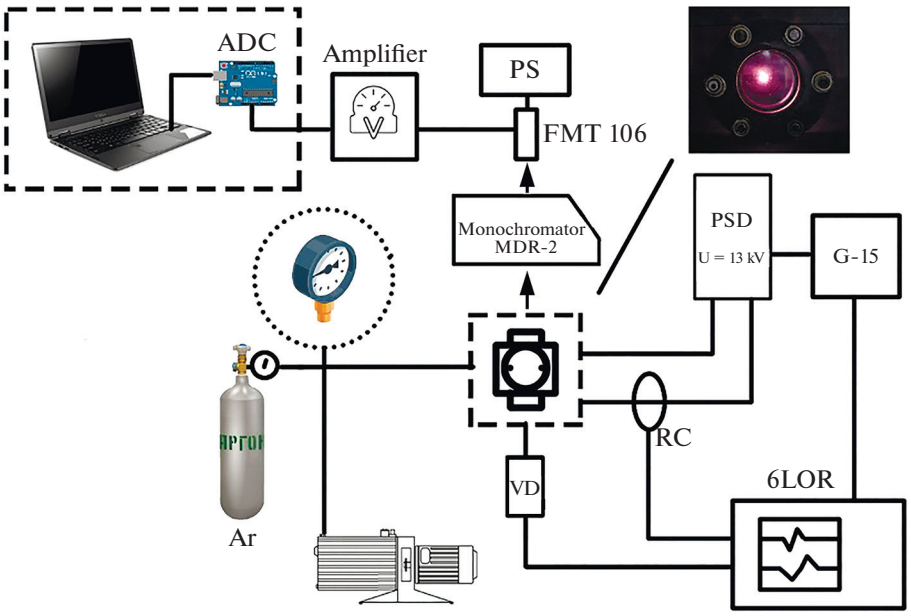

Fig. 1. Diagram of an experimental setup for studying overstressed nanosecond discharge in gases: PSD – discharge power supply; FMT 106 – photoelectric multiplier tube; PS – power supply unit; ADC – analog-to-digital signal converter; RC – Rogovsky coil; VD – voltage divider to determine the voltage drop in the circuit; G5-15 – pulse generator, and 6LOR – broadband oscilloscope.

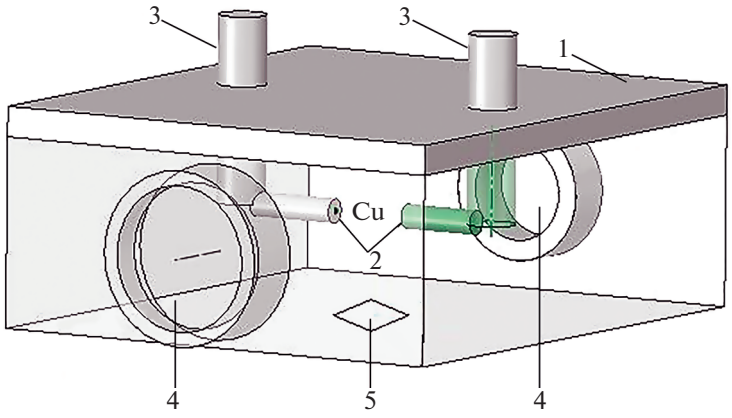

Fig. 2. The structure of the module with a system of electrodes for overstressed nanosecond discharge in gases: *1* – the body of the discharge chamber; *2* – electrodes; *3* – high-voltage inputs; *4* – quartz windows for monitoring the discharge and recording emission spectra, and *5* – a substrate for deposition of thin films.

chamber. The distance between the copper electrodes was $d = 2$ mm.

The experimental setup included a discharge module (Fig. 2) which was powered by a repetitively pulsed high-voltage source, a system for recording optical radiation based on an MDR-2 mono-chromator, and a system for recording pulsed electrical and optical characteristics (current, voltage, and oscillograms on the most intense spectral lines and bands) in time based on a 6LOR multichannel wideband oscilloscope.

The discharge module was connected to a vacuum system using metal tubes, which provided a range of argon operating pressures from 1–200 kPa, but the overwhelming majority of experimental results were obtained at p (Ar) = 101 kPa.

To ignite the discharge, bipolar high-voltage pulses with duration of 50–100 ns and an amplitude of ± 20–40 kV were applied to the electrodes of the discharge cell. The voltage pulse repetition rate was chosen equal to 80–100 Hz since at high frequencies the discharge module was significantly heated. Oscillograms of volt-

