THE INFLUENCE OF NITROGEN ADMIXTURE ON CONCENTRATION OF AN ELECTRONIC–EXCITED HELIUM ATOMS IN ATMOSPHERIC PRESSURE GLOW DISCHARGE

V.I. Arkhipenko, A.A. Kirillov, L.V. Simonchik, S.M. Zgirouski

Institute of Molecular and Atomic Physics NASB, Minsk, Belarus, simon@imaph.bas-net.by

The concentrations of the low-excited helium atoms in states 2^1S, 2^1P, 2^3S and 2^3P were determined in the atmospheric pressure glow discharge in helium (99.98%He) and in helium with a nitrogen admixture (99.5%He + 0.5%N_2). It was shown that the adding of nitrogen into helium leads to the drastically reduction of concentration of both the low-excited helium atoms and ions.

PACS: 52.80.Hc

1. INTRODUCTION

There is large increasing interest in atmospheric pressure glow discharges (APGD) because they can be used for a wide range of technological applications without the need of vacuum systems. Nonthermal atmospheric plasmas of the APGDs are typically generated using voltage excitation at dc, at the 50 or 60Hz mains frequency, or at higher frequencies - from kilohertz to megahertz - between two electrodes of different configuration (plane—plane, pin—plane, microhollow cathode,..). Some of fundamental properties of APGD plasmas have been characterized experimentally including discharge dynamics, optical emission, and densities of the charged and excited particles.

As a rule, helium in mixture with other gases is used as working gas in discharges at atmospheric pressure [1, 2]. However, at present there isn't a complete understanding of the gas discharge physics in gas mixtures at atmospheric pressure. As it was shown in [2], for example, a small addition of other gases (~1%) into working gas of barrier discharge plays a significant role in stability of this discharge. On the base of numerical calculation it is approved in [3] that even the residual gas admixtures with concentration ~$0.5 \cdot 10^{-4}$ % exert a significant influence on both the plasma composition and the gas discharge parameters in atmospheric pressure discharge.

The self-sustained normal dc APGD in helium [4] is a convenient object for investigations of the kinetics of the weakly ionized nonequilibrium plasma at atmospheric pressure having a complicated composition. In present paper the concentrations of the low-excited helium atoms in states 2^1S, 2^1P, 2^3S and 2^3P were determined in the APGD in helium (99.98%He) and in helium with a nitrogen admixture (99.5%He + 0.5%N_2). The low-excited levels of helium are participated at many plasma chemical reactions. Therefore their concentrations are one of major parameters of nonequilibrium plasma of the APGD.

2. EXPERIMENTAL SETUP

The self-sustained normal dc APGD was created in the pressurized chamber between two electrodes: the weakly rounded tungsten anode (diameter 6 mm) and flat circular steel cathode (diameter 30 mm). Interelectrode gap was about 4 mm. The impurity concentrations in helium flow (H_2, N_2, O_2, Ar, CO_2, CO, Ne, H_2O) were not exceeding 0.02 %. The mixture of helium with admixture of nitrogen (in ratio 99.5%He: 0.5%N_2) was prepared in gas-cylinder in advance. Experiments were fulfilled at discharge current of 1 Ampere. An interelectrode voltage was ~ 200 V and ~240 V in APGD in helium and in mixture, correspondingly. The intensive water cooling of cathode was ensured due to its special design.

Schematic diagram of experimental apparatus for an absorption line profile registration is shown in fig. 1. The discharge image was focused on entrance slit of a double grating monochromator (MDD 500x2) of high resolution.

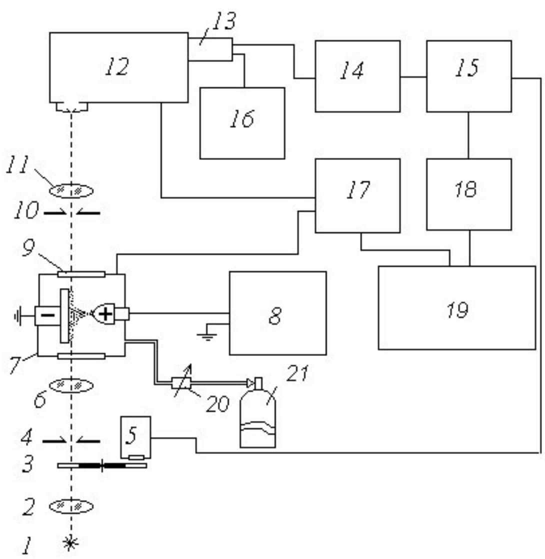

Fig.1. Schematic diagram of the experimental apparatus: 1 – probing emission source, 2, 6, 11 – lens, 3 –chopper, 4, 10 – slit-diaphragms, 5 -- synchronous sensor, 7– discharge chamber, 8 – discharge power supply, 9– windows, 12 – monochromator, 13 – photomultiplier, 14 – selective amplifier, 15 – synchronous detector, 16 – high-voltage power supply, 17 – control block, 18 – A/D converter, 19 – computer, 20 – flowmeter, 21 – gas-cylinder

20 – flowmeter, 21 – gas-cylinder.

