



Determination of gas temperature of high-frequency low-temperature electrodeless plasma using molecular spectra of hydrogen and hydroxyl-radical

Z. Gavare*, A. Svagere, M. Zinge, G. Revalde, V. Fyodorov

Institute of Atomic Physics and Spectroscopy, University of Latvia, Skunu Street 4, LV-1050 Riga, Latvia

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ABSTRACT

In this study we determine gas temperature of He+H₂, Ar+H₂ and Ne+H₂ high frequency electrodeless lamps using intensity distribution in rotational spectra of hydrogen molecule and hydroxyl radical. The measurement results show that OH rotational spectra can be successfully used for estimation of gas temperature of the high-frequency electrodeless lamps at very small amounts of hydrogen present in plasma. The analysis of the measurement results leads to the conclusion that in “pure” plasma the applied energy is used to heat plasma, while for the plasma with hydrogen addition the energy is used in chemical reactions.

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1. Introduction

Our work is concerned with development and manufacturing of electrodeless high-frequency low-temperature light sources (HFEDL). Such light sources are widely used as bright radiators of narrow spectral lines in different types of scientific devices, for instance, in atomic absorption spectrometers. For each particular use these light sources have to be optimized, and it is of great importance to use reliable methods for these investigations and further on for improvement of the light sources.

Although knowledge of temperature of heavy particles (hereinafter referred to as gas temperature) in plasma is not sufficient to understand and explain different processes, its measurement is important for the optimization of the plasma conditions in the light source.

It is well known that for the discharge gas temperature estimation in HFEDLs one can use the high-resolution

spectroscopy method of the emission spectral line shape measurements using Fabry–Perot interferometer and Zeeman spectrometer together with non-linear spectral line shape modeling [1]. However, the method of the line shape modeling is very complicated due to the necessity to solve the incorrect inverse task [2]. It is of great interest to find other methods and verify their applicability for the determination of gas temperature in HFEDLs. In the case of hydrogen containing plasmas, one of the commonly used techniques for the determination of gas temperature is based on the measurements of the intensity distribution in the rotational bands of hydrogen molecule [3–14], for instance (0–0), (1–1), (2–2), and (3–3) Q-branches of Fulcher- α ($d^3\Pi_u^- \rightarrow a^3\Sigma_g^+$) electronic transition [5–14]. In one of our previous works we showed that this method can be successfully used for the hydrogen containing HFEDLs [13]. It has been observed that hydrogen diffuses out of the lamp through the walls during the lamp operating time. As a result it is not possible to register Fulcher- α Q-branches in the spectrum of plasma; although the hydrogen is present in the plasma [13] (it is still possible to register atomic lines of hydrogen). However,

* Corresponding author. Tel.: +371 67225907.

E-mail address: zanda.gavare@gmail.com (Z. Gavare).

one can try to register the UV spectrum of OH radical ($A^2\Sigma^+ \rightarrow X^2\Pi$; $v' = v'' = 0$) at 306.4 nm. It is known that this band can be observed even at very small amounts of hydrogen [15] and it can be used for the gas temperature determination, too [16–23]. In [24] we used OH rotational spectra to determine gas temperature of dumbbell-type HFEDLs; but it is of interest to compare the values of gas temperature determined using different methods to verify the relation between the rotational and translational temperatures.

In order to validate the usage of OH rotation spectra to determine gas temperature, in this study we compare these values with those determined using Doppler broadening of the spectral line. Since one of our previous works [13] showed, that HFEDLs can be successfully used to investigate the influence of hydrogen addition to the argon electrodeless plasma, another task in this study was to determine the hydrogen influence on helium plasma.

2. Theoretical background

2.1. Gas temperature determination from the Doppler broadening of the spectral line

For the determination of gas temperature from the Doppler profile of the spectral line it is necessary to register the spectral line shape using Fabry–Perot interferometer. The problem is that spectral line profile, registered by means of the interferometer, differs significantly from the real one. To determine the real spectral line profile, it is necessary to solve the inverse task. In this work we used another method, namely, the line fitting by means of a non-linear multiparameter chi-square fit.

