INVESTIGATIONS ON THE PARAMETERS OF MULTICOMPONENT GAS-METAL PLASMA FORMED IN THE MIXED MOLECULAR GASES

Yu.V. Kovtun, E.V. Syus'ko, E.I. Skibenko, A.I. Skibenko, A.N. Shapoval, V.B. Yuferov

National Science Center "Kharkov Institute of Physics and Technology", Kharkov, Ukraine

E-mail: Ykovtun@kipt.kharkov.ua

Studies have been made into the parameters of dense, multicomponent gas-metal plasma formed in the operating medium consisting of molecular gases such as nitrogen and oxygen, and also, the component metal *Ti.* Charge and elemental compositions of the generated plasma have been determined. Measurements were taken for the parametric and time dependences of the discharge current and the average density of plasma formed in the pulsed reflex discharge.

PACS: 52.80.-s; 52.80.Sm

INTRODUCTION

The plasma generated in the reflex discharge resides in the electric **E** and magnetic **B** crossed fields, resulting in the rotation of the electronic and ionic components of plasma. So, the reflex discharge is one of the possible variants of creating rotating-plasma devices [1]. The use of these devices for separating the substance into mass groups and elements is one of the factors that stimulates the investigation of the rotating plasma [2, 3].

Previously (e.g., see [4-7]), experimental studies have been made on the multicomponent gas-metal plasma, which was generated in the high-power pulsed reflex discharge in the operating medium that included the gas components (H_2, *Ar*, 88.9%*Kr*-7%*Xe*-4%N_2-0.1%O_2) and the component metal coming to the discharge due to *Ti* cathode sputtering. The experiments have registered the existence of two modes of pulsed discharge burning, which differed from each other in the value of maximum attainable plasma density and its dynamics of variation [6]. In such a case, the observed difference in the plasma density and its dynamic behavior is attributed to different physical mechanisms of plasma generation in the two modes. The first mode is characterized by the formation of dense multicomponent gas-metal plasma, the density of which reaches $\leq 2\cdot10^{14}$ cm^{-3} with a high degree of ionization (of about 100 %). In the second mode, the maximum plasma density ranges between $\sim 2\cdot10^{12}$ and $9\cdot10^{12}$ cm^{-3}, with the degree of ionization of $\leq$ 25 %.

The studies of the multicomponent gas-metal plasma generated in the molecular gas mixture are of definite interest. Firstly, at plasma separation of spent nuclear fuel, the multicomponent plasma will comprise molecular oxygen compounds. Secondly, in the multicomponent gas-metal plasma having the mixture of such molecular gases as nitrogen and oxygen, the plasma-chemical reactions may result in the synthesis of various compounds [8]. Thirdly, in the rotating plasma devices, including those for substance separation, modes of off-normal (emergency) operation may arise, For example, there may be a full or partial seal failure (leaks) in the vacuum chamber. This would lead to the inlet of free air to the plasma volume. In turn, this may cause changes in the mode of plasma device operation right down to emergency shut-down.

So, the present paper reports the experimental results for the multicomponent gas-metal plasma formed in the operating medium consisting of molecular gases such as nitrogen and oxygen, and also, the component metal *Ti.* Basically, it is the first mode of high-power impulse reflex discharge burning that was realized here.

1. EXPERIMENTAL INSTALLATION AND DIAGNOSTIC TECHNIQUES

The parameters of the multicomponent gas-metal plasma were investigated at the "MAKET" facility [7] representing providing the high-power impulse reflex discharge. The vacuum chamber was pumped down to a pressure of $1.33\cdot10^{-4}$ Pa; then the igniting gas mixture was let into the chamber. The gas-metal plasma was formed as a result of the capacitor bank discharge (C = 560 μF, $U_0 \leq$ 5 kV) between two cold cathodes, each of diameter Ø = 10 cm, and the anode, Ø = 20 cm. The cathodes were made from monometallic *Ti,* and atmosphere air was used as an igniting gas mixture with the main components [9]: N_2 (78.084 %), O_2 (20.946 %), H_2O vapors (~3%), *Ar* (0.934 %), CO_2 (0.038 %).

