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THE MEASUREMENT OF PULSED GAS DISCHARGE PARAMETERS BY MEANS OF Fe I LINES IN ARGON AND ARGON-HYDROGEN MIXTURE

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Abstract. In this study, the optical emission spectroscopy (OES) is used to measure the pulsed atmospheric pressure gas discharge parameters in argon and argon-hydrogen mixture. The discharge source has a needle-to-cylinder configuration (NTC). When the stainless steel cathode (SS) is employed, a numerous metallic lines are identified in the discharge spectra (Cu, Zn, Fe, Cr, V, W) in the 260-800 nm spectral range. The thorough study of electron temperature T_e by means of Fe I lines intensity distribution is done. The addition of hydrogen (3% by Vol.) changed the yield of metallic lines. The results showed that T_e=7000 K-9000 K, which was also confirmed using an independent line ratio method. Several, multicomponent, broad Fe I lines are recorded in the scope of this study. The Stark shift of the broadest Fe I line component is used to estimate electron number density N_e in NTC discharge. The N_e results will be compared with Fe I line deconvolution results.

1. INTRODUCTION

The versatile continuous, pulsed and RF gas discharges, in which the excitation/ionization of metal atoms occurs, are used as analytical plasma sources and the environment for plasma-surface interaction. The metallic spectral lines are present also in OES spectra as a result of liquid samples treatment in case of solution-cathode-glow-discharge (SCGD) and in case of low pressure GD source (e.g. Grimm-type cell) for the depth profiling of conductive thin layered surfaces, see reference list in (Jovović 2023).

An atmospheric pressure needle-to-cylinder (NTC) discharge source driven by μs-pulsed generator unit has been studied by means of OES in our Lab, see e.g. (Jovović and Majstorović 2022, 2024). The NTC discharge structure comprises near-cathode region, negative glow (plasma column) and anode region. Decreasing the gap between metallic cathode and graphite anode results in formation of intensive ionizing near-cathode zone, serving as a stable, reproducible source of excited and ionized metallic particles. In that case, one may use the single-frame-spectra approach on numerous Fe I lines to measure T_e applying the atomic state distribution function (ASDF) which reflects the dependence of the population of atoms and ions on their perspective internal states energy. This means that for each fixed position of the diffraction grating, the spectrum is taken by CCD camera covering 18 nm range in the first diffraction order. In addition, the Fe I lines recorded at 538.3 nm, 539.3 nm and 542.4 nm, recorded in the second diffraction order, has asymmetric line shape indicating the existence of low- N_e and high- N_e NTC discharge regions. From the shift of broadest Fe I line component, the highest N_e is estimated.

2. EXPERIMENTAL

The schematic drawing of NTC source and experimental setup is shown elsewhere, see e.g. (Jovović and Majstorović 2024). The source consists of needle-type cathode (diameter 4 mm) and a cylindrical graphite anode (diameter 25 mm). In the present study, the cathode is made of stainless steel (2.07 % V, 3.97 % Cr, 14.84 % Mo, 13.84 % W and Fe balance). The pure argon gas (Ar 99.999 %) and argon-hydrogen gas mixture (Ar +3 % H_2 by vol.) are employed. The discharge is driven by pulsed voltage power supply that consist of DC power supply (Kepco, 0-2 kV, 0-100 mA), the bank of capacitors, HV switching unit (MOSFET technology) and rectangular pulses generator (2-999 μs pulse width, 0.1-100 % duty cycle). During the pulse, the voltage jumps in the range 900-1300 V for 16.7 % duty cycle. The radiation from NTC source was recorded by means of 2 m Ebert type spectrometer (Carl Zeiss, O.P. f/28, an inverse linear dispersion of 0.74 nm/mm) equipped with a thermoelectrically cooled, back-thinned Hamamatsu CCD camera ($T = -10$ °C, 2048 × 512 pixels, 12 μm pixel size).

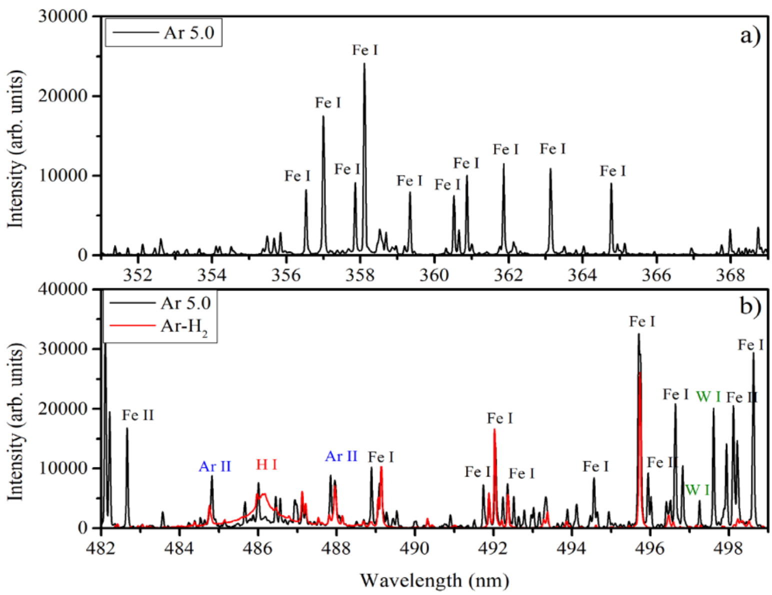

Figure 1: a) The spectrum of Fe I lines in the range 351-368 nm emitted from argon NTC discharge and b) the comparison between NTC spectra in argon and argon-hydrogen mixture in the range 482-499 nm. Experimental conditions: pulse width of 50 μs, duty cycle of 16.7 %.

