ISSN 1068-3755, Surface Engineering and Applied Electrochemistry, 2023, Vol. 59, No. 3, pp. 362–367. © Allerton Press, Inc., 2023.
Russian Text © The Author(s), 2023, published in Elektronnaya Obrabotka Materialov, 2023, No. 3, pp. 116–122.

Gas Discharge Point Source of UV Radiation Based on Argon–Copper Gas–Vapor Mixture

A. K. Shuaibov[a, *], R. V. Gritsak[a], A. I. Minya[a], Z. T. Gomoki[a], A. A. Malinina[a], A. N. Malinin[a], and M. I. Vatrala[a]

[a] *Uzhgorod National University, Uzhgorod, 88000 Ukraine*

**e-mail: alexsander.shuaibov@uzhnu.edu.ua*

Received February 23, 2022; revised July 15, 2022; accepted July 19, 2022

Abstract—The characteristics of a pulse periodic source of a long-range UV radiation with overvoltage pumping by a bipolar discharge of nanosecond duration between copper electrodes in argon at atmospheric pressure are investigated. Copper vapors were introduced into the discharge due to the ectonic mechanism when a sufficient amount of the electrode material vapors is introduced into the discharge gap due to microexplosions of inhomogeneities of the surface of metal electrodes in a strong electric field of an overvoltage high-current nanosecond discharge. The characteristics of an overvoltage nanosecond discharge at a distance between the electrodes of 2 mm are studied. The emission spectra of the discharge were analyzed, and the intensity of the UV radiation of a point emitter was optimized depending on the supply voltage of the high-voltage modulator and the repetition rate of discharge pulses. The identification of the emission spectra of plasma made it possible to establish the main excited plasma products that form the spectrum of the UV radiation of the plasma. The study of the spectral characteristics of plasma based on gas-vapor mixtures “copper–argon” showed that the most intense were the spectral resonance lines of the copper atom and ion. It was found that a space-uniform overvoltage nanosecond discharge was ignited between copper electrodes at an interelectrode distance of 2 mm. It was found that the maximum value of the average UV power at p(Ar) = 101 kPa was observed for the UV-A range.

Keywords: overvoltage bipolar discharge of nanosecond duration, plasma, argon, copper, radiation

DOI: 10.3103/S1068375523030158

INTRODUCTION

To design pulse-periodic sources of ultraviolet (UV) radiation, the channel stage of spark discharge in inert high-pressure gases is of significant interest since it has a high radiation intensity in the range of wavelengths 200–250 nm [1]. However, when investigating short-pulse discharges (τ = 50–150 ns) in gases with ectonic mechanism of introducing the vapors of electrode material, it is important to optimize the device operation modes at the channel stage of discharge development [2].

The earlier research of spark high-pressure discharge [3] showed that the average gas density in its plasma channel at the late stages was $\approx 5 \times 10^{-6}$ g/cm^3, and this corresponded to the plasma medium concentration at the level of $\approx 10^{17}$ cm^{-3}. The average plasma temperature was as high as 40×10^4 K [4].

The results of the studied unipolar nanosecond discharge between copper electrodes at the distance between the electrodes d = 1–2 mm in air at the atmospheric pressure show the existence of intense spectral lines Cu I, Cu II in the range of wavelengths 200–230 nm. A transfer to a bipolar nanosecond discharge in gases at the atmospheric pressure was performed for the uniform application of electrode material in our following investigations [5–7].

Thus, it was established in [5] that this discharge is a source of copper oxide nanostructures that can be deposited on a solid substrate arranged at the discharge. It was shown in [6] that this discharge in air at the atmospheric pressure is a source of UV radiation fluxes of atoms, copper ions, and copper oxide nanoparticles, which can be used in medicine, biology, and biomedical engineering. To produce a flow of pure copper nanostructures together with the UV radiation flux, it is promising to investigate the characteristics of the bipolar nanosecond discharge between copper electrodes in argon.

In [7] are presented the study results for the characteristics of the overvoltage bipolar discharge at the atmospheric pressure between the electrodes of aluminum for τ = 50–400 ns when a sufficiently intense radiation of little nanoparticles of aluminum oxide and UV radiation of atoms and aluminum ions was found.