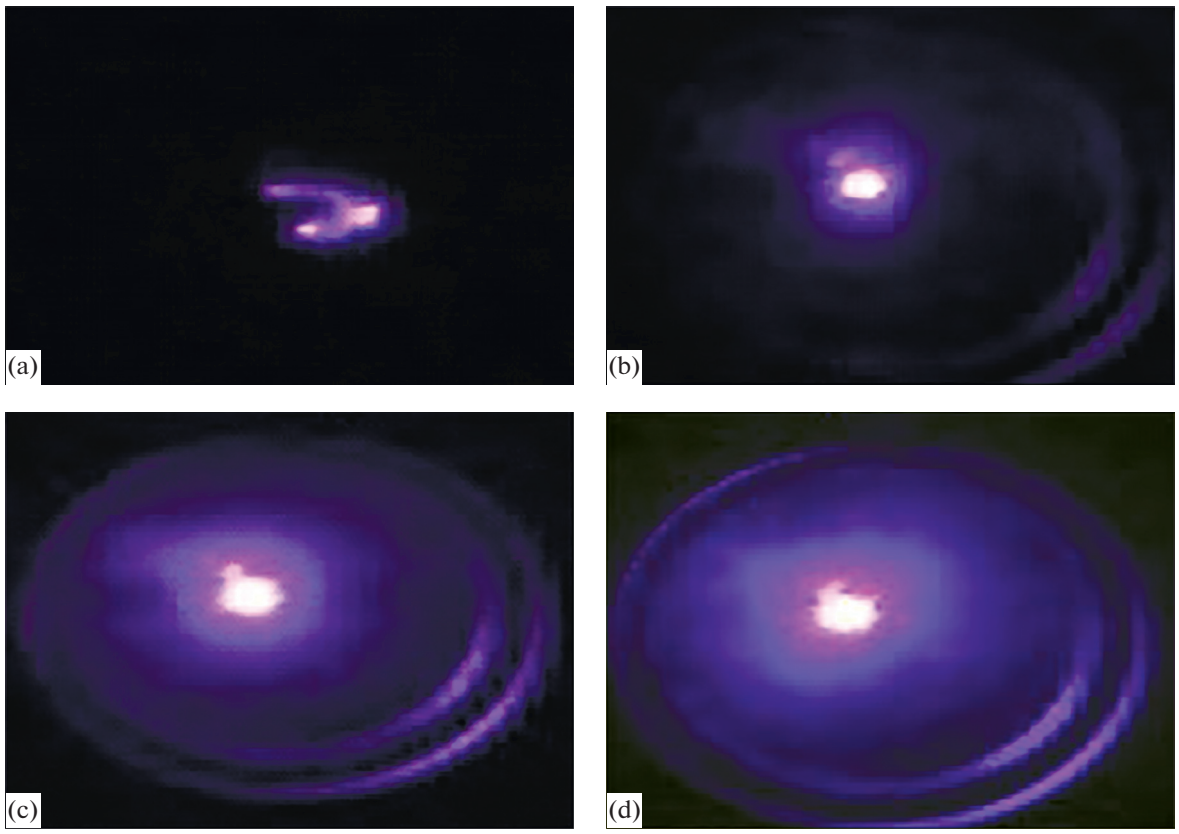

Fig. 3. Images of a high-voltage nanosecond discharge between copper electrodes in argon at different pressures: (a) 6.7 kPa; (b) 50.57 kPa; (c) 1017 kPa, and (d) 151.57 kPa, at $d = 2$ mm.

age pulses across the discharge gap and oscillograms of current pulses were recorded using a broadband capacitive voltage divider, a Rogowski coil, and a 6LOR-04 broadband oscilloscope with a time separation of 2–3 ns.

A fairly uniform discharge with a duration of about 100–200 ns with an amplitude of current pulses of 50–200 A and a plasma volume of 10–500 mm^3 was ignited between the tips of the copper electrodes. At an interelectrode distance of 2 mm, the discharge gap was overstressed, which created favorable conditions for the formation of a beam of runaway electrons of a high energy and accompanying X-ray radiation [15].

Plasma radiation, recorded in the spectral range λ = 196–663 nm, entered the entrance slit of a spectrometer with a 1200-line diffraction lines/mm. An FEU-106 photomultiplier tube connected to a *dc* amplifier was used at the output of the spectrometer to detect radiation. The signal from the amplifier was fed into an analog-to-digital converter and then to a computer for processing.

The measurement of the radiation power of the discharge plasma was carried out using an ultraviolet meter of the absolute radiation power TKA-PKM, which made it possible to carry out measurements in the spectral range of 200–400 nm. The structure of the discharge module is shown in Fig. 2.

The discharge chamber was evacuated with a foreline pump to a residual pressure of 10 Pa, and then argon was fed into the chamber to a pressure of 101 kPa. The diameter of the cylindrical copper electrodes was 5 mm, and the radius of curvature of their working end surface was the same and equal to –3 mm.

The discharge volume depended on the voltage pulse repetition rate. The point discharge mode was achieved only at voltage pulse repetition rates in a range of $f = 40$–150 Hz. With a short-term increase in frequency to 1000 Hz, the volume of the plasma of the gas-discharge emitter grew up to 100–130 mm^3.

SPATIAL AND ELECTRICAL CHARACTERISTICS

Figure 3 shows the time-averaged photographs of a high-voltage nanosecond discharge at a pressure *p* (Ar) 101 kPa, which were obtained with a camera time exposure of $\approx$ 1.

At an argon pressure of 6.7 kPa, a high-voltage nanosecond discharge looked like a bright central part with a diameter of about 2 mm, which was equal to the interelectrode distance from which two jets of green

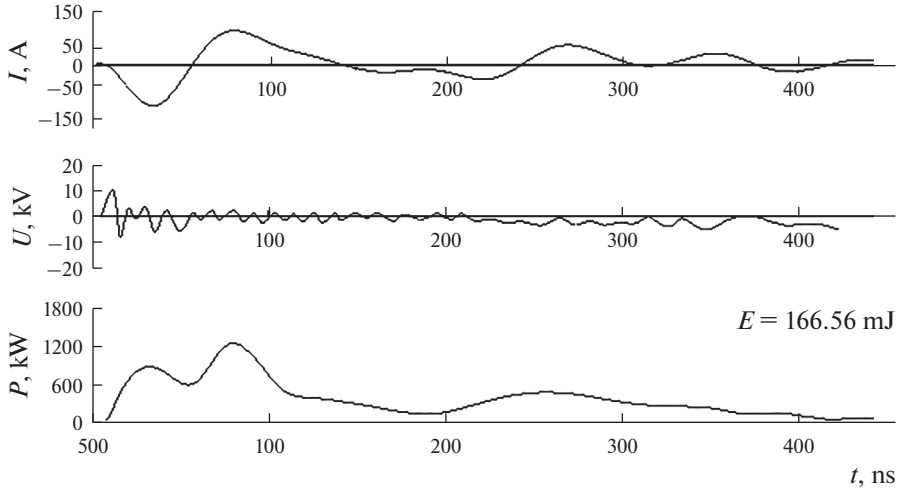

Fig. 4. Temporal dependences of the pulsed electric power of the discharge, the voltage between the copper electrodes, and the current of the nanosecond discharge at the interelectrode distance $d = 2$ mm and p (Ar) = 101 kPa.

plasma flowed out. With an increase in the argon pressure to 101–151.5 kPa, the diameter of the bright central part increased by a factor of 2–3 and, accordingly, the diameter of the halo that covered it increased.

Figure 4 shows oscillograms of voltage, current, and pulse power for a high-voltage nanosecond discharge between copper electrodes in argon at atmospheric pressure.

Oscillograms of voltage and current were in the form of oscillations decaying in time, which was caused by the mismatch of the output resistance of the high-voltage modulator with the load resistance. The total duration of the voltage oscillations across the gap and the discharge current reached 450 ns, with the duration of individual voltage oscillations 7–10 ns, and the current oscillations with duration of ≈ 70 ns. Short-term oscillations showed up better in voltage waveforms. On the oscillograms of current, they were partially integrated in time due to a large time constant of the Rogowski coil used in these studies.