The total broadening of the spectral line consists of (1) *natural broadening*, (2) *Doppler broadening*, (3) *Van der Waals broadening*, and (4) *resonance broadening*. The line shapes caused by natural broadening, resonance broadening, as well as by broadening due to the interaction with surrounding particles, are all described by the Lorentz function.

The basic broadening of the spectral line in low-pressure plasma is the Doppler broadening, characterizing the temperature of emitting atoms with the Maxwellian distribution of velocities. In that case, the spectral line shape has a Gaussian profile.

The resulting line is a convolution of the both shapes, the Gaussian and the Lorentzian, and is described by the Voigt function. To obtain the real Doppler width of the spectral line, it is necessary to fit the line profile and deconvolute from the Lorentz function, which in the case of Fabry–Perot measurements is mainly composed of the instrumental function [25]. Then gas temperature can be derived from the well-known formula:

$$T = \mu \left(\frac{\Delta\lambda_D}{\lambda_0} \frac{1}{7.16 \times 10^{-7}} \right)^2, \quad (1)$$

where T is the gas temperature, $\Delta\lambda_D$ is the Doppler broadening of the spectral line, λ_0 is the transition wavelength and μ is the atomic mass.

2.2. Gas temperature derivation from the intensity distribution in rotational spectra of molecule

Generally, for gas temperature determination from rotational spectra of OH radical, the following relation is used [26,27]:

$$\lg\left(\frac{I\lambda}{A}\right) = \text{const} - \frac{0.625}{T^{\text{rot}}} E_i, \quad (2)$$

where in *const* all factors, which are independent of rotational quantum number N , are combined, I is the measured line intensity, T^{rot} is the rotational temperature, E_i is the excited state energy, λ is the transition wavelength and A is the transition probability.

The determination of the gas temperature from measured intensity distribution in rotational bands of hydrogen molecule is based on a certain theoretical model, which involves series of assumptions about the mechanism of the excitation–deexcitation of rotational–vibrational levels of hydrogen. According to this model, the translational gas temperature may be in equilibrium with rotational temperature determined from either upper or lower energy level depending on processes in plasma under study. In case of low-pressure plasmas (typically, at pressures lower than 1 Torr), the rotational temperature obtained from the lower level can be considered as a valid estimation of gas temperature [5,7,8,28].

In this study we use simple model, according to which the intensity of the rotational–vibration transition is related to rotational temperature with following expression [5–13]:

$$\ln\left(\frac{I}{g_{a,s}(2N+1)v^4}\right) = \text{const} - \frac{E_{XON}}{T^{\text{rot}}}, \quad (3)$$

where E_{XON} is the ground state energy, v is the transition frequency and $g_{a,s}$ is the statistical weight of the n, v, N rotational–vibrational level of the homonuclear molecule depending on the magnitude of the nuclear spin and the symmetry in respect to permutation of the nuclei (a or s). $g_{a,s}$ is equal to unity for heteronuclear molecules.

Formulas (2) and (3) show, that in practice it is possible to determine rotational temperature by plotting $\lg(I\lambda/A)$ vs. E_i (or $\ln(I/g_{a,s}(2N+1)v^4)$ vs. E_{XON}), and fitting the data with linear function. Rotational temperature can be determined from the slope of the fitted line. The necessary transition parameters for the lines of (2–2)Q branch of Fulcher- α band can be found in [13,29–31], and for the lines of OH (A–X) (0–0)Q₁ branch in [26,27,32]. This data is compiled in Table 1.

