^{12.1} The study of characteristics of a surface discharge on the barium titanate

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The studies of characteristics of a surface discharge on the ferroelectric ceramics $BaTiO_3$ are presented. The emission spectrum of the discharge was studied using the optical emission spectroscopy method in the residual gas pressure range of $10^{-5}-10^{-3}$ Torr. The spectrum consisted of lines identified with neutral atoms and ions of elements (Ti, Ba, O) presented in the ferroelectric and residual atmosphere (N). The influence of the discharge current and the pressure of residual atmosphere on the intensity of spectrum lines have been determined.

Keywords: ferroelectric, ion source, emission spectroscopy, pressure.

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Plasma cathodes based on ferroelectric ceramics [1,2] may be used as efficient ion sources [3]. Plasma is formed as a result of discharge along the surface of a ferroelectric ceramic material under the influence of control voltage pulses. This discharge is established at triple points (metal-dielectric-vacuum contact) and is characterized by a low ignition voltage (units to tens of kilovolts) [4.5], which is attributable to a high electric field intensity. Owing to the presence of micropoints on the ceramic surface and the amplification of the tangential component of the electric field, the intensity at triple points reaches $10^4 - 10^5$ V/cm [6]. The discharge proceeds in an incomplete regime and has a multichannel structure [1], ensuring high uniformity of the plasma layer on the surface of a large-area cathode. Since the mentioned ceramics are low-erosion materials, cathodes may withstand as much as 10^6 pulses [6].

The formation of plasma is accompanied by a flux of neutral particles [7], which leads to an increase in pressure at the cathode surface. Depending on the parameters of control voltage pulses (amplitude, pulse duration, repetition rate), an increase in pressure may induce an enhancement of plasma density at the ceramic surface [8]. Therefore, when ferroelectric ceramics are used as an ion source, one needs to take into account the influence of residual atmosphere and the parameters of control voltage pulses on the formation of plasma. In the present study, the ionic and atomic spectral composition of plasma was examined as a function of electrophysical characteristics of the discharge and the residual atmosphere pressure.

A plasma source [9] based on barium titanate $(BaTiO_3)$ was constructed for experiments (Fig. 1, *a*). Our design differs from similar sources [3] in that the ferroelectric ceramic layer consists of several cylindrical segments. The diameter and thickness of each of them are 40 and 20 mm, respectively. This approach provides an opportunity to control the magnitude of discharge current as a function of the intrinsic capacitance of the plasma source and alter the surface area of ceramics. Individual segments were

secured to a flat dielectric base and connected by a common electrode (substrate). An external (front) electrode in the form of a stainless steel mesh with a cell size of 5×5 mm, a wire diameter of 0.5 mm, and an overall diameter of 120 mm was positioned on the opposite side of the ceramic layer. The mesh was fixed rigidly on the ceramic surface relative to the dielectric base (see Fig. 1, *a*) and was grounded (had zero potential). The total surface area of barium titanate under the mesh was 60 cm^2 .

The substrate voltage forms when the power supply capacitance is discharged into intrinsic capacitance C_c of the plasma source (Fig. 1, b). The actuation of gas discharger GD is associated with a certain delay t_d of the discharge current formation (Fig. 1, b). A relative substrate voltage variation of $\sim 2 \, \text{kV}$ is observed within an interval of $\sim 100 \, \text{ns}$ where the discharge current remains close to zero. A sharp increase in discharge current indicates the emergence of plasma on the ceramic surface. As plasma channels form and expand, capacitance C_c of the plasma source increases and reaches a virtually constant level of 25 nF within the voltage front (Fig. 1, b). The discharge voltage and current are oscillatory in nature, which is attributable to the mismatch of capacitances of the power supply and the plasma source and to the presence of inductors in the source circuit. The discharge current is limited in magnitude by the capacitance of the plasma source and reaches 900 A at a substrate voltage of $-10 \,\text{kV}$.

The spectral composition of plasma [10] was investigated within the following pressure range: $10^{-5}-10^{-3}$ Torr. An extremely low pressure in the chamber was established using a NVDM-250 diffusion steam-oil pump; the pressure was increased by supplying gas continuously from the atmosphere. The pressure was monitored in the region of the front electrode (Fig. 1, *a*) with a PMI-10-2 [11] sensor using a Meradat-VIT19IT2 vacuum gauge. The relative sensitivity coefficient of PMI-10-2 was taken equal to unity, since the gas contained components with both high and low ionization potentials. Optical spectra were



Figure 1. a — Diagram of the experimental setup. 1 — Substrate, 2 — dielectric base, 3 — barium titanate, 4 — front electrode, GD — controlled gas discharger, and C_c — intrinsic capacitance. b — Oscilloscope records of current and voltage. 1 — Substrate voltage, 2 — discharge current, 3 — cathode capacitance, and t_d — delay.