For a discharge in argon at atmospheric pressure at $d = 2$ mm, the amplitude of the largest voltage drop on the electrodes was achieved at the initial stage of the discharge and was ≈ ± 7–8 kV, current ± 100 A, and pulsed power −1.2 MW, which provided an energy contribution to one discharge pulse into the plasma about ≈ 167 mJ (Fig. 4).

OPTICAL CHARACTERISTICS

Figure 5 shows the emission spectra of a high-voltage nanosecond discharge between copper electrodes, which was ignited at p (Ar) 101 kPa and the distance between the electrodes of 2 mm, and the results of its interpretation are summarized in Table 2. When identifying spectral lines in the spectra, the respective reference books were used [16–18].

A feature of these emission spectra was the presence of a continuum against the background of which all spectral lines and bands were observed. The nature of this continuum under the conditions of the present experiment is associated with both thermal and recombination plasma radiation. The shape of the continuum with a broad maximum at 400–450 nm correlated well with the results known from publications elsewhere. For example, in [10, 11] for a spark discharge in argon at atmospheric pressure between aluminum electrodes at $d = 10$ mm, an intense continuum was recorded in the emission spectrum in the wavelength range of 350–460 nm with a maximum at $\lambda = 420$ nm. The continuous spectrum of plasma radiation began to be recorded after a time interval of Δt 40 min from the beginning of the discharge when the intensity of the argon ion lines began to increase sharply. The maximum intensity of the continuous spectrum Ar 11 spectral lines was achieved after a time interval Δt 300–400 ns from the onset of a sharp increase in current [11].

At the stage of slow expansion of the spark channel, starting from the time instant −500 ns, the intensity of the continuum decreased, as did the intensity of the ionic spectral lines of argon, and the intensity of the atomic lines: 394.9; 308.2 nm Ar 1 and lines of the electrode material (281.6; 289.1 nm Ar 11) grew [11].

In the emission spectra of the discharge plasma (Fig. 5) on a mixture of argon with copper vapor in the UV wavelength range of 214–330 nm (lines 1–23 in Table 1), emission was observed at the transitions of the copper atom and a singly charged ion. The most intense ionic spectral line was the line with λ 227.62 nm Cu 11, and the atomic intense were the resonance spectral lines of the copper atom with λ 324.75 and 327.39 nm Cu 1, in which the lower energy level was the ground level. The highest energy of the upper level

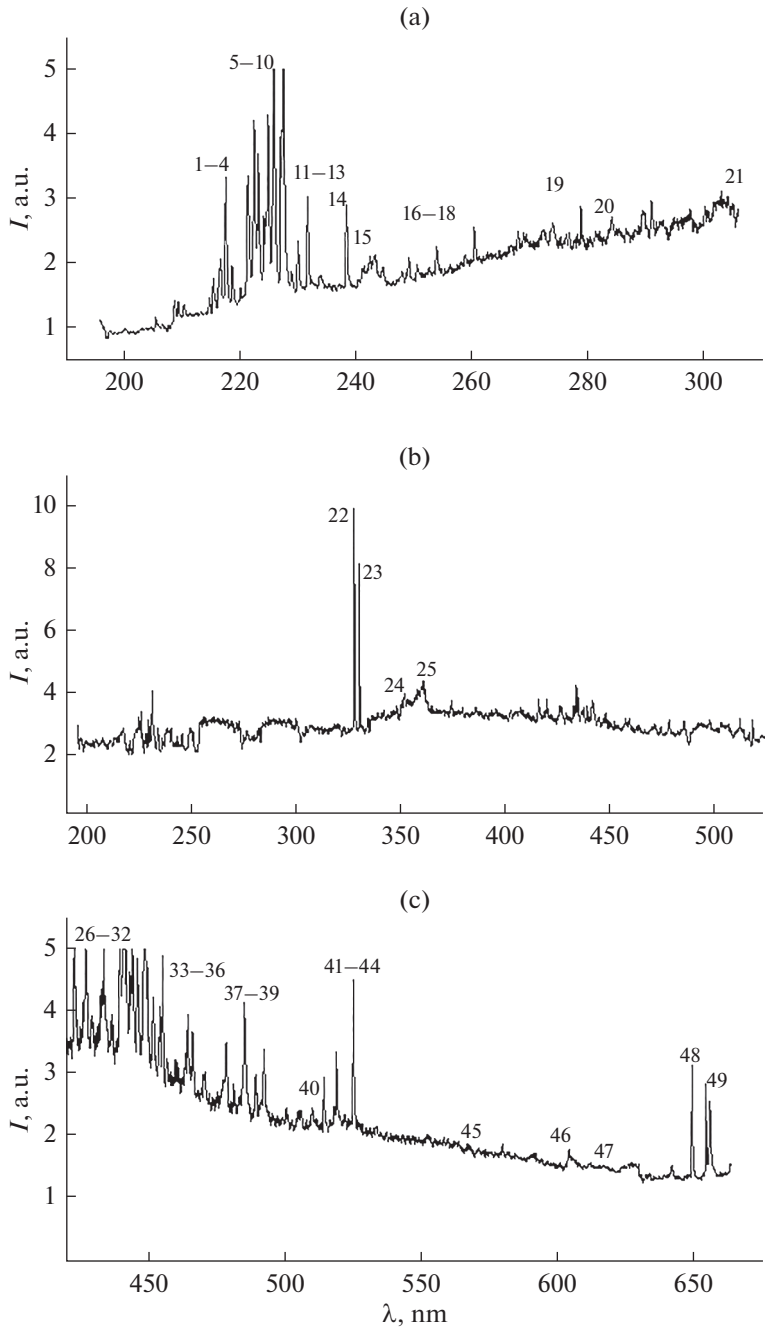

Fig. 5. Emission spectra of a high-voltage nanosecond discharge in argon between copper electrodes at *d* 2 mm and *p* (Ar) 101 kPa.

for the Cu 11 lines was E_{up} = 18.77 eV, and for atomic lines the maximum energy of the upper level was E_{up} = 7.18 eV. In the UV spectrum, bands of the second positive system of the nitrogen molecule were also observed since in these experiments the discharge chamber was pumped out only up to a residual air pressure of ≈ 10 Pa. In the wavelength range of 360–659 nm, radiation was observed mainly at the Ar 1 and Ar 11 transitions, as well as an intense spectral line of the hydrogen atom (λ = 656.29 Hα), which was due to the residual air pressure in the chamber after it was evacuated by a foreline pump.