3. RESULTS AND DISSCUSSION

The example of SS NTC discharge spectrum in the range 351-368 nm is given in Fig. 1a while the comparison between NTC spectra in argon and argon-hydrogen mixture in the range 482-499 nm is shown in Figure 1b. The hydrogen admixture selectively affects the absolute and relative intensities of Fe I lines, which has the impact on the accuracy of analytical results. The results of T_e measurement in both working gases versus the central wavelength of studied CCD spectral range are given in Figure 2. The advantage of single-frame-spectrum approach for T_e measurements is that there is no need for the spectral sensitivity curve. One may notice the influence of hydrogen admixture on distribution and density of points in Figure 2. The T_e values shown in Figure 2 are in agreement with T_e obtained using line ratio method (Jovović 2023).

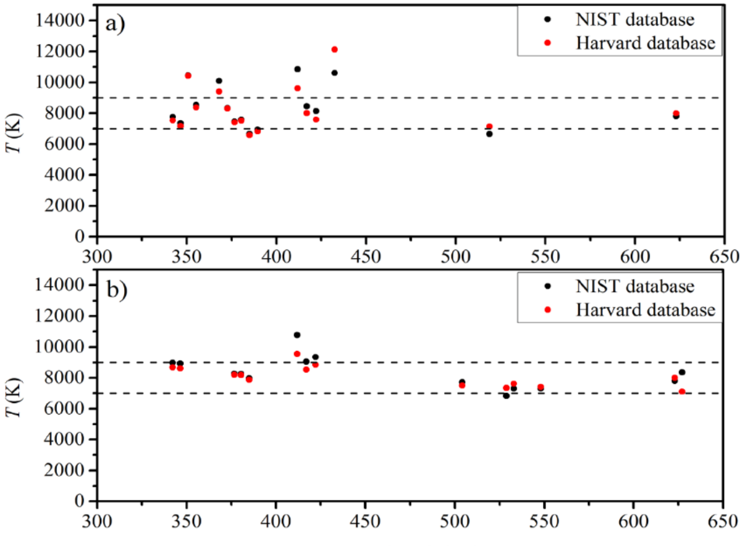

Figure 2: The T_e (K) measured from a single-frame spectrum versus the central wavelength of the studied spectral range in a) Ar and b) Ar+3% H_2 for two A_{ik} databases.

The blue-shifted wing of Fe I 538.3 nm and Fe I 542.4 nm line and the red-shifted wing of Fe I 539.3 nm line, see Fig. 3, indicate the presence of high density plasma (the shifted broadest fit component). Since we work at atmospheric pressure one needs to take into account Van der Waals shift as well. The comparison between experimental shifts measured in (Konjević et al 2002) (N_e=1×10^{16} cm^{-3}; T_e=6000 K) for the studied Fe I lines and Stark shifts determined in our work, is given in Table 1. From Table 1, one may notice that d_e values are one order of magnitude higher than d_m which gives N_e≈ 1.2×10^{17} cm^{-3} (Fe I 538.3 nm), N_e≈ 5×10^{16} cm^{-3} (Fe I 539.3 nm) and N_e≈ 8.9×10^{16} cm^{-3} (Fe I 542.4 nm). Although a rough estimate, these values show a reasonable agreement with the fitting results of Fe I lines in Figure 3, which will be shown during Poster session.

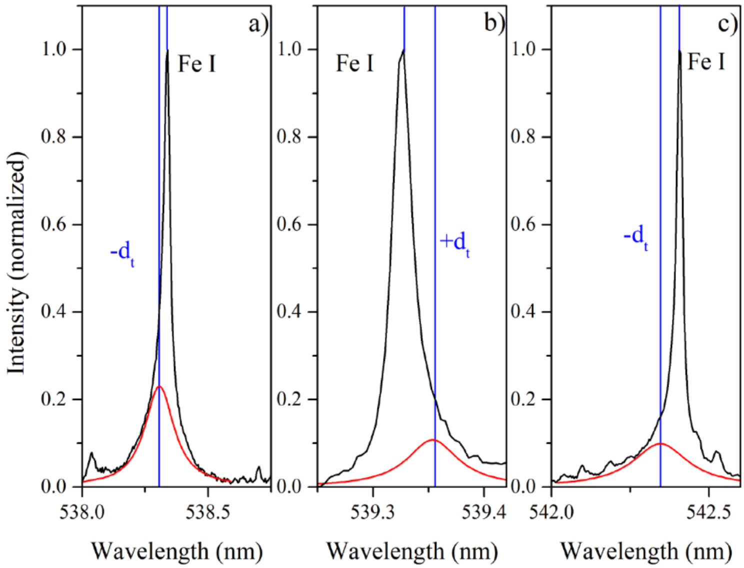

Figure 3: The normalized spectra and a broad fit component of a) Fe I 538.3 nm line, b) Fe I 539.3 nm line and c) Fe I 542.4 nm line recorded from argon and argon-hydrogen NTC discharge.

Line	Transition	d_m (nm)	d_e (nm)
Fe I 538.3 nm	$3d^7 4p - 3d^7 4d$ $z^5G^o - e^5H$	-0.003	-0.0352
Fe I 539.3 nm	$3d^6 4s4p - 3d^6 4s5s$ $z^5D^o - e^5D$	0.005	0.0254
Fe I 542.4 nm	$3d^7 4p - 3d^7 4d$ $z^5G^o - e^5H$	-0.007	-0.062

Table 1: The values of Stark shift d_e determined from the broadest Fe I component in Figure 3 and d_m taken from (Konjević et al 2002).

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