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Optical characteristics of UV–VUV lamps on the electronic-vibrational transitions of the hydroxyl radical pumped by a nanosecond capacitive discharge

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Abstract: This paper presents the results of a study of UV and VUV low-pressure lamp pumped by nanosecond capacitive discharge. This lamp works on the mixtures of helium and argon with water vapor (H_2O) or 'heavy' water (D_2O). At the capacitive low-pressure lamp emission spectra are mainly narrow band 308 nm OH (A–X) and large band in the VUV spectral range: 140–200 nm OH (C, B, A). Found, that the increasing the partial pressure of H_2O and D_2O (>130–150 Pa) led to a decrease in the intensity of VUV radiation of hydroxyl radicals. In the transition from He/H_2O mixture to He/D_2O mixture, the optimal pressure of helium is decreased. Reduced partial pressure of argon led to increased intensity of radiation of hydroxyl in the VUV range. The study of emission characteristics of OD* – lamps on a mixture of He/D_2O and the OH* – lamps on a mixture of He/H_2O showed that the intensity of the bands OD (X, C, A) is 1.5 times larger than the intensity of the bands OH (X, C, A). The maximum absolute intensity of UV–VUV radiation of the lamp (at a frequency of 1000 Hz) reached 1 W.

1 Introduction

In recent years, new efficient sources of ultraviolet and vacuum ultraviolet radiation from exciplex molecules and halogen molecules are widely used in science, photobiology, photochemistry and environment [1, 2]. The working environment of these lamps unlike of mercury lamps is environmentally friendly and does not create problems with recycling of used emitters. The working medium of the excimer lamps is composed of a costly inert gases (xenon, krypton) and agressive halogen molecules (Cl_2, Br_2 etc.). Therefore, relevant is the search for new, more cheap and less aggressive fluids for the new UV–VUV lamps.

The previously developed gas-discharge sources (for the region of 110–190 nm) overlap the spectral range in the form of individual narrow bands, so promising is the study of broadband VUV radiation.

Recently, the UV radiation used for disinfection of water, air, sterilisation of packing materials for food products etc. [3]. The absorption spectrum of a DNA molecule has two peaks near the wavelengths of 200 nm and in the band of 250–270 nm. The inactivation of viruses and cells of different origins is promising using gas discharge lamps working on molecules with maximum radiation $\lambda < 310$ nm [2]. In addition, radiation in the spectral range of 180–190 nm passes through the atmosphere and can be used in different non-vacuum techniques.

Such media for low-pressure lamps may be mixtures of inert gases with water vapor. These sources are excited by the DC glow discharges. It was obtained in [4–6] that lamp of longitudinal low-pressure glow discharge based on the He (or Ar)/H_2O mixtures emits system on the bands 309 nm OH (X–A) and in the range 190–140 nm OH (X, C, B). Adjusting the partial pressure of water vapor gave the prevalence of UV or VUV radiation of the lamp. The results of the simulation of physics processes in the lamp glow discharge on a mixture of He/H_2O are presented in [7]. To increase the lifetime of the lamp working on the water vapor one can use a capacitive discharge in which discharge electrodes are not in contact with the plasma. This lamp emits a pulsed radiation whose time duration is determined mainly by the parameters of the source ignition of the discharge lamp. Previous research studies have shown that the energy of UV and VUV pulses can be increased significantly by the use of nanosecond high-voltage pulses for lamp pumping.

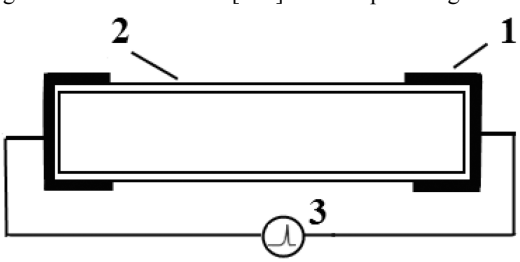

Fig. 1 *Scheme of the lamp pumped by a nanosecond capacitive discharge: 1: electrodes; 2: quartz tube with sealed ends; 3: discharge power supply*

In the present work, we present the results of studies of the optical characteristics of the lamp pumped by the nanosecond capacitive discharge working on the mixtures of He and Ar with H_2O and D_2O.

2 Experimental setup

The repetitively pulsed capacitive discharge is ignited in a cylindrical tube made of quartz. The inner diameter of the tube is 0.5 cm and its length is 20 cm. The short-wave radiation (in the range of 200–140 nm) is registered through the window of lithium fluoride. The tube is docked with the front part of the one-vacuum monochromator through the open end and LiF-window. The gases and water vapor are injected into the tube through the special gas mixing system. Vapours of distilled (H_2O) or 'heavy' (D_2O) water are letting from the pre-pumped of the glass containers with liquid. Electrodes in the lamp are the nickel sheet rings with the width of 2 cm. They are placed outside of the discharge tube (Fig. 1).