The multicomponent gas-metal plasma density was investigated by microwave methods of plasma diagnostics, namely, the microwave interferometry method and the microwave signal cutoff technique. The plasma probing was performed across the plasma column using the ordinary O-wave with the wavelength λ=0.8 cm. Along with the density measurements, the discharge current was measured by means of the Rogowski loop (coil). The signals were registered by the digital oscilloscope *Velleman PCS* 500 (50 MHz).

To determine the charge and elemental compositions of plasma, the noncontact passive method of optical emission plasma spectroscopy [10] was used. The method is based on the registration of the spontaneous emission spectrum from excited atoms, molecules and ions of plasma, followed by identification of lines and their groups in accord with belonging to certain particles and their quantum states responsible for the transitions. The line spectrum of the plasma was registered in the spectral range from 214 to 673 nm by the *SL*-40-2 Type spectrometer. That permitted a simultaneous registration of the spectrum in visible and near ultraviolet bands. The spectrometer was operated in the accumulation mode, i.e., it detected the integral line emission spectrum from the plasma in all the time of plasma existence. The emission was registered through the diagnostic window made from the fused quartz glass *KU1*.

ISSN 1562-6016. ВАНТ. 2017. №1(107)

The identification of spectral lines was based on the data of refs. [10-14].

2. EXPERIMENTAL RESULTS AND DISCUSSION

Fig. 1 shows the time dependence of the mean plasma density. By convention, the time behavior of the mean gas-metal plasma density can be divided into three stages [4, 5]. The first stage consists in the plasma creation and its density increase up to $N_e = 1.7\cdot10^{13}$ cm^{-3} (see Fig. 1, the time interval t_1). In this stage, the gas gap breakdown takes place, and the self-maintained discharge with the cold cathodes gets ignited. The formation of weakly ionized plasma $N_e < 1\cdot10^{11}$ cm^{-3} occurs due to ionization of neutral molecules (atoms) by primary electrons emitted from the cathode surfaces. Note that the plasma electrons can also contribute to the gas ionization. In this case, the cathode sputtering takes place and the sputtered atoms come to the plasma. As calculations made in ref. [7] show, at the density $N_e = 1\cdot10^{11}$ cm^{-3} the degree of *Ti* ionization comes up to $3\cdot10^{-3}$. Later on, as a highly ionized plasma is formed with the density reaching $N_e = 1.7\cdot10^{13}$ cm^{-3}, a high degree of ionization (≈ 0.98) of the sputtered cathode material (*Ti*) is observed [7]. Fig. 2,*a* shows the time t_1 of achieving the plasma density $N_e = 1.7\cdot10^{13}$ cm^{-3} versus the discharge voltage. As is obvious from Fig. 2,*a*, the time t_1 only slightly changes in the given discharge voltage range. For example, at U_{max} = 3.8 kV we have $t_1 \approx 90$ µs, and at U_{max} = 4.2 kV the time t_1 is ≈ 86 µs.

The second stage presents the existence of the dense ($N_e \geq 1.7\cdot10^{13}$ cm^{-3}) highly ionized plasma (see Fig. 1, the time interval t_2). In this stage, a further increase in the plasma density takes place up to the maximum value, which is determined by the balance between the processes of plasma particle production and losses. In this case, the plasma density can attain $N_e \sim 10^{14}$ cm^{-3} and more [7]. At a later time, the processes leading to plasma particle losses become predominant, and the plasma density decreases. Here we note that out of the processes leading to plasma losses, it is the processes of plasma recombination and diffusion across the magnetic field with leaving for the magnetic mirror that are dominant. Fig. 2,*b* shows the time t_2 of dense plasma lifetime as a function of the discharge voltage which changes only slightly in the given range.

The third stage presents the plasma density decrease and decay (see Fig. 1, the time interval t_3). The analysis of the time dependence of the reciprocal plasma density N_e^{-1} has shown that at the initial stage of the plasma decay (t = 2.2 to 2.9 ms) the process of recombination is dominant (the linear law of density variation [15]). Then, starting with $t > 2.9$ ms, the N_p^{-1} value changes by the exponential law. This indicates that the main channel of plasma particle losses is diffusion [15]. Fig. 2,*c* shows the lifetime t_3 of the mean plasma density $1.7\cdot10^{13}$ cm$^{-3} \geq N_e \geq 1\cdot10^{12}$ cm^{-3} as a function of the discharge voltage. Here, with an increasing discharge volage, the lifetime t_3 of the plasma density decreases from 10 down to 8.9 ms. This behavior can be attributed to anomalous plasma decay in the reflex discharge [16, 17].