3. Experimental part

3.1. Plasma under study

Plasma under study was several high-frequency electrodeless lamps (HFEDLs) with different fillings: (1) He + H₂ ($p_{\text{He}} \approx 0.9$ Torr, $p_{\text{H}_2} \approx 0.1$ Torr), (2) Ar + H₂ ($p_{\text{Ar}} \approx 0.9$ Torr, $p_{\text{H}_2} \approx 0.1$ Torr), (3) Ne + H₂ ($p_{\text{Ne}} \approx 0.9$ Torr, $p_{\text{H}_2} \approx 0.1$ Torr).

Table 1Transition parameters for the lines of (A–X) (0–0)Q₁ branch of OH radical [26,27,32] and H₂ Fulcher- α (d³ Π_{u^-} → a³ Σ_g^+) (2–2)Q branch [13,29–31].

OH (A ² Σ^+ → X ² Π) (0–0)Q ₁				H ₂ Fulcher- α (d ³ Π_{u^-} → a ³ Σ_g^+) (2–2)Q				
Line	λ , nm	E , cm ⁻¹	A , 10 ⁸ s ⁻¹	Line	λ , nm	E_{XON} , cm ⁻¹	N	$g_{a,s}$
Q ₁ (4)	308.328	32,779	33.7	Q ₁	622.4815	118.50	1	3
Q ₁ (5)	308.517	32,948	42.2	Q ₂	623.0258	354.35	2	1
Q ₁ (6)	308.734	33,150	50.6	Q ₃	623.8391	705.54	3	3
Q ₁ (8)	309.239	33,652	67.5	Q ₄	624.9150	1168.78	4	1
Q ₁ (9)	309.534	33,952	75.8	Q ₅	626.2495	1740.21	5	3
Q ₁ (10)	309.859	34,283	84.1					

These lamps were manufactured at the *Institute of Atomic Physics and Spectroscopy, University of Latvia*. The cylindrical lamps with diameter of 2 cm and the length of 4 cm were placed into an induction coil and an inductive coupled discharge was excited by means of a high-frequency field of about 100 MHz frequency. The power of the discharge was changed by varying the applied current i into the coil in the region 80–200 mA. The gas temperature dependence from the current i in the induction coil was investigated.

Initially, in emission spectra of He+H₂ and Ar+H₂ HFEDLs the molecular spectra of H₂ could be observed [13], and, the gas temperature could be determined using Fulcher- α Q branches. However, during the operating time, hydrogen diffuses through the walls, and the changes in the spectra can be observed, namely, after longer operating time Fulcher- α bands cannot be registered anymore, in the same time we can observe OH band at 306.4 nm. OH molecule was created from H₂, which was filled in the lamp and O₂, which was emitted directly from the SiO₂ glass walls of the lamp during the discharge [24].

3.1.1. Ar+H₂ HFEDL

In one of our previous works [13] we reported the study of hydrogen influence on argon high-frequency electrodeless plasma, by organizing the experiment in the following way: (1) during the first measurement session the Fulcher- α (2–2)Q branch spectra was registered and rotational temperature determined; (2) then the HFEDL was operated so long until the hydrogen was diffused out from the lamp through the walls. The diffusion of the hydrogen was indicated registering the spectra in the same spectral region as before, and during this, second, measurement session, the gas temperature was determined using the Doppler broadening of argon lines. Further measurements of this HFEDL showed, that although the molecular spectra of hydrogen could not be registered anymore, the hydrogen was still present in plasma (since the atomic lines of hydrogen could be still observed), and at 306.4 nm we could identify intense emission of OH. In this study we determined rotational temperature for this HFEDL from OH emission and compared to the gas temperature values determined from Doppler profiles.

3.1.2. He+H₂ HFEDL

In this work we use He+H₂ HFEDL to estimate the hydrogen influence on helium plasma. In order to achieve

it, the work was organized in similar to described in [13] with argon–hydrogen plasma: (1) during the first measurement session the Fulcher- α (2–2)Q branch spectra was registered and rotational temperature (equal to gas temperature) determined; (2) then the HFEDL was operated so long until the hydrogen was diffused out from the lamp, and during this measurement session the gas temperature was determined from relative intensities of lines of OH (A–X) (0–0)Q₁ branch.