This structure of the emitter minimises contact of discharge plasma with the metal electrodes. In this experiment, we used experimental setup, which was also used to study of the lamp on a mixture of helium and water vapor with pumping by a nanosecond barrier discharge. The characteristics, parameters of the plasma of lamp of the barrier discharge, are shown in [8]. The study of VUV spectrum of the discharge is performed using the one-vacuum monochromator and a photomultiplier with LiF-window

(FEU-142). The double diffraction monochromator (MDR-2) and a photomultiplier FEU-106 are used for the study of the spectral range of 200–400 nm. The signal from the photomultiplier is amplified by an amplifier and is recorded by an electronic potentiometer. Calibration of the registration system of the relative intensity of the radiation depending on the wavelength of the radiation is conducted by the deuterium lamp emission and the discharge in hydrogen (VUV range). The accuracy of measurement of the relative intensity of UV–VUV a lamp radiation is 5–7%. Accuracy of measurement of partial pressure of water vapor is 10 Pa and of the pressure of inert gases is 250 Pa. The absolute emission power of the lamp is estimated using the 'Kvarts-01' taking into account the experimental geometry and the spectral sensitivity of the head of the probe.

The amplitude of the voltage pulses having time duration of 50 ns is 25–40 kV. The pulse repetition rate is varied in the range 50–1000 Hz.

3 Optical characteristics

The repetitively pulsed nanosecond discharge in He/H_2O (D_2O) uniformly fills the entire volume of the discharge tube. The color of electric discharge working with He is white and working with Ar is pink and with the shades of purple.

Figs. 2–4 show the typical emission spectra of lamp working on the mixtures of He and Ar with vapors of H_2O or D_2O.

Fig. 2 shows the emission spectrum of the lamp on the He/D_2O mixture. The main emitters in the UV–VUV spectral range are the hydroxyl bands OD (A, C, X). The major bands of emission in the spectral range $\Delta\lambda = 175$–225 nm are 181.0, 187.1, 189.1 nm. Previously, these bands were also observed in the longitudinal DC glow discharge in [5, 7]. In the UV, a wavelength range $\Delta\lambda = 270$–315 nm is the most intense band with the maximum at λ ; 310 nm (OD) (X–A; 0.0). We observed less intense bands with the maxima at $\lambda = 283.7$ nm OD (X–A, 1.0) and $\lambda = 297.6$ nm OD (X–A; 3.2).

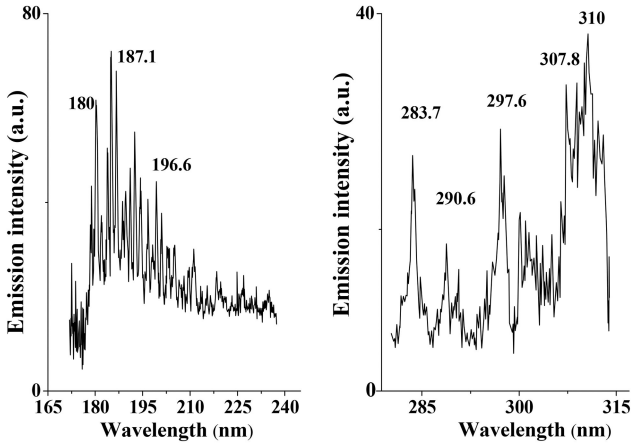

Fig. 2 *Emission spectrum of the capacitive discharge lamp working on the mixture p(He)–p(D_2O) = 8.0–0.13 kPa*

Comparison of the emission spectrum of the OD*-lamp He/D_2O with the emission spectrum of the OH*-lamp He/H_2O with the same composition (under the same conditions of the discharge ignition) has shown that the intensity of all bands OD (X, C, A) is 1.5 times larger than for bands of OH (A–C, A). The increase in the partial pressure of vapor water and 'heavy' water beyond the limits of 130–150 Pa led to the decrease in the intensity of VUV radiation of the hydroxyl radicals.

Fig. 3 shows the effect of the He partial pressure on the spectral characteristics of the OH*-lamp on the He/H_2O mixture. The band OH 156 nm (X–B) is seen in the spectrum for the reduced helium pressure. The increase of the partial pressure of helium from 5 to 21 kPa led to the decrease of the intensity of the emission of bands of the OH radical.