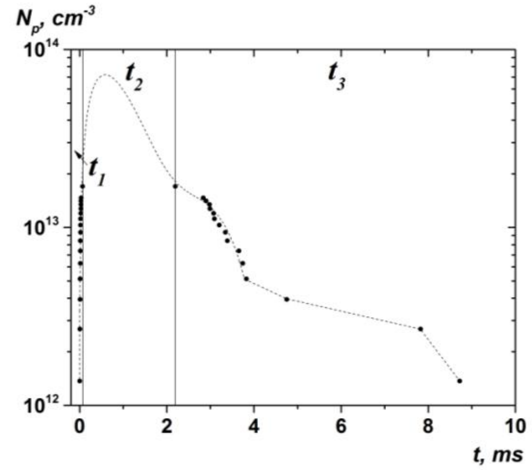

Fig. 1. Time dependence of the mean plasma density ($U_{dis.}$ = 3.8 kV, U_B = 1.5 kV; p = 0.8 Pa)

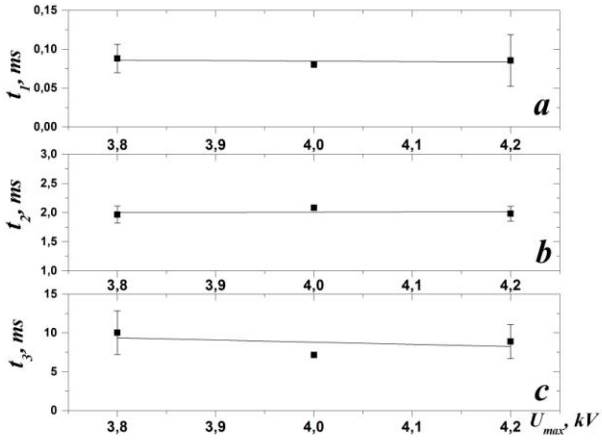

Fig. 2. Time t_1 of plasma density attainment $N_e = 1.7\cdot10^{13}$ cm^{-3} (a); lifetime t_2 of plasma density $N_e \geq 1.7\cdot10^{13}$ cm^{-3} (b); lifetime t_3 of the mean plasma density $1.7\cdot10^{13}$ cm$^{-3} \geq N_e \geq 1\cdot10^{12}$ cm^{-3} (c) as functions of the discharge voltage

However, the authors of ref. [17] observed the reduction of plasma density fluctuations, and consequently, the plasma stabilization as the discharge voltage increased.

The discharge current measurements versus applied voltage (Fig. 3,*b*) have shown that the peak current value I_{max} grows linearly with an increase in the applied voltage (Fig. 3,*b*), and reaches I_{max} = 1.77 kA at U_{max} = 4.2 kV. The time t_{Imax} of attaining I_{max}, being in fact the time of the leading edge of the discharge current, changes only slightly in the given discharge voltage range (Fig. 3,*a*).

The measured spectrum of line emission from reflex discharge plasma is given in Fig. 4. It exhibits a series of lines of excited Ti II ions: the 338.3769 nm line corresponds to the ground-state transition $3d^2(^3F)4s\ a^4F_{3/2}$ - $3d^2(^3F)4p\ z^4G^o_{5/2}$; 308.8042 nm, 334.9405 nm, 336.1227 nm, 337.2798 nm lines correspond to the energy-level transitions $3d^2(^3F)4s\ a^4F$ -$3d^2(^3F)4p\ z^4D^o$; the 323.4513 nm line corresponds to the transition transition $3d^2(^3F)4s\ a^4F$ -$3d^2(^3F)4p\ z^4F^o$; lines 368.5205 nm and 375.93 nm – to the transitions to the levels $3d^2(^3F)4s\ a^2F_{7/2}$, respectively, with $3d^2(^3F)4p\ z^4D^o_{5/2}$ and $z^2F^o_{7/2}$.