When the distance between the electrodes is 2 mm and a bipolar generator of high-voltage nanosecond pulses is used, one or several (with a strong overstress of the discharge gap) cathode spots are formed, moving one towards the other.

In [19], similar results were reported for studying the optical characteristics of the cathode part of a unipolar high-voltage nanosecond discharge with dura-

tion of 20 ns (pulse current 100–150 A, $f = 25$ Hz) in air at atmospheric pressure. However, the use of a unipolar discharge and air instead of argon leads to a different distribution of the emission intensity of the spectral lines of copper, as indicated by a comparison of the data from Tables 1 and 2.

For a high-voltage nanosecond discharge in argon at atmospheric pressure, some optical and gas-dynamic characteristics of the cathode spot plasma were given in [10, 11]. However, in those experiments carried out at an interelectrode distance 10 mm between aluminum electrodes, the radiation of the cathode spot plasma with a diameter of ≈ 0.2–0.3 mm and, attached to it, the diffuse discharge plasma, which occupied most of the discharge gap, was clearly recorded. The formation of cathode spots in an overstressed nanosecond discharge is explained by the explosive emission model [3] which is based on the introduction of significant energy at the microtips of the cathode surface with their subsequent heating and explosion. The cathode spot was formed simultaneously with the onset of a sharp increase in current and a drop in voltage across the gap, and at the initial stages of formation it was hemispherical and the expansion rate was low – $\approx 2.5 \times 10^6$ cm/s. The kinetic energy of 50–100 eV corresponded to this speed of movement of the cathode spot at which an ecton with the number of electrons 10^{11}–10^{12} and a bunch of metal vapors were formed. At the initial stage of the discharge, the radius of the cathode spot did not prevail ≈ 10^{-6} m, and with an increase of time t from 5 to 50 ns, it also went up – from 0.5 to 4.3×10^{-4} cm [10]. The electron temperature of the near-cathode argon plasma at atmospheric pressure, which was estimated from the relative emission intensity of the spectral lines of argon, at the initial moments of its formation (in the time interval ≈ 30–40 ns) reached 5 eV, and at subsequent moments of time (τ = 50–500 ns) decreased from 4.2 to 3.4 eV [11]. The main part of the discharge energy is introduced into the cathode spot in the plasma phase, therefore, in the plasma bunch, it scatters; the energy is first introduced into the electronic component with the subsequent transfer of this energy from electrons to ions. Therefore, the mechanism of formation of excited copper ions in plasma can be determined by the processes of their excitation by electrons from the ground state of the corresponding ion. The effective cross sections for the excitation of transition metal ions by electron impact are large and, for example, for zinc ions they reach 10^{-16} cm^2 [20]. Data on the effective cross sections for the excitation of a copper ion in the ground energy state by electron impact are absent in the studied respective literature, which may be due to the difficulty of obtaining stable beams of copper ions suitable for measuring the corresponding effective cross sections.

The reason for such processes is a high concentration of electrons in the plasma of overstressed discharges of nanosecond duration with an ectonic mechanism of electrode sputtering, which reaches 10^{17} cm^{-3} [21]. Estimates of the concentration of electrons in a spark discharge in argon at atmospheric pressure, carried out in [11] using the experimentally determined half-width of the spectral line Ar 11: $\Delta\lambda$ = 0.42 nm, showed that it is ≈ 2.4×10^{18} cm^{-3}.

Figures 6 and 7 show oscillograms of radiation at the transitions of the most intense spectral (resonance) lines of the copper atom and the bands of the second positive system of the nitrogen molecule at argon pressures of 6.7 and 101 kPa.

At an argon pressure of 6.7 kPa (Fig. 6), the luminescence maximum at the transition with λ = 327.39 nm of Cu I was observed at $t \approx 60$ ns, and for the line with λ = 324.75 nm Cu I – at t ≈ 80 ns. The emission maxima for the bands of the nitrogen molecule were observed at a time of $t \approx 70$ ns. For nanosecond discharges in mixtures of argon with small admixtures of nitrogen or air, an efficient process of energy transfer from metastable argon atoms to a nitrogen molecule is characteristic [22]. Thus, the rate constant for the transfer of energy from metastable Ar ($^3P_2$) atoms to a nitrogen molecule with its excitation N_2 (C) is ≈ 3.5×10^{-11} cm^3/s [23]. This can lead to a time delay with the luminescence of the bands of the second positive system of the nitrogen molecule relative to the pulsed energy contribution or the current pulse.

The main (the first in time from the beginning of the discharge ignition) luminescence maxima at the resonance transitions of the copper atom correlated with the first two maxima of the pulsed power of the electric discharge (t = 50 and 80 ns). At medium argon pressures, the most probable mechanisms for the population of the upper energy levels for spectral lines with λ = 324.75 nm and 327.39 nm of Cu I can be the processes of direct and stepwise excitation by electron impact. For the oscillogram of the line of the Cu I atom with λ = 324.75 nm, the second maximum of the luminescence intensity was practically absent, and, for the line with λ = 327.39 nm, it had a low intensity. This may be due to the strong self-absorption of radiation at the late stages of the spatially uniform stage of the discharge, due to the fact that for these copper lines the lower energy level is the main one (see Table 2) and the accumulation of copper vapors in the discharge gap. More intense were the second maxima for emission in the bands of the nitrogen molecule (t = 230, 270 ns), which were observed in the afterglow from the main maximum of the pulsed power (pulse t = 150 ns, Fig. 6).

