Optical emission spectroscopy of tungsten cathodic arc plasma at different argon and nitrogen pressure

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The vacuum-arc discharge has numerous applications in both physics and applied technologies [1]. The vacuum-arc deposition method is commonly utilized in the industry to modify material surfaces and apply functional coatings. The optical spectrum of tungsten plasma was mainly investigated in the high vacuum region and also in various gases at pressures close to atmospheric pressure [2, 3]. Although the medium vacuum range is used for coating applications. Therefore, studies of the optical spectrum of tungsten cathodic arc at these pressures are of interest. The optical emission spectroscopy (OES) is a non-contact method that can be used over a wide range of temperatures, plasma densities, and neutral gas pressures. This is important in studying metallic plasmas both in vacuum and in neutral and reactive gas environments.

The optical emission spectrum of steady-state tungsten cathodic arc discharge plasma in an industrial commercial "Bulat-6" installation (see fig. 1) at different argon and nitrogen pressure was investigated [4]. A vacuum cathodic arc source with a magnetic stabilization field and tungsten cathode (diameter 60 mm) was used in the experiments. The induction of the magnetic field created by this coil was 6 mT. The arc current was I_d = 130 A. The gas pressure range in the experiments was from 0.05 Pa to 5 Pa. The vacuum chamber was pumped using a diffusion pump with a pumping speed of 2500 l/s. The pressure in the vacuum chamber was measured with ionization gauge PMI-10-2. The plasma lines spectrum was registered by optical emission spectrometer in the spectral range from 214 to 673 nm with the Type SL-40-2-3648 USB spectrometer (spectral resolution <0.6 nm) SOLAR TII. The shortest optical axis-cathode distance was about 250 mm. The spectral lines were identified according to the data from [5, 6].

In a vacuum arc discharge, the plasma is created by a cathode spot or spots, producing a plasma flow and a flow of neutral and macroparticles [1]. This is the main feature that differentiates a vacuum arc discharge from other types of discharge. The formed plasma fills the interelectrode space, where it interacts with the gas target. This interaction is especially intense when the concentration of neutral gas particles (gas pressure) increases. As a result of this interaction, the temperature of electrons, the charge of metal ions, and the ion energy
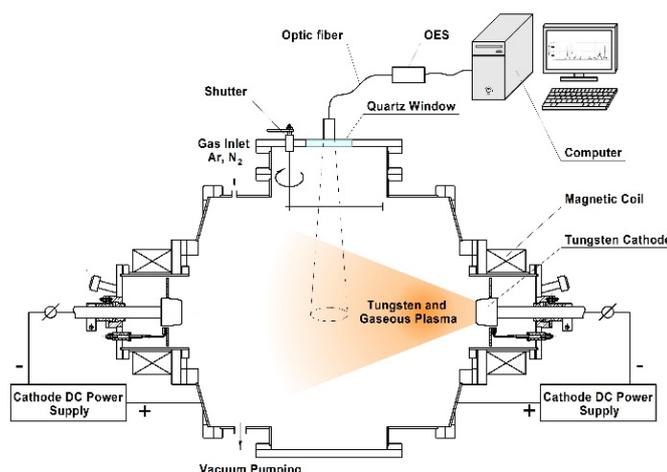
distribution function change.

Fig. 1. Schematic diagram of the "Bulat-6" cathodic-arc evaporation deposition equipment and optical spectrum measurements.

Fig. 2 (a) shows the optical spectrum of arc discharge plasma in the range of 214-673 nm for two argon pressure values. The spectrum observes lines of excited atoms and ions of the tungsten cathode material, for example, the W^* (W I) atom at 400.87 nm, 407.43 nm, 429.46 nm, and the single-charge ion W^{+*} (W II) at 265.8 nm, 276.42 nm, 239.71 nm. There are also lines of excited atoms and ions of the main argon gas Ar^* (Ar I) 415.85 nm, 420.06 nm, and Ar^{+*} (Ar II) 476.48 nm, 480.6 nm. The formation of excited argon atoms and ions is related to the processes of excitation and ionization by electron impact. The formation of argon ions is also possible in the processes of charge-exchange tungsten ions on argon atoms. The appearance of spectral lines of the hydrogen atom H^* (H I) in the spectrum is related to dissociative processes involving hydrocarbons (CH_y, C_xH_y) and water (H_2O) present in the residual atmosphere. Figure 2 shows that the intensity of spectral line emission increases with increasing argon pressure. Note that the intensity of spectral line emission depends on particle concentration, electron density, and electron temperature. An increase in argon concentration leads to an increase in the probability of excitation and ionization. This leads to an increase in plasma density, which is observed in vacuum arc discharges. However, this causes the electron temperature to decrease. An increase in argon concentration also leads to an increase in the probability of tungsten ions being charge-exchange on argon atoms and ions.