The experimental results for the studied spark channel characteristics in argon at the atmospheric pressure between the electrodes of aluminum both in magnetic field and without it are presented in [8]. The results of systematic research of the influence of the longitudinal magnetic field on the main characteristics of spark discharge in argon at the atmospheric pressure with aluminum vapor impurities that were introduced into the plasma when forming the ectons on the electrode surfaces are presented in [9]. It is shown that the magnetic field shifts the maximum of the spectral density of radiation into the UV range of the spectrum, decreases the rate of the spark channel expansion and the losses for the transverse radiation, and increases the specific electrical power of the discharge, the conductivity and the temperature of plasma at its arc stage.

No investigation results for the characteristics of the UV emitter with overvoltage pumping by the bipolar discharge of the argon–copper gas–vapor mixture at the atmospheric pressure can be found in the literature; thus, it is an urgent task to optimize this source of UF radiation in order to be applied in medicine, biology, and biomedical engineering.

The paper presents the results of investigation of spacial, electrical, and optical characteristics of the overvoltage nanosecond bipolar discharge at the atmospheric pressure in argon with copper vapor impurity, which was introduced into plasma due to the formation of ectons on the working surfaces of copper electrodes in a strong electric field.

EXPERIMENTAL

The parameters of the overvoltage nanosecond discharge were studied using the discharge module whose diagram is presented in [1, 5, 6]. The overvoltage nanosecond discharge between the electrodes of copper was ignited in the sealed chamber of plexiglas at the distance between copper electrodes $d = 2$ mm and the pressure of argon 6.7; 101 kPa. The experimental setup contained the discharge module, the registration system for an optical radiation based on the MDR-2 monochromator, and the registration system for pulse, electrical, and optical characteristics. Bipolar high voltage pulses 50–150 ns long and $\pm(20–40)$ kV in amplitude were applied to the discharge cell electrodes to ignite the discharge. The voltage pulse repetition frequency was chosen as 40–150 Hz. The oscillograms of voltage pulses and current pulses were recorded using a wide-band capacitive voltage divider, the Rogowski toroidal coil with the time separation at a level of 2–3 ns.

A uniform discharge 100–400 ns long with a current pulse amplitude 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 overvoltaged, which created favorable conditions for the formation of a beam of runaway electrons with high energy and accompanying X-ray radiation [10, 11].

The discharge plasma radiation power was measured using the TKA-PKM ultraviolet meter of absolute radiation power, which makes it possible to carry out measurements within the spectral range from 200 to 400 nm.

The air in the discharge chamber was pumped out using the backing pump up to a residual pressure of 10 Pa. The diameter of copper cylindrical electrodes was 5 mm, and the radius of curvature of their working end surface was the same (3 mm). The discharge capacity depended on the voltage pulse repetition frequency. The “point discharge” mode was achieved only at the voltage pulse repetition frequencies within the range $f = 40–150$ Hz. At a short-term increase in frequency up to 1000 Hz, the capacity of the gas-discharge emitter plasma grew to 120–150 mm^3.

CHARACTERISTICS OF UV EMITTER

Figure 1 represents the time-averaged pictures of the overvoltage nanosecond discharge at the pressures p(Ar) = 7.6; 101kPa taken at a photocamera time exposure of ≈ 1 sec. At the pressure of argon 6.7 kPa, the overvoltage nanosecond discharge had a form of a bright central part approximately 2 mm in diameter that was equal to the interelectrode gap from where two jets of green plasma flew out. With the increase in argon pressure up to 101 kPa, the diameter of the bright central part grew by a factor of two to three.

The oscillograms of voltage and current were in the form of time-fading oscillations due to the inconsistency in the output resistance of the high-voltage modulator with the resistance to load. The full duration of voltage oscillations in the gap and the discharge current reached 450 ns at the duration of single oscillations of voltage 7–10 ns, and the current oscillations had a duration of ≈70 ns. For the discharge in argon at the atmospheric pressure at $d = 2$ mm, the amplitude of the largest voltage drop on the electrodes was achieved at the initial stage of discharge, and it was ≈ ±7–8 kV, the current was ±100, and the pulse power was 1.2 MW, which ensured the energy contribution during one discharge pulse into plasma of approximately ≈167 MJ.