Emission from the transitions of the copper atom was in the afterglow of the first maximum of the pulsed power, weak second luminescence maxima were recorded only for spectral lines with λ = 306.34, 521.82 nm Cu I at t = 150 ns. It is likely that, at atmospheric pressure of argon, the duration of the diffuse stage of the high-voltage nanosecond discharge was over 100 ns, and the second maxima of the lumines-

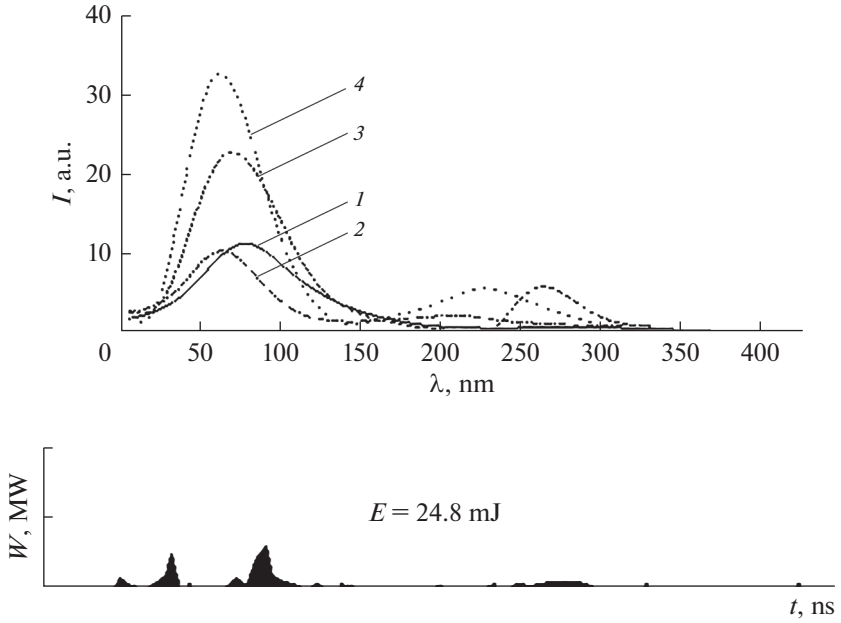

Fig. 6. Oscillograms of luminescence at the transitions of the copper atom and nitrogen molecule in a high-voltage nanosecond discharge at an Argon pressure of 6.7 kPa: *1* – 324.75 nm Cu I; *2* – 327.39 nm Cu I; *3* – 357.69 nm N_2; *4* – 441.67 nm N_2 and a pulse waveform power.

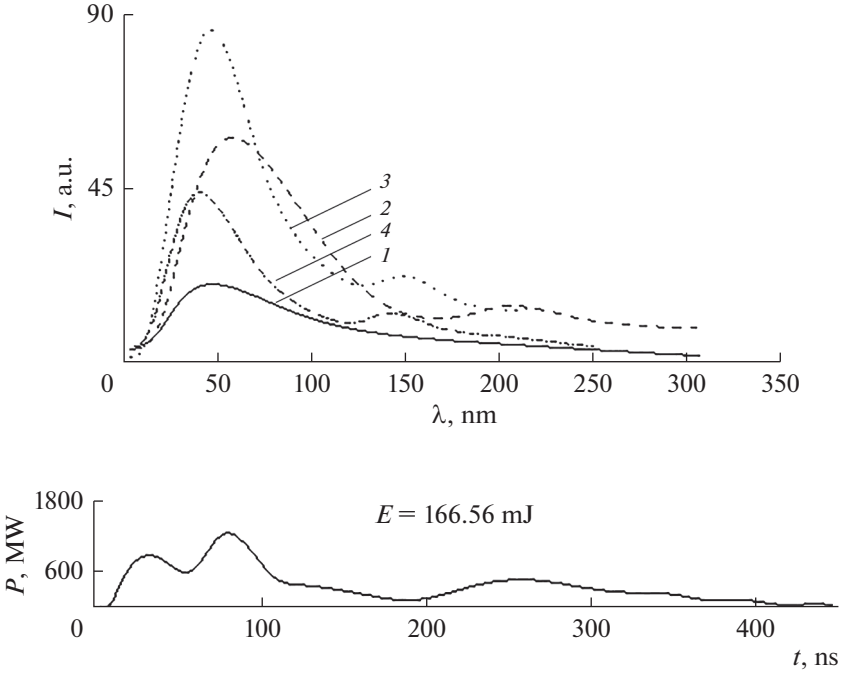

Fig. 7. Oscillograms of luminescence at the transitions of Cu and Ar atoms: *1* – 249.1 nm Cu I; *2* – 306.34 nm Cu I; *3* – 470.23 Ar 1; *4* – 521.82 nm Cu I in a discharge at an Argon pressure of 101 kPa and an oscillogram of the pulsed power.

cence of the lines of the copper atom appear already at the contracted stage of the discharge. Such a picture of the development of those discharges was established in [24] during high-speed photography of the discharge with a nanosecond temporal resolution. The absence of a repeated maximum in the oscillogram of the 249.1 nm Cu I spectral line, of which the lower is the main energy level of the copper atom, may be due to the pro-

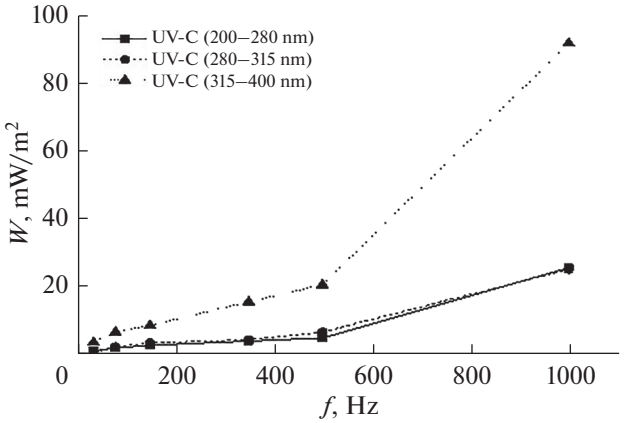

Fig. 8. Dependence of the intensity of UV radiation: UV-C, UV-B, and UV-A ranges of high-voltage nanosecond discharge, on the repetition rate of voltage pulses at a charging voltage of $U = 13$ kV ($f = 80$ Hz, p (Ar) = 101 kPa, and $d = 2$ mm).