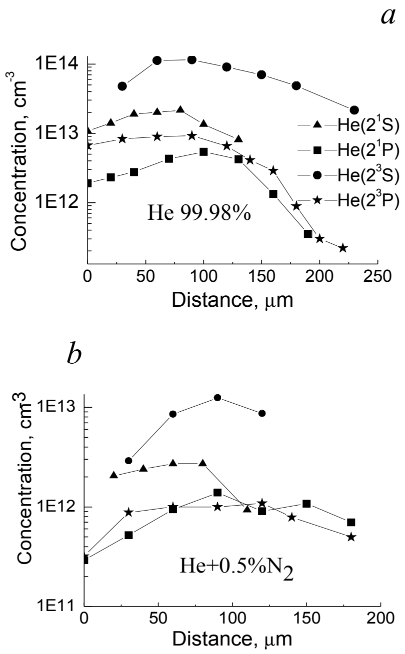

Fig. 3. Axial distributions of the exited helium atom concentrations in helium dischsrge (a) and in helium mixture discharge (b)

A Gaussian instrumental profile was ~ 0.01 nm. To determine the concentrations of the excited helium atoms the absorption spectroscopy method was used. The APGD was highlighted parallel to the cathode surface by a probing emission. A halogen incandescent lamp KGM-12-50 was used as probing light source for getting the absorption lines in visible spectral region. The original light source on base of arc with thermionic cathode [5] was used in the UV region. A probing light emission was focused in plasma volume located on the discharge axes and than was collected on entrance slit of monochromator together with a discharge emission. To obtain the needed resolution in axial direction two slit-diaphragms were used. The electrical signal proportionate to the probing emission intensity was separated from a common photomultiplier signal due to both the modulation of the probing emission and the using of a selective amplifier. To make better a signal/noise ratio the synchronous detection was used as well.

The spectra of the probing light emission before and after passing through plasma were registered in experiments. Calculation of the excited helium atom concentrations in states 2^1S, 2^1P, 2^3S and 2^3P was fulfilled by numerical integration of the measured absorption line profiles at the wavelength 501.6, 667.8, 388.9 and 587.6nm, correspondingly. An axial concentration distribution was obtained due to a moving of discharge chamber by step motor in the discharge axes direction.

3. EXPERIMENTAL RESULTS AND DISCUSSION

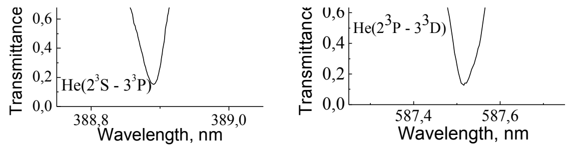

Fig. 2. The transmission line profiles at the wavelength 501.6 nm (a), 667.8 nm (b), 388.9 nm (c) u 587.6 nm (d), solid curve – discharge in helium, dashed one in mixture

The emission spectrum of the APGD plasma consists of the intensive lines of neutral helium atoms. The more weak lines of the hydrogen, nitrogen, oxygen and other elements are observed. The molecular bands are taken place in spectrum as well. The $N_2^+(B^2\Sigma_u^+ - X^2\Sigma_g^+)$ electron-vibration bands (1,0) 358.2 nm, (0,0) 391.4 nm, (0,1) 427.8 nm and (0,2) 470 nm of first negative system of nitrogen have higher intensities. The ($A^2\Sigma^+$-$X^2\Pi_i$) electron-vibration bands (0,0) 308.nm, (1,1) 314,3 nm of hydroxyl are intensive in UV region. More intensive spectra are registered in negative glow.

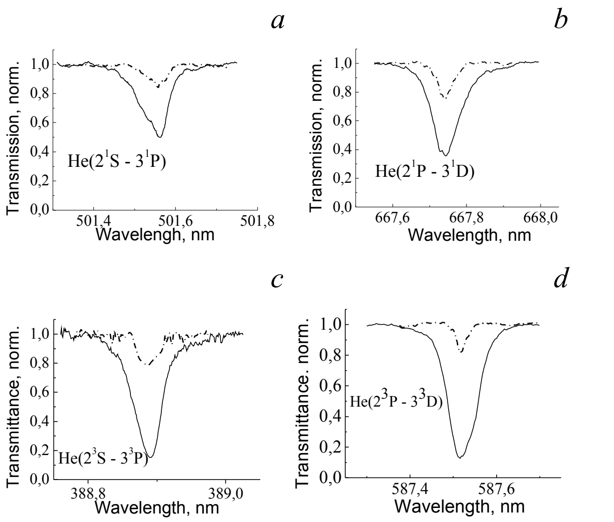

Fig. 2. The transmission line profiles at the wavelength 501.6 nm (a), 667.8 nm (b), 388.9 nm (c) u 587.6 nm (d), solid curve – discharge in helium, dashed one in mixture