Figure 2 represents the parts of the radiation spectrum of the nanosecond high-voltage discharge between copper electrodes in argon (p(Ar) = 101 kPa) at an interelectrode distance of $d = 2$ mm, and the results of its identification are tabulated.

The interpretation of the spectrum of plasma radiation of the nanosecond discharge between copper electrodes are presented in Fig. 2.

The peculiarity of these radiation spectra is the existence of the continuum against the background of which there are all the spectral lines and bands. The

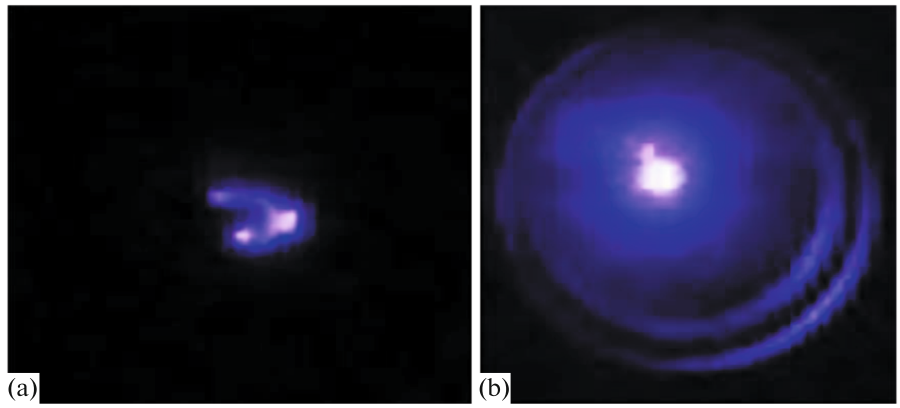

Fig. 1. Pictures of overvoltage nanosecond discharge between copper electrodes in argon at: p(Ar) = (a) 6.7 and (b) 101 kPa for $d = 2$ mm.

nature of this continuum under the conditions of our research is associated with the heat and recombination radiation of plasma. The form of the continuum with a wide maximum at 400–450 nm is in good agreement with the results known in the literature.

Thus, for the spark discharge in argon at the atmospheric pressure between the electrodes of aluminum at d = 10 mm in the radiation spectrum there was recorded in [7] an intense continuum in the wavelength range from 350 to 460 nm with the maximum at $\lambda = 420$ nm.

On the parts of the radiation spectrum of the discharge plasma (Fig. 2) on the mixture of argon with copper vapors in the UV range of wavelengths from 214 to 330 nm (lines 1–23, see Table 1) there was observed the radiation on the transitions of atom and single-charge copper ion. The most intense ion spectral line was a line with $\lambda = 227.62$ nm Cu II, and the most intense atomic lines were the resonant spectral lines of copper with $\lambda = 324.75$ and 327.39 nm Cu I, for which the main level is the lower energy level.

The highest energy of the upper level for lines Cu II was $E_{\text{upper}} = 18.77$ eV, while the maximum energy of the upper level was $E_{\text{upper}} = 7.18$ eV for the atomic lines. In the UV spectrum, there were also observed two bands of the second positive system of the nitrogen molecule since the evacuation of air from the discharge chamber appeared in these experiments only up to the residual pressure of air ≈10 Pa. In the range of wavelengths 360–659 nm, there was radiation mainly on transitions Ar I, Ar II, and also one intense spectral line of hydrogen atom ($\lambda = 656.29$ H_{α}) due to the residual pressure of air in the chamber after its pumping using the backing pump.

At an interelectrode distance of 2 mm and the usage of the bipolar generator of high voltage nanosecond pulses, there is formed one or several (at a strong overvoltage of the discharge gap) cathode spots which move towards one another.