The optical spectrum of vacuum-arc discharge plasma in the range of 214–673 nm for two nitrogen pressure values is shown in Fig. 2 (b). The spectrum shows spectral lines of excited nitrogen molecules N_2^* Second Positive System 337.13 nm, 357.69 nm, 380.49 nm, and First positive system 590.6 nm, 595.901 nm, 606.966 nm, 632.285 nm.
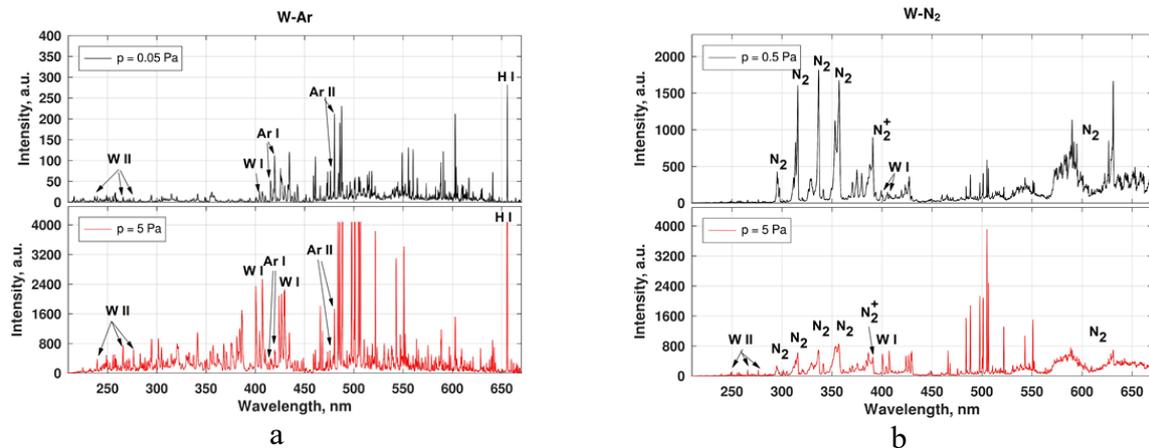
Fig. 2. Optical emission spectrum in the range of 214–673 nm for different argon pressures (a) and in the range of 214–673 nm for different nitrogen pressures (b).

Spectral lines of excited molecular ions N_2^{+*} First negative system 391.44 nm, 427.81 nm, 419.91 nm are also observed. The formation of excited nitrogen atoms and ions, as in the case of argon, is related to the processes of excitation and ionization by electron impact and the processes of charge-exchange tungsten ions on nitrogen molecules (atoms). Fig. 2 (b) shows that the intensity of the nitrogen spectral lines at 5 Pa is lower than at 0.5 Pa. A similar situation was seen with a vanadium vacuum-arc in nitrogen [4]. The spectrum in Fig. 2 (b) also shows the same lines of excited W^* (W I) atoms and W^{+*} (W II) ions of the tungsten cathode material as in the case of argon plasma (see Fig. 2 (a)). For comparison, Fig. 3 shows the normalized intensity of spectral lines for tungsten-argon plasma and tungsten-nitrogen plasma. Fig. 3 shows that the same W I lines are observed in both spectra.

