Study of spatial distributions of highly ionized nonequilibrium helium plasma at atmospheric pressures

V F Chinnov¹, D I Kavyrshin¹, A G Ageev², O V Korshunov¹, M A Sargsyan¹ and A V Efimov¹

¹ Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg 2, Moscow 125412, Russia
² National Research University Moscow Power Engineering Institute, Krasnokazarmennaya 14, Moscow 111250, Russia
E-mail: dimakav@rambler.ru

Abstract. Experimental study of helium plasma in the state of quasistationary heating under atmospheric pressure was made. The plasma state is shown to fail to be described by Saha–Boltzmann approximation at high ionization levels \( \alpha_i = 0.5–0.9 \), temperatures 2.5–4.0 eV and electron concentrations about \( 10^{17} \) cm\(^{-3}\). The deviation from the equilibrium state of the plasma is caused by lack of spatial uniformity due to charged particles loss by ambipolar diffusion. In order to thoroughly study the temporal changes of plasma radiation characteristics, spectroscopic analysis was carried out with DFS-452 spectrometer and high-speed CMOS camera Andor iStar attached to its output. The system yields the spatial resolution of 30–50 \( \mu \)m and temporal resolution of 5–50 \( \mu \)s. Electron concentration \( n_e \) was measured from the half-width of the local He\(_I\) spectrum line contours having dominant quadruple Stark effect with well-known constants. In order to determine the temperature of heavy particles, Doppler component of He\(_I\) line triplet at 1083 nm was studied. The temporal evolution of the following important characteristics has been determined for helium plasma during pulsed heating: current power, intensities of a number of He\(_I\) and He\(_{II}\) spectral lines, electron temperatures and concentrations.

1. Introduction
During the recent years, experimental works showing substantial deviations of highly ionized plasma state from the local thermodynamic equilibrium state have been published [1–4]. It is of interest to investigate the possibility of achieving double ionization of single-ionized helium plasma by means of quasistationary impulse heating. In order to study the reasons behind nonequilibrium of highly-ionized helium plasma, experimental investigation of spatial (radial) distributions of plasma parameters is needed since spatial inhomogeneity and large values of transport coefficients—thermal conductivity, ambipolar diffusion—are the probable causes of the observed nonequilibrium [2].

2. Experimental setup and method for obtaining emission spectra
Highly ionized helium plasma was generated on a DC electric arc plasmatron with expanding anode channel, \( d = 4–8 \) mm, and vortex arc stabilization [5], helium flow 0.1–0.3 g/s, arc length
10 mm and arc current from 200 to 400 A providing plasma temperature from 2 to 2.5 eV. The plasma was heated further with electric impulses with the same polarity as the main arc current, duration of 1000–1500 µs and peak current up to 4000 A. Plasma was heated in this quasistationary manner to temperatures up to 4 eV. Electrical schematics of the combined arc generation circuit are shown in figure 1.

The investigated zone of the arc was located 1 mm from the tip of the tungsten cathode, its radiation was observed through two symmetrically located quartz looking windows in the plasmatron nozzle wall. The emission spectra were registered with an optic fiber three-channel spectrometer AvaSpec and DFS-452 spectrograph with a high-speed camera Andor attached to its output. The AvaSpec spectrometer is able to register emission spectra in a wide wavelength range of 200–1100 nm allowing to get low-resolution overview spectra. The input slit of the DFS-452 spectrometer was set to be narrow, about 20–25 µm with the attached camera pixel size being 25 µm. Thus, the instrument function of the DFS-Andor was $\delta_{\text{inst}} \approx 0.03$ nm allowing to resolve the contours of helium spectral lines with widths of about 0.1 nm. A selection of most information-bearing spectrum segments obtained with the systems described above is given in figures 2 and 3. Figure 3 shows plasma spectrum with spatial resolution over plasma radius which allows us to determine radial distributions of plasma parameters.

A characteristic feature of the spectrum in the 440–480 nm area is that double increase in arc electric current strength (from 200 to 400 A) causes intensity increase of He I lines and continuum only by 10–20% and sharp four-fold increase in He II line intensity at 468.5 nm.

3. Study methods and results of the stationary discharge

3.1. Measurement system calibration

The measurement system was calibrated with reference radiation source—tungsten strip lamp in wavelength range of 300–1100 nm and deuterium lamp in wavelength range of 190–350 nm. Measuring device sensitivity dependencies on wavelength obtained from calibration were used to convert the relative spectral radiation intensities produced by the devices into absolute spectral radiation intensities of the plasma. Absolute population values for a number of excited states for a set of arc currents were obtained from the measured emission spectra of stationary and nonstationary plasma. The tasks performed on the obtained spectra were:

- Matching experimentally obtained and reference data spectral lines;
- Determining relative and absolute radiation intensity of the spectral lines;
- Analysis of their contours and their means of broadening.