For the high-voltage nanosecond discharge in argon at the atmospheric pressure, some optic and gas-dynamic characteristics of the cathode plasma are

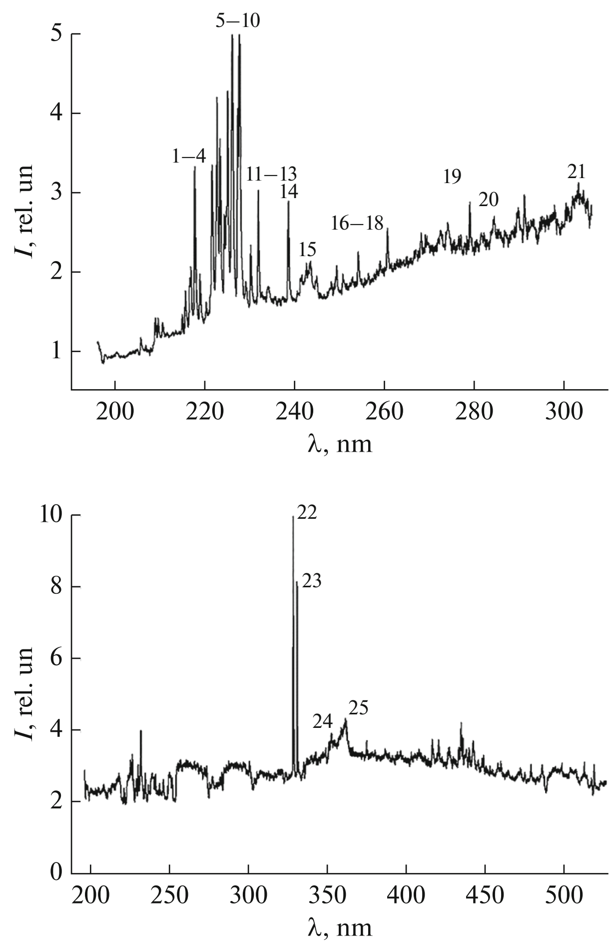

Fig. 2. Spectrum sections of radiation of nanosecond discharge in argon between copper electrodes at d = 2 mm and p(Ar) = 101kPa.

Table 1.