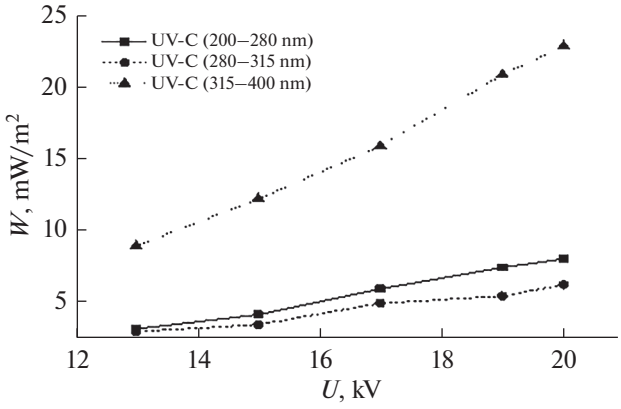

Fig. 9. Dependence of the intensity of UV radiation: UV-C, UV-B, and UV-A ranges of high-voltage nanosecond discharge, on the value of the charging voltage on the working capacitor of the high-voltage modulator (f = 80 Hz, p (Ar) = 101 kPa, and $d = 2$ mm).

cess of self-absorption of radiation at the channel stage of the discharge when the concent-ration of copper vapors in plasma increases.

Radiation at the transition of the argon atom with $\lambda = 470.23$ Ar 1 nm, also observed in the afterglow of the first maximum of the pulsed power of the discharge, can be determined by the recombination mechanism of population of its upper energy level.

The radiation of the plasma products of an overstressed discharge in argon at atmospheric pressure correlated with the data for a discharge in argon between electrodes from chalcopyrite [25], and in a mixture of nitrogen with oxygen between aluminum electrodes [26].

Figures 8 and 9 present the results of optimization of the average intensity of UV radiation of the discharge depending on the pulse repetition rate (Fig. 8) and on the value of the charging voltage across the working capacitor of the high-voltage modulator (Fig. 9).

The maximum values of the average power of UV radiation in argon at an argon pressure of 101 kPa for various UV ranges were: UV-C (200–280 nm) – 67 mW/m^2, UV-B (280–315 nm) – 65 mW/m^2, and UV-A (315–400 nm) – 204 mW/m^2 (at $U = 20$ kV, $f =$ 1 kHz). With a decrease in the argon pressure to 6.7 kPa, the maximum value of the average power of UV radiation in the UV-C spectral range (200–280 nm) became 4.4 W/m^2, in the UV-B (280–315 nm) range – 4.4 mW/m^2, and in the UV-a (315–400 nm) range – 11.7 mW/m^2 (at $U = 20$ kV, and $f = 1$ kHz).

With an increase in the pulse repetition rate from 40 to 1000 Hz, the highest increase in the discharge radiation intensity in the UV-A (315–400 nm) range, in particular, in the frequency range $\Delta f = 350$–1000 Hz, it increased from 8 to 95 mW/m^2. In the UV-B and UV-C ranges, an increase in the radiation power density was lower and was observed in a range of 3.0–25.0 mW/m^2. An increase in the intensity of UV radiation of the discharge, depending on the value of the charging voltage across the working capacitor of the high-voltage modulator at a fixed repetition rate was less effective and was in a range of 8.0–24 mW/m^2. In this case, the relative ratios between the intensities of radiation in the ranges of UV-C, UV-B, UV-A were the same as for the corres-ponding dependences on frequency.

PARAMETERS OF THE DISCHARGE PLASMA IN A MIXTURE OF ARGON WITH COPPER VAPOR AT ATMOSPHERIC PRESSURES (COMPONENT RATIO 101 kPa : 30 PA AND 202 kPa : 30 Pa)

Plasma parameters were determined numerically and calculated as total integrals of the EEDF. The EEDF values were found by solving the kinetic Boltzmann equation in a two-term approximation. The EEDF calculations were carried out using the program [27], where the effective cross section database also includes the effective cross sections for the interaction of electrons with copper atoms.

The durations of voltage oscillations in the gap with a half-period of ~ 10 ns in a diffuse plasma medium are in an electric field which changes slower than the settling time of the electron distribution function. It is known that the settling time of the quasi-stationary distribution (τ) of electrons is approximately equal to the relaxation time of the mean electron energy [28]. Estimation of the value of τ for the presented here experiment gives a value of ~ $1–10^{-13}$ s, which is significantly lower than the duration of 10 ns. Therefore, the authors consider that for the experiment discussed here, it is possible to use the standard program for

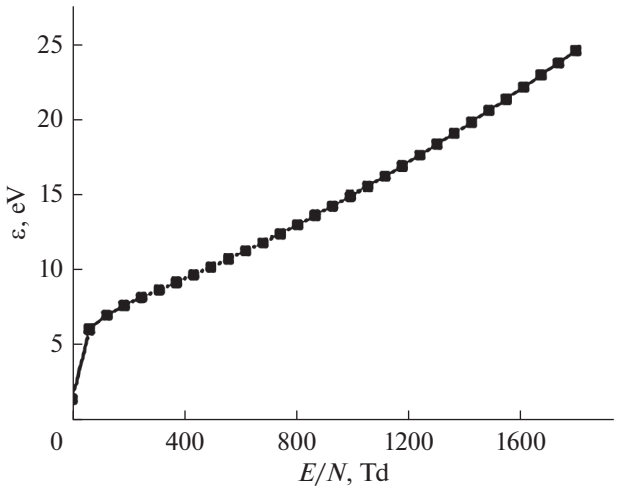

Fig. 10. Dependence of the mean energy of electrons in plasma of a vapor-gas mixture Cu:Ar = 30–101000 Pa at a total pressure of p = 101.030 kPa on the reduced electric field strength.

solving the stationary kinetic Boltzmann equation for the EEDF Bolsig+.

On the base of the obtained EEDF, a number of plasma parameters were determined depending on the magnitude of the reduced electric field – the parameter E/N (the ratio of the electric field strength (E) to the total concentration of argon atoms and a small admixture of copper vapors (N)). The range of variation of the parameter E/N = 1–1800 Td (1 – 10^{-17}–1.8 – 10^{-15} V/cm^2) included the values of the parameter E/N which were realized in the experiment. For a discharge in a vapor-gas mixture Cu : Ar = 30 : 101000 Pa at a total pressure p = 101.030 kPa, the values of the E/N parameter were 1676 Td and 807 Td at 100 ns and 480 ns from the beginning of the discharge, respectively, and for discharge in a vapor-gas mixture Cu : Ar = 30 : 202000 Pa at a total pressure p = 202.030 kPa, the values of the E/N parameter were 1242 Td and 621 Td, respectively. In the integral of collisions of electrons with atoms and molecules, the following processes are taken into account: elastic scattering of electrons by Cu and Ar atoms, excitation of the energy levels of the copper atom (threshold energies 1.500 eV, 3.800 eV, 5.100 eV), ionization of the copper atom (threshold energy 7.724 eV), excitation energy level of the argon atom (threshold energy 11.50 eV), ionization of the argon atom (threshold energy, 15.80 eV), electron-electron and electron – ion collisions.