3.1.3. Ne+H₂ HFEDL

In emission spectra of Ne+H₂ HFEDL the H₂ molecular bands could not be observed, however the OH rotational band at 306.4 nm could be registered. The gas temperature was determined using the relative intensities of (A–X) (0–0) Q₁-branch of OH radical.

3.2. Gas temperature derivation from the Doppler broadening of the spectral lines

For the gas temperature determination from the Doppler profile, profiles of Ar 772.4 nm (Ar I 1 s₅–2 p₇) line were registered by means of Fabry–Perot interferometer. The light, collected from the lamp, is transmitted through the pressure-scanned interferometer, focused on a monochromator, amplified and registered by means of a photomultiplier. Line profiles of Ar 772.4 nm line were recorded using mirrors with a dielectric coating and a 1.4 cm spacer (free spectral range of 0.36 cm⁻¹).

The profiles were fitted and deconvoluted from the Lorentz function, mainly composed of the instrumental function in our case [25], to obtain the real Doppler profile. Deconvolution was done using *Origin* software.

3.3. Rotational temperature determination from the rotational spectra of hydrogen molecule

The light from the lamp was imaged on the entrance slit of the spectrometer (*JobinYvon SPEX 1000M*, focal length 1 m) and detected by means of a charge-coupled device matrix detector (2048 × 512 *Thermoelectric Front Illuminated UV Sensitive CCD Detector, Symphony*). The emission spectra of the discharge under study have been recorded in the wavelength range $\lambda = 620$ –630 nm, containing first five lines of the (2–2)Q branch of the Fulcher- α band system of the hydrogen molecule.

An example of the registered spectra and a typical semilogarithmic plot of the Fulcher- α (2–2)Q branch lines