No.	λ_{tabl}, nm	$I_{exp.}$ rel.un.	Object	E_{lower}, eV	E_{upper}, eV	Configuration and term of the lower level	Configuration and term of the upper level
1	214.89	1.78	Cu II	1.39	7.18	$3d^84s^2\ ^2D$	$3d^95f\ ^2F^o$
2	216.50	2.07	Cu I	0.00	5.72	$3d^{10}4s\ ^2S$	$4p'\ ^2D^o$
3	217.49	3.34	Cu II	8.92	14.61	$4p\ ^1F^o$	$4d\ ^1G$
4	220.05	1.63	Cu II	9.06	14.70	$4p\ ^3D^o$	$4d\ ^3F$
5	221.02	3.34	Cu II	3.26	8.86	$4s\ ^1D$	$4p\ ^3D^o$
6	221.45	4.21	Cu I	1.39	6.98	$4s^2\ ^2D$	$4p''\ ^2P^o$
7	223.84	3.69	Cu I	1.64	7.18	$4s^2\ ^2D$	$5f\ ^2F^o$
8	224.70	4.30	Cu II	2.72	8.23	$4s\ ^3D$	$4p\ ^3P^o$
9	226.30	5.00	Cu I	1.64	7.12	$4s^2\ ^2D$	$7p\ ^2P^o$
10	227.62	5.05	Cu II	2.98	8.42	$4s\ ^3D$	$4p\ ^3P^o$
11	229.43	1.89	Cu II	2.83	8.23	$4s\ ^3D$	$4p\ ^3P^o$
12	230.131	2.34	Cu I	1.64	7.02	$4s^2\ ^2D$	$4p''\ ^2D^o$
13	236.98	1.82	Cu II	3.26	8.49	$4s\ ^1D$	$4p\ ^3F^o$
14	239.26	2.86	Cu I	1.64	6.82	$4s^2\ ^2D$	$6p\ ^2P^o$
15	244.16	2.12	Cu I	0.00	5.08	$4s\ ^2S$	$4p'\ ^4P^o$
16	249.21	2.08	Cu I	0.00	4.97	$4s\ ^2S$	$4p'\ ^4P^o$
17	254.48	2.25	Cu II	8.52	13.39	$4p\ ^3F^o$	$5s\ ^3D$
18	261.83	2.55	Cu I	1.39	6.12	$4s^2\ ^2D$	$5p\ ^2P^o$
19	279.17	2.86	Cu II	14.33	18.77	$4d\ ^3G$	$6f\ ^3H^o$
20	282.43	2.5	Cu I	1.39	5.78	$4s^2\ ^2D$	$4p'\ ^2D^o$
21	306.34	3.13	Cu I	1.64	5.68	$4s^2\ ^2D$	$4p'\ ^2P^o$
22	324.75	9.98	Cu I	0	3.82	$3d^{10}4s\ ^2S$	$4p\ ^2P^o$
23	327.39	8.19	Cu I	0	3.39	$3d^{10}4s\ ^2S$	$4p\ ^2P^o$
24	354.89	4.10	N_2	Second positive system $C^3\Pi_u{}^+$-$B^3\Pi_g{}^+$ (3;2)			
25	357.69	4.37	N_2	Second positive system $C^3\Pi_u{}^+$-$B^3\Pi_g{}^+$ (0;1)			
26	420.06	5.04	Ar I	11.55	14.50	4s [½]^o	5p [2½]
27	425.93	4.99	Ar I	11.83	14.74	4s' [½]^o	5p' [½]
28	430.01	3.95	Ar I	11.62	14.51	4s [1½]^o	5p [1½]
29	433.35	5	Ar I	11.83	14.69	4s' [½]^o	5p' [1½]
30	436.37	3.97	Ar I	11.62	14.46	4s [1½]^o	5p [½]
31	442.39	5.13	Ar I	11.72	14.52	4s' [½]^o	5p' [1½]
32	451.07	4.25	Ar I	11.83	14.58	4s' [½]^o	5p [½]
33	459.60	3.25	Ar I	11.83	14.52	4s' [½]^o	5p' [1½]
34	460.95	3.32	Ar II	18.45	21.14	$4s'\ ^2D$	$4p'\ ^2F^o$
35	462.84	3.96	Ar I	11.83	14.51	4s' [½]^o	5p [2½]
36	470.23	3.03	Ar I	11.83	14.46	4s' [½]^o	5p' [1½]
37	483.66	4.18	Ar I	12.91	15.47	4p [½]	9s [1½]
38	487.62	3.01	Ar I	12.91	15.45	4p [½]	7d [1½]

Table 1. (Contd.)

No.	λ_{tabl}, nm	$I_{exp.}$ rel.un.	Object	E_{lower}, eV	E_{upper}, eV	Configuration and term of the lower level	Configuration and term of the upper level
39	492.10	3.39	Ar I	13.09	15.61	4p [2½]	10d [3½]
40	511.82	2.47	Ar I	13.09	15.52	4p [2½]	6d' [2½]^o
41	516.22	2.98	Ar I	12.91	15.31	4p [½]	6d [½]
42	518.77	3.36	Ar I	12.91	15.30	4p [½]	5d' [1½]^o
43	521.82	2.4	Cu I	3.82	6.19	$3d^{10}4p\ ^2P^o$	$3d^{10}4d\ ^2D$
44	525.27	4.53	Ar I	13.09	15.45	4p [2½]	7d [3½]
45	570.02	1.96	Cu I	1.64	3.82	$3d^94s^2\ ^2D$	$3d^{10}4p\ ^2P^o$
46	601.36	1.92	Ar I	13.08	15.14	4p [2½]	5d [½]^o
47	617.01	1.84	Ar I	13.17	15.18	4p [1½]	7s [1½]^o
48	653.81	3.17	Ar I	13.08	14.95	4p [2½]	4d' [½]^o
49	659.61	2.85	Ar I	13.08	14.95	4p [2½]	4d' [½]^o