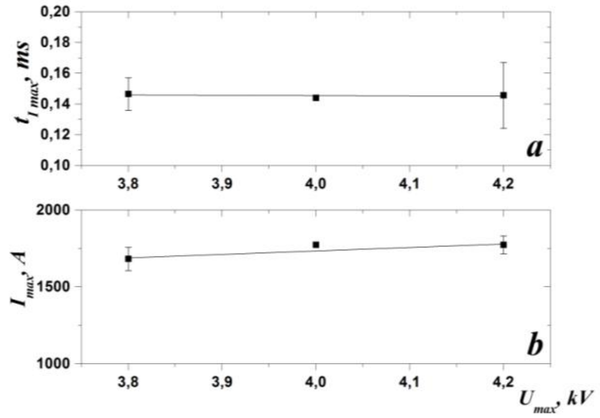

Fig. 3. Peak current values (b) and current-rise time (a) versus applied voltage

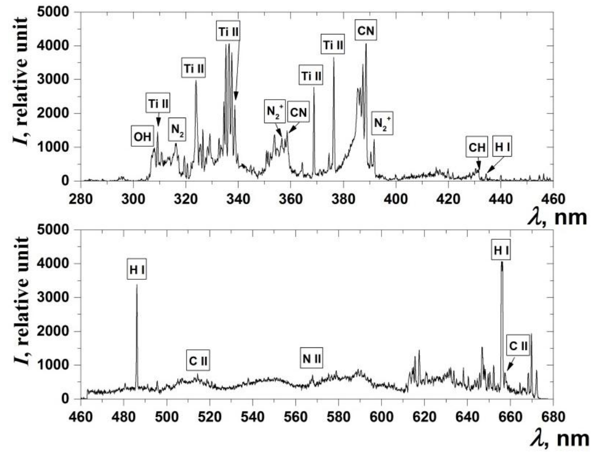

Fig. 4. Spectrogram of integral line emission from plasma (air + Ti). ($U_{dis.}$ = 4 kV, U_B = 1.3 kV; p = 0.7 Pa)

The presence of Ti^+ ions in the spectrum gives evidence of the entry of the cathode material into the plasma, and of its subsequent ionization, as has been observed previously, too, in [4, 5]. The spectrum shows the lines of molecular nitrogen (the basic air component): one of the positive system ($B^3\Pi_g$–$A^3\Sigma_u^+$) N_2, and one of the negative system ($B^2\Sigma_u^+$–$X^2\Sigma_g^+$) N_2^+; and also, the nitrogen ion line *N II* (567.956 nm). Besides, the emission spectrum (see Fig. 4) from the reflex discharge plasma exhibits he lines of the following atoms (ions): hydrogen (H_α – 656.28518 nm; H_β – 486.12789 nm, H_γ – 434.0462 nm), excited ions *C II* (514.516 nm, 657.805 nm); radicals: *OH* ($A^2\Sigma^2$–$X^2\Pi$), *CH* ($A^2\Delta$–$X^2\Pi$), the violet system ($B^2\Sigma^+$–$X^2\Sigma^+$) *CN*. The *H* and *OH* owe their presence in the plasma to the molecular dissociation of H_2O present in the discharge chamber. The inflow of *C, CH,* and *H* to the plasma is due to the occurrence of dissociation processes with the participation of hydrocarbons (CH_y, C_xH_y). The main contributor of hydrocarbons to the vacuum chamber is considered to be diffusion of the products of diffusion pump oil decomposition (cracking). In spite of the use of traps in diffusion pumps, which sufficiently reduce the operating fluid vapor flow to the vacuum chamber, the presence of hydrocarbons is practically always observed in vacuum chambers [18-20]. In the multicomponent plasma, various plasmochemical processes may occur, resulting, in this particular case, in the occurrence of the CN cyanogen radical produced due to the reactions of nitrogen with carbon-containing compounds. Furthermore, in the multicomponent gas-metal plasma, the synthesis of various compounds, e.g., *TiN*, may also take place [8, 21, 22]. According to the data of ref. [23], the optical spectrum of *TiN* lies in the spectral range 588…685 nm for the transitions $A^2\Pi$ - $X^2\Sigma$ and $B^2\Sigma$ - $A'^2\Delta$. The spectrum analysis of the multicomponent gas-metal plasma of the high-power impulse reflex discharge (see Fig. 4) has shown the presence of the lines in the spectrum, which may be related to the *TiN* lines, though in the given case they cannot be identified unambiguously as TiN lines. It should be noted that after realization of a few discharge pulses in the air+*Ti* plasma the vacuum chamber surface takes on golden color typical for *TiN* coatings.