Figure 2. a — Pressure in a series of pulses (the substrate voltage is -10 kV); b — optical spectrum of plasma at a pressure of $1 \cdot 10^{-5}$ Torr.

recorded with an AvaSpec 3648 spectrometer within the 200–800 nm range. The signal integration time was 1 s. Individual lines of atoms and ions in the emission spectrum were identified by comparing them with data from the NIST Atomic Spectra Database (ver. 5.4) [12]. When lines with close centers associated with different components overlapped, the predominant element was chosen according to the tabular intensity values and probabilities of electron transitions between the upper and lower energy levels of atoms. The magnitude of instrumental line broadening was determined in preliminary calibration using a neodymium laser with a fundamental wavelength of 1064 nm tuned to the second and third harmonics (with the corresponding wavelengths of 532 and 355 nm) and a helium-neon laser with a wavelength of 632.84 nm.

The formation of plasma on the ferroelectric ceramic surface is accompanied by an intense flux of neutral particles, which is produced during evaporation of the ceramic material and desorption of gas from its surface [10] under the influence of the explosive electron emission current [1]. Neutral particles get ionized under the effect of an electron avalanche forming on the surface of ferroelectric ceramics in a tangential electric field. The flux of neutral particles raises the gas pressure at the front electrode surface. In a series of individual pulses with 5-s-long intervals between them, the pressure at a distance of 50 mm from the front electrode surface increases from $3 \cdot 10^{-4}$ to $2 \cdot 10^{-3}$ Torr (Fig. 2, *a*).

The typical plasma glow spectrum (Fig. 2, *b*) consists of lines of neutral atoms and ions of ferroelectric ceramic elements (Ti, Ba, O), nitrogen ions and atoms, and broad bands in the ranges of 410-440, 500-520, and 530-570 nm, which may be attributed to carbon [13], nitrogen, and oxygen [12,14–19] molecules. The emergence of lines of ions and atoms of nitrogen is associated with



Figure 3. a — Ratio of intensities of spectral lines of ions and atoms: Ti I (455.47 nm), Ti II (430.19 nm), Ti III (397.0 nm), Ba I (553.30 nm), Ba II (413.30 nm), and N II (422.77 nm); b — dependences of ion line intensities I_i on pressure: I — Ti II (325.43 nm), 2 — N II (422.77 nm), and 3 — Ba II (389.18 nm).

the ionization of gas adsorbed on the ceramic surface. According to [5], gas ionization occurs in a near-surface layer with a thickness of several monolayers. The lines of titanium and barium atoms and ions produce the dominant contribution to the spectrum. The spectral lines of oxygen ions (O II, O III) have lower intensities than the lines of titanium and barium ions. The intensity of lines of nitrogen atoms and ions is also lower. Broad molecular bands are very weak (Fig. 2, b), and their contribution to the emission spectrum is negligible. The low intensity of O II and O III ion lines may be attributed to the fact that their upper energy levels are positioned higher than those of barium and titanium; i.e., the energy of plasma electrons is insufficient to excite oxygen atoms efficiently.

The intensity of spectrum lines depends on the discharge current. As the current increases from 450 to 900 A due to the substrate voltage rise, the intensity of the lines of ions and atoms of Ti, Ba, and N increases, indicating the growth of plasma temperature and concentration. A relation between the intensities of ion I_i and atom I_0 lines in the spectrum in the form of power function $I_i \sim I_0^a$, where *a* is the exponent of power, was established (Fig. 3, *a*). The variation of intensities of Ti and Ba ion lines correlates with the change in intensity of N ion lines correlates with the change in intensities Ti and Ba ion lines.

The elemental composition of plasma remains unchanged within the pressure range of $10^{-5}-10^{-3}$ Torr [9]: the spectra are dominated by the lines of Ti, Ba, O, and N atoms and ions. As the pressure increases at a given substrate voltage (Fig. 1, *b*), a local change in the intensity of atomic and ionic lines occurs (Fig. 3, *b*), but no persistent growth or reduction of the intensity of spectral lines is observed. The intensities of Ti II, Ba II, and N II lines correlate with each other and vary amid the backdrop of a pulsed increase in pressure at the ceramic surface. The pressure affects

the frequency of collisions of electrons with molecules and atoms of matter at the ceramic surface. This affects the processes of excitation and relaxation of the energy levels of atoms and ions, leading, in turn, to a change in the intensity of spectral lines.