Figure 10 shows the dependence of the mean energy of electrons in the plasma of the vapor-gas mixture Cu : Ar = 30–101000 Pa at a total pressure p = 101.030 kPa on the reduced electric field strength.

The mean energy of the discharge electrons in the vapor-gas mixture Ar : Cu = 101 kPa – 30 Pa increased from 1.49 to 24.72 eV, and for the mixture Ar : Cu = 202 kPa–30 Pa it also increased from 1.64 to 24.73 eV at an increase in the reduced electric field strength from 1 to 1800 Td (Fig. 10). At the same time, an increase in the rate of its change was observed in the range of the parameter E/N = 1–63 Td.

Table 3 shows the results of calculating the transport characteristics of electrons: mean energy (ε), temperature (T) and drift velocity ($V_{dr.}$) of electrons for a discharge in two mixtures of copper vapors with argon.

The electron drift velocity is 9.0 × 10^5 m/s for a plasma field strength of 4 × 10^7 V/m and 1.0 × 10^6 m/s, as well as for a plasma field strength of 2 × 10^7 V/m, which was reached after the instant τ = 100 ns from the beginning of the breakdown of the interelectrode gap (the value of the voltage pulse amplitude decreases to 20000 V, Fig. 10 for a discharge in a mixture: Cu – Ar = 30 Pa – 101 kPa. And for a mixture of Cu – Ar = 30 Pa – 202 kPa $V_{dr.}$ = 7.0 × 10^5 m/s and 1.1 × 10^6 m/s, respectively. The value of the electron concentration is 5.3 × 10^{19} m^{-3}–3.8 × 10^{19} m^{-3} at a current density (7.65–6.12) 10^6 A/m^2 on the surface of the radiation source electrode (0.196 × 10^{-6} m^2) for the reduced electric field strength E/N = 1676 Td, which existed on the discharge gap in the first 100 ns and for the reduced electric field strength E/N = 807 Td, which existed on the discharge gap after τ = 100 ns for a discharge in a Cu : Ar = 30 Pa – 101 kPa mixture. For a discharge in a Cu : Ar = 30 Pa – 202 kPa mixture, the value of the electron concentration was in a range 8.2 × 10^{19} m^{-3}–3.5 × 10^{19} m^{-3} at a current density (9.2–6.1) × 10^6 A/m^2 on the surface of the radiation source electrode (0.196 × 10^{-6} m^2) for the reduced electric field strength E/N = 1242 Td, which existed in the discharge gap during the first 100 ns, and for the reduced electric field strength E/N = 621 Td, which existed in the discharge gap after a time of 100 ns.

Specific power losses of the discharge in a mixture of copper vapors with argon (Ar : Cu = 101 kPa–30 Pa) for inelastic processes of collisions of electrons with the components of the mixture for a reduced electric field strength of 1667 Td are maximum for ionization of Argon atoms (Fig. 11), and for its excitation an energy level with a threshold energy of 11.5 eV, they reached 34% and 16%, respectively. For the discharge in the second mixture (Ar : Cu = 202 kPa – 30 Pa), the specific losses of the discharge power were 37 and 21% at the reduced electric field strength of 1242 Td, respectively.

For the reduced field strength E/N = 807 Td, which arose after a time of 100 ns from the beginning of the breakdown of the interelectrode gap, in the first mixture the specific losses of the discharge power were 39 and 28%, respectively, and for the discharge on the second mixture after a time of 100 ns, they were 39 and 33% for E/N = 621 Td.

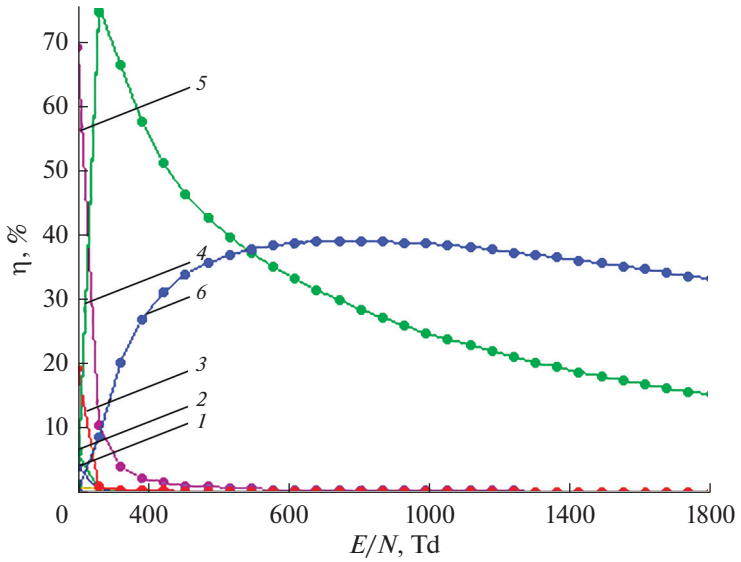

Fig. 11. Dependency of the specific discharge power losses for the processes of collisions of electrons with Cu and Ar atoms on the E/N parameter in plasma based on a Cu : Ar = 30–101 000 mixture at a total pressure of p = 101.030 kPa: *1* – excitation of the energy level of the Cu atom (threshold energy 1.5 eV); *2* – excitation of the state of the Cu atom (threshold energy 1.5 eV); *3* – elastic scattering by Ar atoms; *4* – excitation of the energy level of the Ar atom (threshold energy 11.5 eV); *5* – excitation of the resonance level of the Cu atom $^2P_{3/2,\,1/2}$ (threshold energy 3.8 eV); *6* – ionization of the Ar atom (threshold energy 15.8 eV).