presented in [8, 9]. However, in these experiments carried out at an interelectrode distance of d = 10 mm, between the electrodes of aluminum there was clearly recorded the radiation of the plasma cathode spot ≈0.2–0.3 mm in diameter and the associated diffusion discharge plasma that occupied the greater part of the discharge gap. The formation of cathode spots in the overvoltage nanosecond discharge is explained by the explosive emission model [2] based on the considerable energy on the micro tips of the cathode surface with the subsequent heating and explosion of them. The cathode spot formed simultaneously with the beginning of a sharp increase in current and voltage drop on the gap, and it was hemispherical in the initial stages of formation and had an expansion rate of ≈2.5×10^6 cm/s. The kinetic energy 50–100 eV, at which the ecton with a number of electrons of 10^{11}–10^{12} and a composite of metal vapors, corresponded to this motion speed of the cathode spot. At the initial stage of discharge, the cathode spot radius was not more than ≈10^{-6} m, and it increased from 0.5×10^{-4} to 4.3×10^{-4} cm during 5–50 ns [8].

The temperature of electrons of the near-cathode argon plasma at the atmospheric pressure that was estimated according to the radiation intensities of the argon spectral lines at the initial moments of its formation (over the time interval ≈30–40 ns) reached 5 eV, and it reduced from 4.2 to 3.4 eV in the following moments of time (τ = 50–50 ns) [8].

Figure 3 represents the optimization results for the average intensity of the discharge UV radiation versus the pulse repetition frequency.

The maximum values of the average power of the UV radiation in argon at the argon pressure 101 kPa for different ranges of the UV radiation was as follows: 67 MW/m^2 for UV-C (200–280 nm), 65 MW/m^2 for UV-B (280–315 nm), and 204 MW/m^2 for UV-A (315–400 nm) (at U = 20 kV, f = 1 kHz).

When the pulse repetition frequency increased from 40 to 1000 Hz, the growth in the discharge radiation intensity was the highest in the UV-A (315–400 nm) range; in particular, in the range of frequencies Δf = 350–1000 Hz, it increased from 15 to 95 MW/m^2. For the ranges UV-B and UV-C, the growth in the radiation power density was much less (from 3.0 to 25.0 MW/m^2).

The increase in the discharge UV radiation intensity depending on the discharge voltage value on the

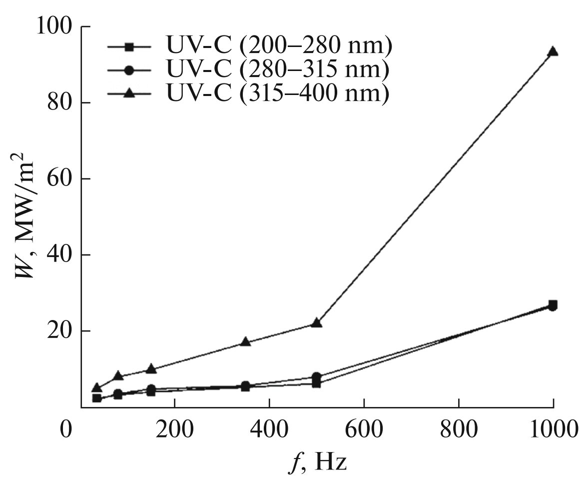

Fig. 3. Intensity of UV-radiation of UV-C, UV-B, and UV-A; ranges of high voltage nanosecond discharge versus pulse repetition frequency at charge voltage U = 13 kV (f = 80 Hz, p(Ar) = 101 kPa, d = 2 mm).

working capacitor of the high-voltage modulator at the fixed repetition frequency was less effective (in the range 8.0–24 MW/m^2). Furthermore, the relative ratios between the intensities of radiation in the ranges UV-C, UV-B, and UV-A were the same as for the corresponding dependences on frequency.

CONCLUSIONS

It is established that, at the pressure of argon 101 kPa, between the electrodes of copper at the interelectrode distance 2 mm, there was ignited a space-uniform overvoltage nanosecond discharge with the pulse electrical power up to 1.2 MW, and the energy contribution into plasma for one pulse was 176 mJ. The investigation of the spectral characteristics of plasma based on the copper-argon vapor-gas mixtures showed that the resonant spectral lines of copper atom were the most intense (λ = 324.75 and 327.39 nm Cu I); of the lines of the singly-charged copper ion in the range 200–330 nm, the line λ = 227.62 nm was the most intense. The copper ion line with λ = 618.86 nm stood out from the spectral lines in the visible range of the spectrum.