CONCLUSIONS

The present studies have demonstrated that under the conditions of a pulsed reflex discharge in the molecular gas mixture (nitrogen and oxygen) a dense ($N_e \sim 10^{13}…10^{14}$ cm^{-3}) multicomponent gas-metal plasma is formed.

The elemental and charge compositions of the generated plasma have been determined to consist of excited titanium ions *Ti II* (cathode material); molecular N_2 and N_2^+ (ignition gas); excited atoms *H*; excited ions *C II*; radicals *OH* and *CH* (H_2O, CH_y, C_xH_y dissociation products); radical of *CN* cyanogen (result of plasmochemical reactions).

Parametric and time dependences of the mean plasma density have been measured experimentally at all stages of the discharge development (build-up, existence, decay.

REFERENCES

1. B. Lehnert. Rotating plasmas // *Nuclear Fusion*. 1971, v. 11, № 5, p. 485-533.
2. R. Gueroult, N.J. Fisch. Plasma mass filtering for separation of actinides from lanthanides // *Plasma Sources Sci. Technol.* 2014, v. 23, № 3, p. 035002.
3. V.B. Yuferov, V.V. Katrechko, A.S. Svichkar, et al. Problems of impurities deducing from multicomponent media at thermal heating, ionization and rotation of plasma in crossed fields // *Problems of Atomic Science and Technology. Series "Vacuum, Pure Materials, Superconductors"*. 2016, № 1(101), p. 124-129.
4. Yu.V. Kovtun, A.I. Skibenko, E.I. Skibenko, et al. Study of the Parameters of Hydrogen–Titanium Plasma in a Pulsed Reflective Discharge // *Plasma Physics Reports*. 2010, v. 36, № 12, p. 1065-1071.
5. Yu.V. Kovtun, A.I. Skibenko, E.I. Skibenko, et al. Study of multicomponent plasma parametrs in the pulsed reflex discharge // *Ukrainian Journal of Physics*. 2010, v. 55, № 12, p. 1269-1277
6. Yu.V. Kovtun, A.I. Skibenko, E.I. Skibenko, V.B. Yuferov. Measurement of the plasma density in two modes of pulsed discharge burning in the Penning ce // *Problems of Atomic Science and Technology. Series "Physics of plasma"*. 2015, № 1 (21), p. 197-200.