In order to determine the amount of spectral line widening due to the instrument function of the AvaSpec spectrometer, it was used to register emission of tantalum electric discharge lamp with hollow cathode. Widths of the least wide lines registered were determined, these widths

![Figure 1. Electrical schematics of the experiment.](image)
Figure 2. Sample spectra regions obtained with AvaSpec (upper) and DFS-452 (lower) systems from chordal observation in 1 mm from cathode for various electric current strengths.

being instrument function. It was found to be 0.15, 0.2, 0.3 nm for the first (UV range), second (400–620 nm) and third (600–1000 nm) channels respectively.

The DFS-452 spectrometer combined with Andor camera allows to resolve spectral lines with widths down to 0.04 nm.
Figure 3. Sample region of 2D-spectrum obtained with DFS-Andor iStar system in 1 mm from cathode, spectrum (a) being for arc current strength of 200 A and (b) for 400 A.

In order to perform absolute calibration of the measurement devices, a comparison between the obtained spectrum and the reference data on their radiation was carried out.

A small ($d = 3$ mm) tungsten incandescent lamp was used to determine the influence of looking window geometry. In order to obtain the passing radiation reduction rate by the looking window, the lamp was placed inside the plasmatron channel to have its emission spectrum registered. After that, its emission spectrum was registered with the same optical system but with the lamp placed outside the plasmatron. The relation between these signals gives the signal weakening coefficient for the plasmatron looking window. Its value was found to be 0.35.

An additional means of the obtained device spectral sensitivity curve validation was comparing the obtained spectra at the wavelengths where they were determined with more than one device.

The performed calibration procedures allow obtaining absolute intensities of He\textsc{i} and He\textsc{ii} spectral lines from their relative spectral intensities.

3.2. Spectroscopy data processing: inverse Abel transformation and radial distributions of spectral line intensities

By utilizing matrix camera, we have studied spatial and time distributions of radiation properties of helium plasma which has allowed us to obtain both stationary distributions of temperature and electron concentration and their evolution during impulse plasma heating.

The data obtained from the DFS-Andor system was processed with a developed program that applied inverse Abel transformation to the data and produced radial distributions of radiation intensity. A single camera shot captures wavelength range of 40 nm containing several He\textsc{i} and He\textsc{ii} lines (see figure 3). It was processed in the following order:

- The data from the camera was supplied in ASCII files in table format with each line corresponding to one radius (that is, distance of observation chordal line from the axis) and each column corresponding to one wavelength, each element giving the current spectral radiation intensity.
- The exact values of radius and wavelength for each table element were obtained through the scale coefficients.
- One of the lines to be processed was manually selected on the image and was split into slices over the radius with fixed wavelength.
• Every slice was processed with inverse Abel transformation converting chordal radiation intensity values into radial ones. Since Abel transformation accepts only one line slope at a time, if the line is symmetrical, only one transformation is sufficient.

• The obtained radial slices were united back into table with each line corresponding to different distances from the arc axis, each column corresponding to different wavelength and values showing radiation intensities. It is then possible to obtain slices over wavelengths for each radius from that table containing radial distribution information: electron temperature and concentration, excited particle concentrations.

The obtained spectral line contours are then processed with the algorithm developed earlier [6] which obtains the aforementioned parameters in the following way:

• The lines are matched with the lines from the database [7];

• Line contour is approximated with Voight function [8, 9] carrying the information within its Gaussian and Lorenzian components: the Gaussian component width is formed by instrumental function and Doppler shifting and the Lorenzian width forming due to Stark effect.

• Line intensity is found by integrating the found Voight function over all wavelengths.

3.3. Electron concentration

Electron concentration $n_e$ was measured from the width of He$_I$ spectral lines with dominating quadruple Stark effect and well-known constants for it (lines at 318.7, 388.8, 402.6, 471.3, 492.2, 501.5, 667.8, 706.5, 728.1 nm and others). The constants were taken from [10,11]. As arc current increases from 200 A to 400 A, electron concentration increases from $6.5 \times 10^{16}$ to $8.0 \times 10^{16}$ cm$^{-3}$, what gives a rather small plasma ionization temperature increase: $T_i = 20000–21000$ K.

The presence of a large number of He$_I$ lines and application of spectrometers with high spectral resolution (better than 0.05 nm) allows to perform multiple and plasma state-independent measurements of electron temperature with error no more than 10% at electron concentrations higher than $10^{16}$ cm$^{-3}$. It is possible to use helium ion lines with linear Stark effect to measure temperature concentration at temperatures higher than 25000 K.