The adding of nitrogen into working gas helium leads to an increase of intensities of the electron systems of molecular nitrogen. An interesting fact is that the intensity of the second positive system of nitrogen increases more quickly in comparison with one of the first negative system of nitrogen while concentration of nitrogen in mixture is increased. At the same time the bands of the second positive system of nitrogen are not observed in helium discharge. Therefore, a significant population of both the $N_2^+(B^2\Sigma^+)$ and the $N_2(C^3\Pi_u)$ levels takes place in the APGD in helium-nitrogen mixture.

The adding of nitrogen in helium influences on populations of the low-excited helium atom levels ($n = 2$) as well. The transmission line profiles at the wavelength 501.6, 667.8, 388.9 and 587.6 nm are presented in fig. 3. These profiles were registered in negative glow. It is seen at the adding of the 0.5% nitrogen in helium a significant decrease of an absorption is observed at all lines.

Using the experimental transmission profile (Fig. 2) the concentration of atoms N_n in corresponding state can be calculated as follow [5]

$$N_n \approx c/(0.026 f_{nm}\, l\lambda_0^2) \int \ln(I_0/I_\lambda)d\lambda, \quad , \qquad (1)$$

where λ_0 – wavelength of corresponding line, l – thickness of absorption layer which was supposed to be homogeneous, c – light velocity, f_{mn} – oscillator force.

The transmission profiles were registered at different distances from cathode. Using these experimental profiles and formula (1) the corresponding axial distributions of concentration of low-excited helium atoms were obtained. They are presented in fig. 3. It can be seen that the concentration of the excited helium atoms in helium-nitrogen mixture (99.5%He + 0.5%N_2) is less one order of magnitude than in case of pure helium. The axial concentration distributions are analogous for discharges in helium and helium-nitrogen mixture. Maximal concentration magnitude takes place in cathode region at distance about 0.1 mm from cathode surface.

Let we make the analysis of the main reactions in the both APGDs. The low-exited levels of helium atoms play an important role in stepped ionization processes. The role of this ionization mechanism in the APGD is more significant in comparison with one in the lower pressure glow discharge. In the electric fields $E/N_0 \leq 10^{-15}$V·cm^2 and at the ionization degree more 10^{-5} a stepped ionization in the APGD plasma is more important than a direct ionization by electron collision even at free exit of light emission [6]. Naturally, a light reabsorption increases the stepped ionization efficiency. The charged particles are produced in follow reactions with participation of excited helium atoms He^* [7]:

$$He^* + He^* \rightarrow He^+ + He + e \,, \qquad (2)$$

$$He^*(n=3) + He \rightarrow He_2^+ + e \,. \qquad (3)$$

The scheme of reactions in gas mixture discharge should be added by the follow reactions describing an interaction of helium atoms and molecules with the nitrogen molecules [7-9]

$$\mathrm{He}^{¿} + \mathrm{N}_2 \rightarrow \mathrm{He} + \mathrm{N}_2^+ + e \;, \qquad (4)$$

$$\mathrm{He}_2\left({}^3\Sigma_u^+\right) + \mathrm{N}_2 \rightarrow \mathrm{He} + \mathrm{He} + N_2^+ + e \;, \qquad (5)$$

$$\mathrm{He}^+ + \mathrm{N}_2 \rightarrow \mathrm{He} + \mathrm{N} + \mathrm{N}^+ + 0{,}3\ eV \;, \qquad (6)$$

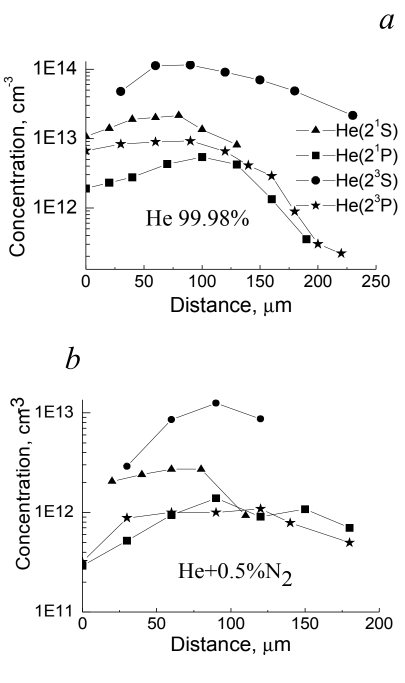

Fig. 3. Axial distributions of the exited helium atom concentrations in helium dischsrge (a) and in helium mixture discharge (b)

$$\mathrm{He}_{2^+} + \mathrm{N}_2 \rightarrow \mathrm{He} + \mathrm{He} + N_2^+ \quad . \tag{7}$$