The maximum value of the average power UV radiation at p(Ar) = 101 kPa was for the UV-A range at the absolute radiation density 204 MW/m^2. The most effective was the increase in the pulse repetition frequency up to 1000 Hz but not the growth in the value of charge voltage on the working capacitor of the high voltage modulator.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

REFERENCES

1. Shuaibov, O.K., Malinina, A.O., and Malinin, O.M., *Novi hazorozryadni metody oderzhannya selektyvnoho ul'trafioletovoho i vydymoho vyprominyuvannya ta syntezu nanostruktur oksydiv perekhidnykh metaliv: monohrafiya* (New Gas-Discharge Methods for Obtaining Selective Ultraviolet and Visible Radiation and Synthesis of Nanostructures of Transition Metal Oxides: Monograph), Uzhhorod: Hoverla, 2019.
2. Mesyats, G.A., Ecton or electron avalanche from metal, *Phys.-Usp.,* 1995, vol. 38, no. 6, p. 601. https://doi.org/10.1070/PU1995v038n06ABEH000089
3. Marshak, I.S., Electric breakdown of gases at pressures close to atmospheric pressure, *Sov. Phys. Usp.,* 1960, vol. 3, no. 4, p. 624. https://doi.org/10.1070/PU1961v003n04ABEH003314
4. Shuaibov, A.K., Laslov, G.E., and Kozak, Ya.Ya., Emission characteristics of the cathode region of nanosecond discharge in atmospheric-pressure air, *Opt. Spectrosc.,* 2014, vol. 116, no. 4, p. 552. https://doi.org/10.1134/S0030400X14030199
5. Shuaibov, A., Minya, A., Malinina, A., Malinin, A., et al., Synthesis of nanostructured transition metal oxides by a nanosecond discharge in air with assistance of the deposition process by plasma UV-radiation, *Adv. Nat. Sci.: Nanosci. Nanotechnol.,* 2018, vol. 9, no. 3, p. 035018. https://doi.org/10.1088/2043-6254/aadc4b
6. Shuaibov, A.K., Minya, A.Y., Malinina, A.A., Malinin, A.N., et al., Study into synchronous flows of bactericidal ultraviolet radiation and transition oxides metals (Zn, Cu, Fe) in a pulsed gas discharge overvoltage reactor nanosecond discharge in the air, *Surf. Eng. Appl. Electrochem.,* 2020, vol. 56, no. 4, p. 510. https://doi.org/10.3103/S106837552004016X
7. Shuaibov, A.K., Minya, A.Y., Malinina, A.A., Malinin, A.N., et al., Study of the formation conditions of aluminum oxide nanoparticles in an overstressed nanosecond discharge between aluminum electrodes in a mixture of nitrogen and oxygen, *J. Metal. Mater. Res.,* 2020, vol. 3, no. 2, p. 37. https://doi.org/10.30564/jmmr.v3i2.2441
8. Kurbanismailov, V.S., Omarov, O.A., and Ragimkhanov, G.B., Radiative and spectral characteristics of a spark channel in argon, *Prikl. Fiz.,* 2014, vol. 2, no. 3, p. 35.
9. Omarov, O.A., Omarova, N.O., Omarova, P.Kh., and Aliverdiev, A.A., Breakdown of high-pressure gases in a longitudinal magnetic field, *Teplofiz. Vys. Temp.,* 2019, vol. 57, no. 2, p. 174. https://doi.org/10.1134/S0040364419020169
10. Tarasenko, V.F., *Runaway Electrons Preconized Diffuse Discharge,* New York: Nova Science, 2014.
11. Beloplotov, D.V., Lomaev, V.I., Sorokin, D.A., and Tarasenko, V.F., Blue and green jets in laboratory discharges initiated by runaway electrons, *J. Phys.: Conf. Ser.,* 2015, vol. 652, p. 012012. https://doi.org/10.1088/1742-6596/652/1/012012

Translated by M. Myshkina