Figure 4 shows an example of $n_e$ radial distributions obtained from analysis of these lines. Differences between results for individual lines did not go over 20%. It can be seen from figure 4 that electron concentration is not much sensitive to arc electric current increase which is being one of the key problems of obtaining high ionization degrees of helium plasma. Electron concentration decreases by one magnitude in the outer arc region as compared to arc center, this decrease being less strong for arc current of 400 A.

Important information is contained within the values of local spectral line intensities. Intensity of an optically thin line gives the population of the corresponding emitting excited energy level:

$$I_{k_i}(r) = \int I_{\lambda} d\lambda = h\nu_0 n_{k_i}(r) A_{ki}.$$ (1)

Thus, spatial and dimensional distributions of $I_{k_i}(r,t)$ allow to make assumptions on the populations of the corresponding emitting excited energy level. Radial distributions of a large number of He$_I$ line intensities were obtained for the stationary discharge in the excitation energy range of 21–24 eV. Radial dependency examples of relative intensities of He$_I$ 471.3 nm line and He$_II$ 468.5 nm line for comparison are given in figure 5.

The following important features require attention: ion line intensity raises with current increase while that of atomic line decreases and atomic line intensity reaches its maximum not in the middle of the plasma arc at 400 A current but in the distance of $r \approx 0.5$ mm from it.

Data sets on absolute and relative population of He$_I$ excited states allow to tell whether these populations follow equilibrium (Boltzmann) distribution law. The overall picture of the
Figure 4. Radial distributions of electron concentration $n_e$.

Figure 5. Radial distributions of relative intensities for spectral lines He\textsc{i} 471.3 nm and He\textsc{ii} 468.5 nm for 200 A and 400 A arc currents.

populations of the emitting helium atom energy states for the currents 200 and 400 A is shown in figure 6.
3.4. Electron and heavy particle temperatures

Nonequilibrium He\textsubscript{I} excited level population was identified from figure 6 data analysis. It can’t be described by “Boltzmann exponent” method [4]. This problem may be solved by using an alternative method for \( T_e \) determination, which is based on measuring the relation between atomic and ion line intensities. Its advantage lies in large energy gap between the considered excited states (over 50 eV) which decreases the influence of nonequilibrium on the final result. The values of electron temperature in the axial region of the stationary plasma arc in helium determined with this method are 2.6 and 2.9 eV for arc currents 200 and 400 A respectively.

An additional method of determining electron temperature is the analysis of the relationship between continuum, atomic and ionic lines intensities changes during the growth of the heating current. In the high ionization region (\( T_e > 1.5 \) eV and \( n_e > 10^{16} \) cm\(^{-3}\)) continuous plasma radiation in the observable wavelength range includes radiation produced by the process of photorecombination onto excited states of He\textsubscript{I} and He\textsubscript{II} and electron bremsstrahlung in the fields of He\textsuperscript{+} and He\textsuperscript{++} ions. According to [12], dependence of continuum radiation intensity on the plasma parameters is as follows:

\[
\epsilon_{\lambda}^{\text{fb,eff}} \approx n_e([He^+] + 4[He^{++}]) \sqrt{T_e}. \tag{2}
\]

Assuming equilibrium composition of helium plasma, it is easy to establish the form of \( \epsilon_{\lambda}^{\text{fb,eff}}(T_e) \) dependence: it is a sharply increasing function in the temperature region of 2.5–4.5 eV reflecting the increasing role of He\textsuperscript{++} ions in continuum radiation as plasma temperature increases.

In order to determine heavy particle temperature, a triplet line He\textsubscript{I} at 1083 nm (transition \( 2\textsuperscript{3}P \rightarrow 2\textsuperscript{3}S \)) should be used. This line allows to expand the range of He\textsubscript{I} excitation energies since the \( 2\textsuperscript{3}P \) state has the lowest excitation energy of \( E^* = 20.96 \) eV of all observable He\textsubscript{I} lines and

![Figure 6. Experimental data on the population of He\textsubscript{I} excited states. The numbers indicate spectrum line wavelengths (nm) corresponding to the respective population value logarithm.](image)
also to determine the Doppler component of the line contour due to high value of its half-width. The values of $T_a$ for arc currents 200 and 400 A were found to be 1.8 and 2.2 eV respectively which indicates the presence of a gap between electron and atom temperatures.

4. Impulse heating

In order to carry out quasistationary heating of electric arc plasma, the duration of the heating impulse $\tau_{imp}$ must be larger than the characteristic relaxation times of electron temperature, energy exchange between electrons and ions and the larger time of two: either electron and ion triple recombination or ambipolar diffusion. $\tau_{imp} \geq 1000 \mu s$ satisfies these conditions.