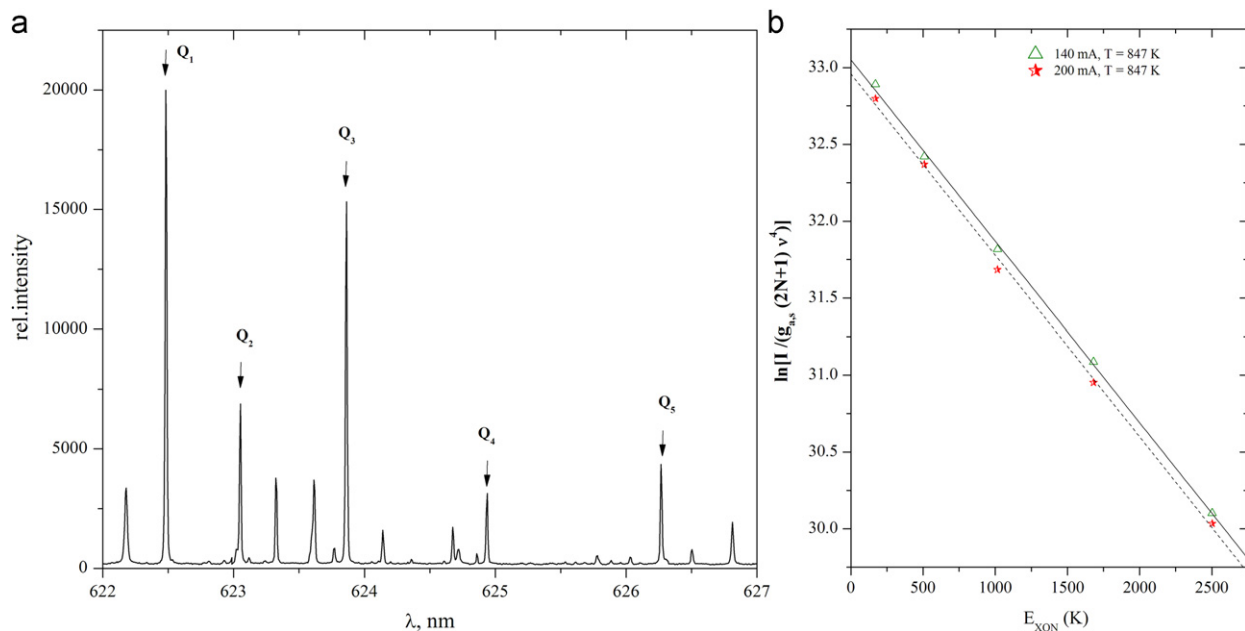


Fig. 1. (a) Example of a typical emission spectra of the hydrogen containing plasma of HFEDL recorded at applied current $i = 180$ mA. First five lines of Fulcher- α (2–2)Q branch are identified. (b) The semilogarithmic plot of scaled intensity of Fulcher- α (2–2)Q lines versus rotational energy of the molecular hydrogen ground state for He+H₂ HFEDL at $i = 140$ mA and $i = 200$ mA.

are shown in Fig. 1(a) and (b), respectively. The linearity of these plots indicates the Boltzmann distribution over the rotational levels.

3.4. Rotational temperature determination from the rotational spectra of OH radical

The experimental setup was the same as for the rotational temperature determination from the rotational spectra of hydrogen molecule. The emission spectra of the discharge under study have been recorded in the wavelength range $\lambda = 306$ – 312 nm. In Fig. 2 an example of the recorded emission spectra with identified OH lines is shown.

From analysis of the recorded OH spectrum, it was concluded that for the rotational temperature derivation only $Q_1(4)$, $Q_1(5)$, $Q_1(6)$, $Q_1(8)$, $Q_1(9)$ and $Q_1(10)$ lines can be used, since $Q_1(1)$ – $Q_1(3)$ and $Q_1(7)$ lines overlap with lines of other branches, and lines starting with $Q_1(11)$ cannot be identified in the spectra due to their overlapping with lines of other branches or due to their small intensities.

4. Results and discussion

4.1. Ar+H₂ HFEDL

Initially this lamp was filled with 0.9 Torr Ar and 0.1 Torr H₂. As it was mentioned before, this lamp was used in previous measurements [13], where it was operated until H₂ was diffused out of lamp, and the emission of hydrogen could not be registered anymore, however we could register OH emission. Since the content of H₂ in the plasma decreases (hydrogen is diffused through the

walls), increases the degree of dissociation of hydrogen molecules (the concentration of hydrogen atoms becomes higher) [33], which can lead to the subsequent formation of OH radical.

In Fig. 3 the dependence of the estimated gas temperature in dependence of the excitation generator current is shown.

The results of both methods are in very good agreement, confirming that the relative intensities of OH (A–X) (0–0)Q₁ branch can be successfully used for HFEDL gas temperature determination.

The uncertainties for both methods are in the range 6–12%. The temperature change is very explicit (from 620 K till 1100 K) by varying excitation generator current from 80 mA till 200 mA.

4.2. He+H₂ HFEDL

In Fig. 4 the temperature dependence on the excitation generator current for the He+H₂ (9:1) HFEDL is shown. Rotational temperature (equal to gas temperature) determined from Fulcher- α (2–2)Q branch does not change significantly by changing the current of excitation generator. For this HFEDL the determined gas temperature was around 680–700 K (the uncertainties did not exceed 15%).

After the first measurement session the lamp was operated until the hydrogen diffused out of the lamp, leading to the changes in the plasma discharge conditions. During the second measurement session gas temperature was determined using OH band. It can be seen that the temperature dependence on applied current is completely different compared to the case when hydrogen is still present in plasma.