7. Yu.V. Kovtun. Features of Dense Plasma Formation in the Reflex Discharge on Gas-Metal Mixes // *Problems of Atomic Science and Technology. Series "Plasma Electronics and New Methods of Acceleration"*. 2013, № 4(86), p. 38-43.
8. I.I. Aksyonov, A.A. Andreev, V.A. Belous, et al. V*acuum arc. Plasma sources, coating deposition, surface modification*. K.: "Naukova dumka", 2012, p. 727 (in Russian).
9. D.R. Lide. *Handbook of Chemistry and Physics*. CRC, 2010.
10. V.N. Ochkin. *Spectroscopy of Low Temperature Plasma*. Weinhaim: "WILEY-VCH", 2009, p. 609.
11. J.E. Sansonetti, W.C. Martin. Handbook of Basic Atomic Spectroscopic Data // *Journal of Physical and Chemical Reference Data*. 2005, v. 34, № 4, p. 1559-2259.
12. A. Lofthus, P.H. Krupenie. The spectrum of molecular nitrogen // *Journal of Physical and Chemical Reference Data*. 1977, v. 6, № 1, p. 113-307.
13. P.H. Krupenie. The spectrum of molecular oxygen // *Journal of Physical and Chemical Reference Data*. 1972, v. 1, № 2, p. 423-534.
14. R.W.B. Pearse, A.G. Gaydon. *The Identification of Molecular Spectra Fourth editio*n. L.: "Chapman & Hall", 1976.
15. J.B. Hasted. *Physics of Atomic Collisions*. London: "Butterworths", 1964.
16. V.G. Grigorenko, L.A. Dushin, O.S. Pavlichenko, A.I. Skibenko. Anomalous discharge plasma decay with oscillating electrons in a strong magnetic field // *Zh. Tekh. Fiz.* 1966, v. 35, № 10, p. 1800-1807.
17. L.A. Dushin, A.I. Skibenko, A.V. Serikov, I.P. Fomin. Modulation of a propagating electromagnetic wave the reflex discharge plasma // *Ukrainian Journal of Physics*. 1974, v. 19, № 1, p. 98-104.
18. E.S. Frolov, V.E. Minaichev, A.T. Aleksandrov, et al // *Vakuumnaya Tekhnika*: Reference book M.: "Mashinostroyenie publ.", 1992, p 480 (in Russian).
19. E.S. Borovik, B.G. Lazarev, N.M. Tsin. Diffusion pump oil decomposition // *Ukrainian Journal of Physics*. 1957, v. 2, № 1, p. 78-86.
20. S.F. Grishin, V.Ya. Chernyshenko, V.B. Yuferov. About hydrogen recycling and gas desorption in the glow discharge // *Ukrainian Journal of Physics*. 1983, v. 28, № 9, p. 1365-1370.
21. A. Anders. *Cathodic Arcs. From Fractal Spots to Energetic Condensation*. Springer: "Science", 2008, p. 540.
22. I.V. Blonskii, A.A. Goncharov, A. V. Demchishin, et al. Plasma-dynamic and optical characteristics of magnetron-type cylindrical gas discharge under conditions of titanium nitride film synthesis // *Technical Physics*. 2009. v. 54, № 7, p. 1052-1057.
23. K. Brabaharan, J.A. Coxon, A.B. Yamashita. Vibrational analysis of the titanium nitride emission spectrum. The $A^2\Pi$-$X^2\Sigma$ and $B^2\Sigma$-$A'^2\Delta$ systems // *Spectrochimica Acta Part A: Molecular Spectroscopy*. 1985, v. 41, № 6, p. 847-850.

Article received 22.11.2016

ИССЛЕДОВАНИЕ ПАРАМЕТРОВ МНОГОКОМПОНЕНТНОЙ ГАЗОМЕТАЛЛИЧЕСКОЙ ПЛАЗМЫ, ОБРАЗОВАННОЙ В СМЕСИ МОЛЕКУЛЯРНЫХ ГАЗОВ

Ю.В. Ковтун, Е.В. Сюсько, Е.И. Скибенко, А.И. Скибенко, А.Н. Шаповал, В.Б. Юферов

Проведено исследование параметров плотной многокомпонентной газометаллической плазмы, образованной в среде рабочего вещества, состоящей из молекулярных газов, таких как азот и кислород, а также металлической компоненты *Ti*. Определены элементный и зарядовый составы образованной плазмы. Измерены параметрические и временные зависимости разрядного тока, средней плотности образованной плазмы в импульсном отражательном разряде.

ДОСЛІДЖЕННЯ ПАРАМЕТРІВ БАГАТОКОМПОНЕНТНОЇ ГАЗОМЕТАЛЕВОЇ ПЛАЗМИ, УТВОРЕНОЇ В СУМІШІ МОЛЕКУЛЯРНИХ ГАЗІВ

Ю.В. Ковтун, Є.В. Сюсько, Е.І. Скібенко, А.І. Скібенко, А.М. Шаповал, В.Б. Юферов

Проведено дослідження параметрів густої багатокомпонентної газометалевої плазми, утвореної в середовищі робочої речовини, що складається з молекулярних газів, таких як азот та кисень, а також металевої компоненти *Ti*. Визначено елементний і зарядовий склад утвореної плазми. Виміряні параметричні і часові залежності розрядного струму, середньої густини утвореної плазми в імпульсному відбивному розряді.