Reaction (4) is in charge of a decrease of the excited helium atom concentrations at a nitrogen adding, because a decrease of the He^* atom concentration due to the temperature growth is neglible. Since a significant part (a few tens of percents) of created molecular ions of nitrogen is in the excited $B^2\Sigma^+$ state, the quenching of excited helium atoms is accompanied by emission in first negative system of nitrogen. The creation of molecular nitrogen ion result in a quenching process of the $He_2(2^3\Sigma_u^+)$ molecule at collision of metastable helium atom with nitrogen molecule (reaction 5). Reactions (6), (7) describe the processes of recharge at collision of both the atomic and molecular helium with nitrogen molecules.

Thus, the adding of nitrogen into helium leads to the reduction of concentration of both the low-excited helium atoms and ions. As result of that, the nitrogen becomes in charge of the maintenance of the APGD in helium-nitrogen mixture even in presence of small nitrogen admixture.

The work was supported by BRFBR (grant T04-181).

REFERENCES

1. S. Kanazawa, M. Kogoma, et. al. Stable glow plasma at atmospheric pressure// *J. Phys. D: Appl. Phys.* 1988, v.21, N5, p.838-840.
2. F. Massines, A. Rabehi, et. al. Experimental and theoretical study of a glow discharge at atmospheric pressure controlled by dielectric barrier// *J. Appl. Phys.* 1998, v.83, N6, p.2950-2957.
3. X. Yuan, L.L. Raja. Role of trace impurities in large-volume noble gas atmospheric-pressure glow discharges // *Appl. Phys. Lett.* 2002, v.81, N 5, p.814-816.
4. V. I. Arkhipenko, S. M. Zgirovski, L.V. Simonchik. Determination of the Metastable Helium Atoms Concentration in an Atmospheric-Pressure Glow Discharge // *J. Appl. Spectr.* 2000, v.67, N4, p.530-533.
5. V. I. Arkhipenko, S. M. Zgirovski, et. al. Light Source with Termionic Cathode// *J. Appl. Spectr.* (71), N1, p. 96-102.
6. A.A. Belevtsev, L.M. Biberman. Step ionization effect on first Townsend coefficient // *Izv. AN SSSR. Energetika i transport.* 1978, N4, p.106-113 (in Russian).
7. B.M. Smirnov. *Excited atoms.* Moscow: "Energoizdat", 1982 (in Russian).
8. F.W. Lee, C.B. Collins. Measurement of the rate coefficients for the de-excitation reactions of $\mathrm{He}\left({}^3\Sigma_u\right)$ metastables with Ne, Ar, N_2, CO, CO_2, and CH_4 // *J. Chem. Phys.* 1977, v.67, N6, p.2798-2802.
9. F.C. Fehsenfeld, A.L. Schmeltekopf et al. Thermal Energy Ion-Neutral Reaction Rates. I. Some Reactions of Helium Ions // *J. Chem. Phys*. 1966, v.44, N11, p.4087-4094.
1.

ВЛИЯНИЕ ДОБАВОК АЗОТА НА КОНЦЕНТРАЦИЮ ЭЛЕКТРОННО-ВОЗБУЖДЕННЫХ АТОМОВ ГЕЛИЯ В ТЛЕЮЩЕМ РАЗРЯДЕ АТМОСФЕРНОГО ДАВЛЕНИЯ

В.И. Архипенко, А.А. Кириллов, Л.В. Симончик, С.М. Згировский

Определены концентрации низких возбужденных атомов гелия в состояниях 2^1S, 2^1P, 2^3S и 2^3P в тлеющем разряде при атмосферном давлении в гелии (99.98%He) и в смеси гелия с азотом (99.5%He + 0.5%N_2). Показано, что добавление азота в гелий приводит к существенному уменьшению как заселенностей нижних возбужденных уровней гелия, так и концентрации ионов гелия.

ВПЛИВ ДОБАВОК АЗОТУ НА КОНЦЕНТРАЦІЮ ЭЛЕКТРОННО-ЗБУЖДЕНИХ АТОМІВ ГЕЛІЮ В ТЛІЮЧОМУ РОЗРЯДІ АТМОСФЕРНОГО ТИСКУ

В.І. Архипенко, А.А. Кирилов, Л.В. Симончик, С.М. Згировський

Визначено концентрації низьких збуджених атомів гелію в станах 2^1S, 2^1P, 2^3S і 2^3P у тліючому розряді при атмосферному тиску в гелії (99.98%He) і в суміші гелію з азотом (99.5%He + 0.5%N_2). Показано, що додавання азоту в гелій приводить до істотного зменшення як заселенностей нижніх збуджених рівнів гелію, так і концентрації іонів гелію.