When studying the combined discharge plasma with impulse heating, we used instruments with good spatial (30–50 $\mu m$) and temporal (5–50 $\mu s$) resolutions to investigate temporal changes of emission parameters of helium plasma, namely, DFS-452 spectrometer with high-performance CCD Andor iStar camera attached to its output. Examples of the obtained helium plasma 2D spectra at different times during impulse heating are given in figures 7a and 7b. Axial shift of ion line maximum relative to atomic lines is observed testifying the existence of spatial transfer processes in radial direction.

The relative intensity method applied to atomic and ion lines allow analyzing the growth of electron temperature during impulse heating of the stationary arc plasma. By applying inverse Abel transformation to the closely located lines He\textsubscript{II} 468.5 nm and He\textsubscript{I} 471.3 nm, it is possible to observe the evolution of intensities of these lines during the impulse. Electron temperature rises from 2.9 eV at the beginning of the impulse to the maximum value of $T_e^{max} \approx 3.7$ eV at $\tau = 800 \mu s$.

Temporal dependence of the experimentally obtained values of impulse current, spectral lines intensities and electron temperature during the impulse heating is given in figure 8. The dependence of electron temperature on experiment time is obtained by applying the relative intensities method to the He\textsubscript{II} 468.5 nm line and the He\textsubscript{I} 492.2 nm line with almost constant radiation intensity. Radiation intensity measurement error does not exceed 10% and
Figure 8. Discharge and plasma parameter changes during impulse heating of stationary arc plasma with the current of 200 A.

$T_e$ determination error is 15–20%. It should be noted that the maximum intensity of HeII 468.5 nm line and the maximum value of $T_e$ are lagging behind the impulse current maximum by 300−400 µs which is equal to the relaxation time of the atomic and ion excited level population changes caused by changes in electric current (energy deposition). The intensity of the atomic line HeI 471.3 nm remains within 15% of its initial value.

Attempts to further heat plasma by increasing the impulse current to 4–5 kA caused intense thermoelectron emission from the cathode and sharp increase in evaporation of materials: tungsten from the cathode and copper from its cooled holder. The influx of easily ionized metal particles causes electron temperature to fall in the observed arc region near the cathode.

5. Conclusion

The most important results of the performed investigation are the following. An experimental study of state of atmospheric pressure helium plasma during stationary and quasistationary heating by electric field with intensity of 30–40 V/cm and specific energy disposition of 100–1000 W/cm³ has been performed. Radial distributions of electron concentrations and intensities of atomic and ion lines under the conditions of high ionization degree $\alpha_i = 0.5$–0.9 of helium plasma with $T_e = 2.5$–3.0 eV $n_e \lesssim 10^{17}$ cm$^{-3}$ have been obtained. Strong spatial inhomogeneity of the plasma was observed manifesting itself as fivefold decrease of electron concentration and ion line intensity at the distance of only 1 mm from the arc axis. This fact confirms the presence of strong diffusion flow of charged particles on the channel walls causing deviations from the local thermodynamic equilibrium state of the plasma. During impulse heating of electrons in the stationary arc discharge, electron temperature increased only by 0.5 eV which is showing the difficulty in obtaining the double-ionized helium plasma under quasistationary conditions.

References

[1] Jonkers J and Van Der Mullen J A M 1999 J. Quant. Spectrosc. Radiat. Transfer 61 703–709
[2] Jonkers J, van de Sande M, Sola A, Gamero A and Van Der Mullen J A M 2003 Plasma Sources Sci. Technol. 12 30–38
[3] Sturgeon R E, Willie S N and Luong V T 1991 Spectrochim. Acta, Part B 46 1021–1031
[4] Isakaev E K, Chinnov V F, Sargsyan M A and Kavyrshin D I 2013 High Temp. 51 141–146
[5] Isakaev E K, Sinkevich O A, Tyuftyaev A S and Chinnov V F 2010 High Temp. 48 97–125
[6] Kavyrshin D I, Chinnov V F and Ageev A G 2015 J. Phys.: Conf. Ser. 653 012115
[7] Kramida A, Ralchenko Yu, Reader J and NIST ASD Team 2015 NIST Atomic Spectra Database (ver. 5.1), [Online]. Available: http://physics.nist.gov/asd [2014, June 9]. National Institute of Standards and Technology, Gaithersburg, MD.
[8] Lochte-Holtgreven W and Ed 1968 Plasma Diagnostics (Amsterdam: Elsevier)
[9] Frish S E 1963 Optical Spectra of Atoms (Moscow-Leningrad: Fizmatgiz)
[10] Griem H 1974 Spectral Line Broadening by Plasmas (New York: Academic)
[11] Konjevic N, Dimitrijevic M S and Wiese W L 1986 J. Phys. Chem. Ref. Data 13 619
[12] Biberman L M, Vorob'ev V S and Yakubov I T 1987 Kinetics of Nonequilibrium Low-Temperature Plasmas (Berlin: Springer-Verlag)