The emission spectrum of plasma of a surface discharge on ferroelectric ceramics BaTiO₃ consists of lines of atoms and ions of elements from the ceramic material and gas adsorbed on its surface. The formation of plasma is accompanied by a local increase in pressure at the ferroelectric ceramic surface. The spectral line intensities fluctuate amid the backdrop of a pulsed increase in pressure in the chamber varying within the range of $10^{-5}-10^{-3}$ Torr. The elemental composition of the spectrum remains stable within the examined range of variation of the discharge current and the residual atmosphere pressure. The composition of plasma spectral lines and their intensity are essentially unaffected by fluctuations of the residual atmosphere pressure within the interval of $10^{-5}-10^{-3}$ Torr.

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Conflict of interest

The authors declare that they have no conflict of interest.

References

- [1] G.A. Mesyats, Phys. Usp., **51** (1), 79 (2008). DOI: 10.1070/PU2008v051n01ABEH006426.
- [2] S.P. Bugaev, G.A. Mesyats, Sov. Phys. Tech. Phys., **12**, 1358 (1968).
- [3] K. Chirko, Ya.E. Krasik, J. Felsteiner, J. Appl. Phys., 91 (12), 9487 (2002). DOI: 10.1063/1.1479465

- [4] H. Riege, I. Boscolo, J. Handerek, U. Herleb, J. Appl. Phys., 84 (3), 1602 (1998). DOI: 10.1063/1.368230
- [5] O. Peleg, K. Chirko, V. Gurovich, J. Felsteiner, Ya.E. Krasik,
 V. Bernshtam, J. Appl. Phys., 97 (11), 113307 (2005).
 DOI: 10.1063/1.1927704
- [6] Y.E. Krasik, A. Dunaevsky, J. Felsteiner, A. Krokhmal, C. Leibovitch, A. Rosenberg, I. Schnitzer, J. Shiloh, IEEE Trans. Plasma Sci., 28 (5), 1642 (2000). DOI: 10.1109/27.901248
- [7] K. Chirko, Ya.E. Krasik, J. Felsteiner, A. Sternlieb, J. Appl. Phys., 92 (10), 5691 (2002). DOI: 10.1063/1.1516259
- [8] A. Dunaevsky, Ya.E. Krasik, J. Felsteiner, A. Sternlieb, J. Appl. Phys., 91 (3), 975 (2002). DOI: 10.1063/1.1425422
- [9] A.V. Stepanov, F.V. Konusov, S.K. Pavlov, V.A. Tarbokov, M.A. Serebrennikov, Nucl. Inst. Meth. Phys. Res. A, 1062, 169223 (2024). DOI: 10.1016/j.nima.2024.169223
- [10] S.P. Bugaev, V.V. Kremnev, Yu.I. Terent'ev, V.G. Shpak, Ya.Ya. Yurike, Sov. Phys. Tech. Phys., 16, 1547 (1972).
- [11] S.V. Korobkov, M.E. Gushchin, A.V. Strikovskiy, K.N. Loskutov, A.A. Evtushenko, Tech. Phys., 64 (1), 27 (2019). DOI: 10.1134/S1063784219010171.
- [12] U. Cvelbar, N. Krstulović, S. Milošević, M. Mozetič, Vacuum, 82 (2), 224 (2007). DOI: 10.1016/j.vacuum.2007.07.016
- [13] R. Engeln, B. Klarenaar, O. Guaitella, Plasma Sources Sci. Technol., 29 (6), 063001 (2020).
 DOI: 10.1088/1361-6595/ab6880
- [14] D. Wu, L. Sun, J. Liu, X. Yu, R. Hai, C. Feng,
 Z. Wang, H. Ding, Phys. Plasmas, 26 (1), 013303 (2019).
 DOI: 10.1063/1.5081969
- [15] V. Milosavljević, M. Donegan, P.J. Cullen, D.P. Dowling, J. Phys. Soc. Jpn., 83 (1), 014501 (2014).
 DOI: 10.7566/JPSJ.83.014501
- [16] S.M. Starikovskaia, K. Allegraud, O. Guaitella, A. Rousseau, J. Phys. D: Appl. Phys., 43 (12), 124007 (2010). DOI: 10.1088/0022-3727/43/12/124007
- [17] J.J. Camacho, M. Santos, L. Díaz, J.M.L. Poyato, J. Phys. D: Appl. Phys., 41 (21), 215206 (2008).
 DOI: 10.1088/0022-3727/41/21/215206
- [18] N. Bolouki, W.-H. Kuan, Y.-Y. Huang, J.-H. Hsieh, Appl. Sci., 11 (13), 6158 (2021). DOI: 10.3390/app11136158
- [19] Y.M. Kim, J.U. Kim, J.G. Han, Surf. Coat. Technol., 151-152, 227 (2002). DOI: 10.1016/S0257-8972(01)01601-2

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