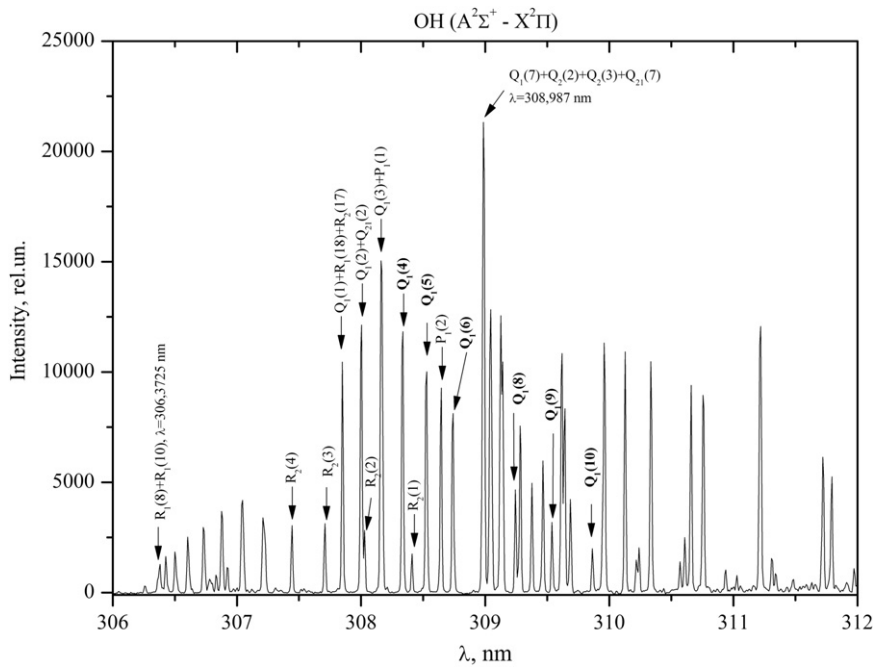


Fig. 2. Typical spectrum of OH radical band at 306.4 nm. He+H₂ HFEDL emission at $i=200$ mA.

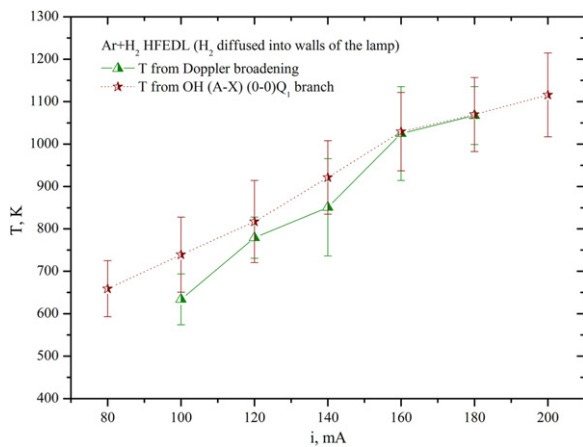


Fig. 3. The temperature dependence on the applied current for Ar+H₂ HFEDL. Gas temperature values obtained using OH (A-X) (0-0)Q₁ branch are compared with ones obtained from Doppler profile of Ar I 772.4 nm line (hydrogen is diffused out of the lamp).

The gas temperature ranging from 530 K to 788 K was obtained, depending on the applied current (80–200 mA). Relative uncertainty of the obtained temperature values is less than 10%, except for temperature value determined at 80 mA being 14%.

4.3. Ne+H₂ HFEDL

As it was mentioned earlier, in the emission spectra of Ne+H₂ HFEDL it was not possible to observe molecular bands of H₂, this could be due to intense collisions between excited neon atoms with hydrogen molecules leading to the dissociation of hydrogen molecule [34,35].