For the Cu atom, they did not exceed 0.07% (for the excitation of the resonance state $^2P_{3/2,\,1/2}$) for the first mixture at E/N = 1667 Td, and for the discharge in the second mixture, 0.06% at E/N = 1242 Td. The maximum specific losses of the discharge power in the first mixture were observed for the excitation of the energy level of the Ar atom with a threshold energy of 11.5 eV and reached – 76% for the reduced electric field strength of 63 Td and 70% for the excitation of the resonance level $^2P_{3/2,\,1/2}$ of the Cu atom for the reduced electric strength field 1 Td, and for the discharge in the second mixture 80% for the excitation of the energy level of the Ar atom with a threshold energy of 11.5 eV for E/N = 63 Td and 61% for the excitation of the resonance state $^2P_{3/2,\,1/2}$ of the Cu atom for E/N = 1 Td. With an increase in the E/N parameter to 1800 Td, the specific discharge power losses in mixtures reached 33% for the process of ionization of Ar atoms by electrons. The rates of an increase and adecrease in the discharge power losses on the processes of excitation of electronic states and ionization and its values (Fig. 11) are associated with the nature of the dependence of the effective cross sections of inelastic processes of collisions of electrons with the components of the mixture on the energy of electrons, their absolute values, as well as with the dependence of the EEDF on the reduced electric field strength and threshold energy of the process. The power losses of the discharge for the excitation and ionization of the Cu atom in the range of values of the reduced electric field strength, at which our plasma source worked (1676–807 Td) and (1242–621 Td), are small because of a low content of Cu vapors in the gas-vapor mixture.

Table 4 shows the highest values of the rate constants of collisions of electrons with atoms of Cu and Ar for three values of the reduced electric field strength, is a measure of the efficiency of the process for a discharge on a mixture Cu : Ar = 30Pa – 101 kPa at p = 101030 Pa and for a discharge in a mixture Cu : Ar = 30 Pa – 202 kPa at p = 202030 Pa. They varied in the range 0.2140×10^{-16}–0.1178×10^{-11} m^3/s for the selected values of the reduced electric field strength. The rate constant of elastic scattering of electrons by the Cu atom is maximum, and with a decrease in the reduced electric field strength, it decreased by almost 1.4 times both for the discharge in the first and second gas-vapor mixtures. The rate constant of excitation of the resonance level of the Cu atom for the discharge in the first mixture is higher and amounted to 0.9495×10^{-12} and 0.7831×10^{-12} m^3/s at the reduced electric field strength at which the plasma radiation source for film deposition operates (E/N 1676 Td and 807 Td).

CONCLUSIONS

Thus, it was found that at an argon pressure of 101 kPa between copper electrodes at an interelectrode distance of 2 mm, a sufficiently uniform in space intense nanosecond discharge with a pulsed electric power of up to 1.2 MW was ignited, and the energy contribution to plasma in one pulse was 176 mJ.

The study of the spectral characteristics of plasma based on vapor-gas mixtures Cu : Ar showed that the

most intense were the spectral resonance spectral lines of the Cu atom (λ = 324.75 and 327.39 nm Cu 1) from the lines of a singly charged Cu ion in a range of 200–330 nm, the most intense was the line λ = 227.62 nm Cu 11. A line with λ = 618.86 nm Cu 11 was distinguished from ionic spectral lines in the visible region of the spectrum. Automatic irradiation of the substrate and film nuclei on the substrate with hard UV radiation of Cu atoms and ions from the discharge plasma is promising for influencing the electrical characteristics of the synthesized films, in particular, reducing their resistance [29].

The maximum value of the average power of UV radiation at p (Ar) = 101 kPa was observed for the UV-A range, and the absolute radiation densities in various UV ranges reached: UV-C – 67 mW/m^2, UV-B – 65 mW/m^2 and UV-A – 204 mW/m^2 (at U_{ch} = 20 kV, f = 1 kHz), for a discharge at p (Ar) = 6.7 kPa, the radiation power decreased by more than an order of magnitude. The most effective was an increase in the pulse repetition rate to 1000 Hz rather than an increase in the value of the charging voltage across the working capacitor of the high-voltage modulator. However, when the UV radiation source operates in the frequency range of 300–1000 Hz, it is necessary to use forced cooling of the discharge device.

In plasma on a gas-vapor mixture Ar:Cu at an Ar pressure of 101 kPa, the average electron energy increased from 1.493 to 24.72 eV, with an increase in the E/N parameter from 1 to 1800 Td. The electron temperature was 71560.4, 152076, and 267 844 K at the values of the parameter E/N = 63 Td, 807 Td, and 1676 Td, respectively. The values of the electron concentration were in a range of 5.3×10^{19} m^{-3}–3.8×10^{19} m^3 at a current density of (7.65–6.12) × 10^6 A/m^2 on the electrode surface with a value of the parameter E/N = 1676 Td, which is realized in the discharge in the first 100 ns and for the reduced electric field strength E/N = 807 Td, which existed in the discharge gap after a time of 100 ns. Specific losses of the discharge power in the vapor-gas mixture Ar:Cu = 101 kPa – 30 Pa for inelastic processes of collisions of electrons with the components of the mixture were the largest for the Ar atom and reached 76% at the value of the parameter E/N = 63 Td and 80% at the same value of the reduced electric field strength for the vapor-gas mixture Ar:Cu = 202 kPa – 30 Pa, and for the Cu atom they had a maximum value of 70% for the excitation of the resonance state $^2P_{3/2,1/2}$ in the mixture Ar:Cu = 101 kPa – 30 Pa for a reduced electric field strength of 1 Td. The rate constants of electronic processes varied in a range of $0.2140 \times 10^{-16} - 0.1178 \times 10^{-11}$ m^3/s. The rate constant of excitation of the resonance level of the Cu atom in the first mixture of Cu vapors with Ar was 0.9495×10^{-12} m^3 /s and 0.7831×10^{-12} m^3/s for the experimental value of the reduced electric field strength E/N 1676 Td and 807 Td, and for the second gas-vapor mixture (p (Ar) = 202 kPa), it had lower values – 0.8916×10^{-12} m^3/s and 0.7184×10^{-12} m^3/s for E/N 1242 Td and 621 Td.

CONFICT OF INTEREST

The authors declare that they have no conflicts of interest.

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