The formation of gases ions may develop in charge-exchange processes involving tungsten ions and gases molecules (atoms). The depending on the defect of energy ΔE, equal to the difference between the binding energies of the electron in the target and the ion (atom) formed, separated into resonant (symmetric) $\Delta E = 0$ and quasi-resonant charge-exchange $\Delta E \approx 0$. In the case of $\Delta E > 0$, it is an exothermic reaction, and in the case of $\Delta E < 0$, it is an endothermic reaction. For an endothermic reaction, the charge-exchange cross section is 0 when $E_{c.m.} < \Delta E$. The estimation of ΔE in the case of charge exchange of W ions on Ar, N_2, N shows that for the W^{2+} ion and higher charges, $\Delta E > 0$ and, accordingly, the exothermic process of charge exchange is possible at any collision energies. At the same time, for W^+, the process $\Delta E < 0$.

In this case, it is important to know the values of the charge-exchange cross section. Unfortunately, there is no complete database on these processes for tungsten. For high-energy collisions of multiply charged W ions with Ar and N, the charge-exchange cross section was

calculated in [7]. In [8], the charge-exchange cross section for W^+ with Ar was measured at a collision energy of 10 keV, which was $3.9 \cdot 10^{-17}$ cm^2. Thus, charge-exchange processes can contribute to the formation of gas ions and, accordingly, to the observed optical spectrum.
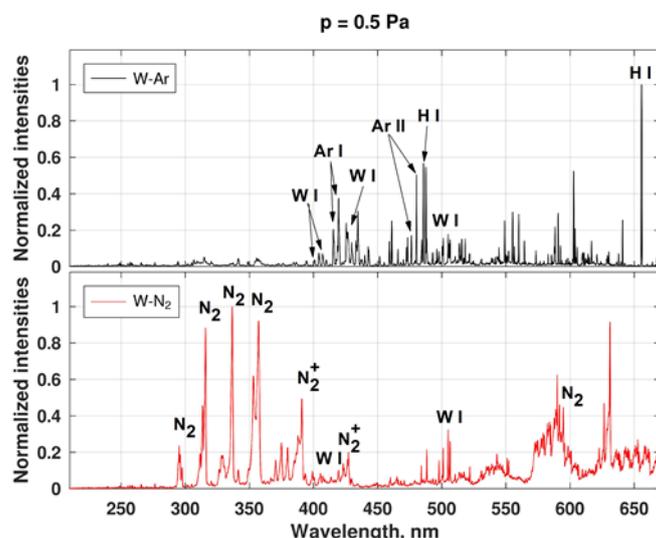

Fig. 3. Optical emission spectrum in the range of 214–673 nm for W cathodic arc in Ar and N_2 for pressure 0.5 Pa.

Optical emission spectroscopy studies of tungsten plasma in a cathodic arc discharge in a argon and nitrogen atmosphere have been carried out. Spectral lines of neutral atoms and ions of the cathode material W, W^+, argon Ar, Ar^+ and nitrogen N_2, N_2^+ were observed in the discharge plasma. The dependence of the intensities of the spectral lines on gaseous pressure differs greatly between argon and nitrogen. This difference can be attributed to the interaction between metal ions and neutral atoms (or molecules), as well as between gaseous ions.

References

[1]   A. Anders, Cathodic Arcs From Fractal Spots to Energetic Condensation (Springer Verlag2008), p. 540.

[2]   N. Sakura et al., J. Phys. D: Appl. Phys. 52, 404002 (2019).

[3]   S. Frohnert, J. Mentel, Contrib. Plasma Phys. 62, 202100216 (2022).

[4]   Yu. V. Kovtun et al., J. Appl. Phys. 134, 243301 (2023).

[5]   Kramida, A., Ralchenko, Yu., Reader, J., and NIST ASD Team (2024). NIST Atomic Spectra Database (ver. 5.12), [Online]. Available: https://physics.nist.gov/asd [2025, June 25]. National Institute of Standards and Technology, Gaithersburg, MD. DOI: https://doi.org/10.18434/T4W30F

[6]   A. Lofthus, P.H. Krupenie, J. Phys. Chem. Ref. Data 6, 113307 (1977).

[7]   Inga Yu Tolstikhina et al., J. Phys. B: At. Mol. Opt. Phys. 45, 145201 (2012).

[8]   M. Imai, Y. Iriki, A. Itoh, Fusion Science and Technology, 63, 392-399 (2013).