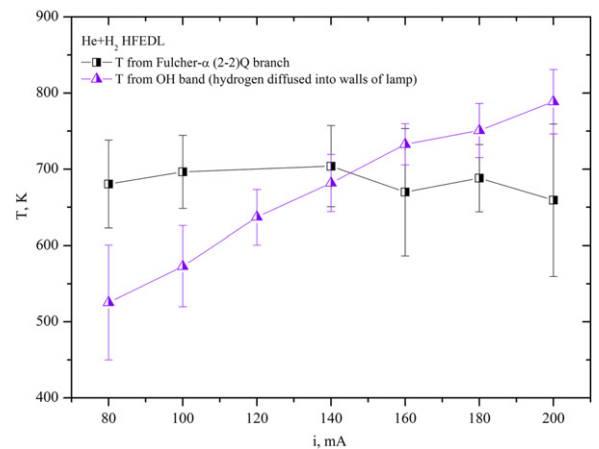


Fig. 4. The temperature dependence on the excitation generator current for the He+H₂ (9:1) HFEDL. Comparison of gas temperature values obtained using H₂ Fulcher- α (2-2)Q branch (1st measurement session) and OH (A-X) (0-0)Q₁ band (2nd measurement session; hydrogen is diffused out of the lamp).

While the excitation generator current values were in range from 100 mA till 140 mA, the emission of OH band was weak in comparison with the neighboring atomic lines of Ne, yet at 200 mA current value the OH emission increased exceeding the intensity of atomic lines of Ne in that region.

It is expected that intensity of atomic spectral lines will also increase by increasing the current of excitation generator, however in the case of Ne+H₂ HFEDL neon lines in the vicinity of OH band (at 306.4 nm) did not increase in intensity, while intensity of Ne I spectral lines in red region increased noticeably. This allows us to

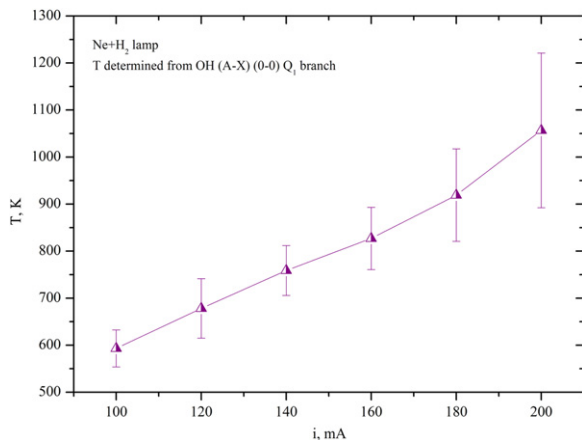


Fig. 5. The temperature dependence on the applied current for Ne+H₂ HFEDL. Temperature values were obtained using OH (A–X) (0–0)Q₁ branch.

conclude that part of excited Ne atoms is quenched by OH radical, leading to the increase in intensity of OH emission.

From emission of OH radical we also determined gas temperature of Ne+H₂ HFEDL. In Fig. 5 one may see temperature dependence on the applied current for this HFEDL. Temperature values were obtained using OH (A–X) (0–0)Q₁ branch. The temperature change was very explicit by varying the current of excitation generator, and values were in range from 580 K till 1080 K. The uncertainties in this case were in range 7–15.5%.

These explicit temperature dependences on excitation generator current determined for Ar+H₂, He+H₂ and Ne+H₂ HFEDLs can be explained by the fact that in “pure” gases the energy is used to heat plasma, as well as for the ionization and excitation of atoms, while in mixtures supplied energy is used more in the chemical reactions. By increasing the applied current we increase the input energy, which in “pure” gases will lead to the increase in gas temperature (this is what we observe for HFEDLs without hydrogen). Similar observations regarding gas temperature were made by authors of [36].

5. Conclusions

Measurement results confirm that OH rotational spectra can be successfully used for the high-frequency electrodeless gas temperature estimation at very small amounts of hydrogen present in plasma, even if spectra of molecular hydrogen cannot be registered.

The analysis of changes we observed in spectra of Ne+H₂ HFEDL leads to the conclusion that in this plasma excited neon atoms supply energy to OH radicals via collisions leading to increase of intensity of OH emission.

The explicit temperature dependence on excitation generator current which was observed for HFEDLs without hydrogen can be attributed to the fact that plasma becomes “pure” and the applied energy is used to heat plasma, while for the plasma with hydrogen addition the energy is used in chemical reactions and the gas temperature stays constant by varying excitation generator current.

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