Fig. 4 shows typical spectrum of the low-pressure lamp working on the mixture of argon and water (H_2O) vapors. The VUV radiation occupies ~70% of the total intensity of the UV–VUV spectrum of the plasma. The dominant lines of this spectrum are the shortwave lines of the hydroxyl radical.

Figs. 5–7 show the main results of optimisation of the lamp radiation intensity working on the mixture of helium and water vapor. To optimise the lamp, we varied the partial composition of the working mixture. At p(He) = 2.6 kPa, the optimum partial pressure of water vapor for the emission bands the hydroxyl OH (X–A) is in the range of 130–150 Pa (Fig. 5). We obtained for p(He) = 2.6 kPa that replacing H_2O by D_2O does not change significantly the character of the dependence of the intensity of the bands of OH (X–A) (Fig. 5).

However, the intensity of the line increases ~1.5 times. We also obtained the peak intensity of the X–C band (176–198 nm) for the water pressure of 20 and 130 Pa.

The obtained decrease of the emission intensity of OH bands X, A, C, B with increasing partial water pressure (Fig. 5) is due to the quenching of the excited states of hydroxyl radical by the water molecules. As in the lamp of DC glow discharge in the He/H_2O mixture, the quenching of states C and B of the radical OH was much stronger than A state.

Fig. 6 shows the influence of the helium partial pressure on the intensities of the OH (X–A) bands (p(H_2O) = 130 Pa).

One can see the first maximum of the UV radiation of OH at p(He) = 0.7–3.5 kPa. This is explained by the high spatial uniformity of the capacitive discharge along the radius of the discharge tube. The second maximum of intensity of the radiation of OH* was observed in the pressure range of helium 5–15 kPa when the plasma emission intensity is the highest from the central part of the discharge tube.

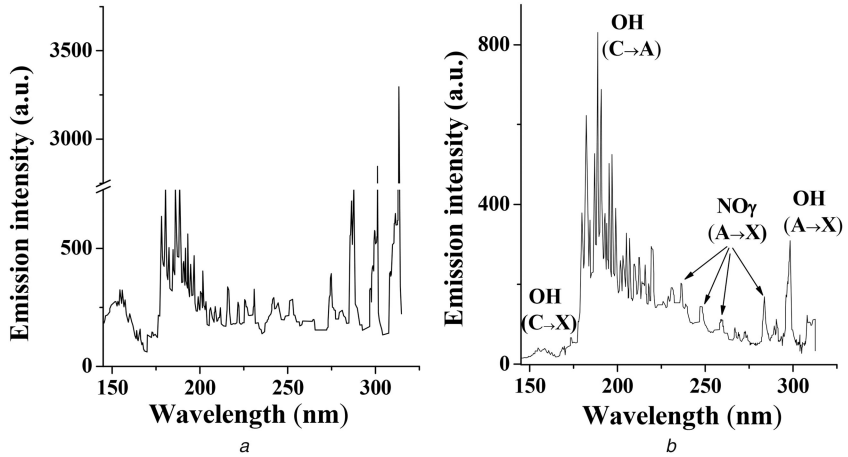

Fig. 3 *Emission spectra of the pulse-periodic UV–VUV lamp working on the mixtures*
(a) p(He)–p(H_2O) = 21.3–0.11, ***(b)*** 5–0.11 kPa

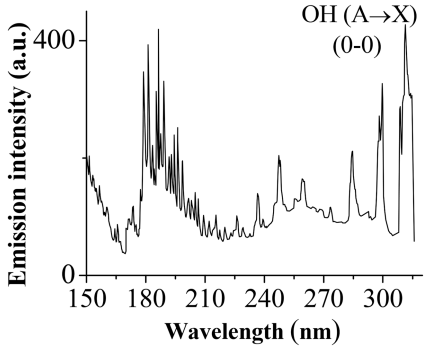

Fig. 4 *Emission spectrum of the pulse-periodic UV–VUV lamp working on the mixture of p(Ar)–p(H_2O) = 1.3–0.13 kPa*

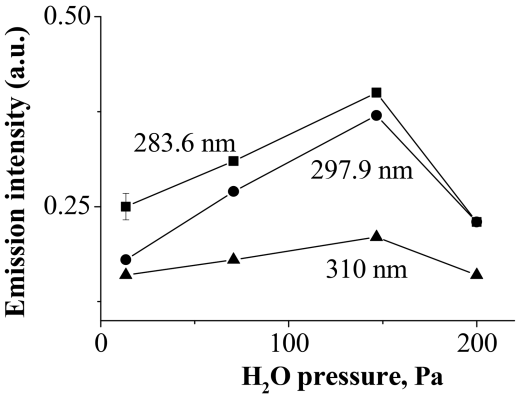

Fig. 5 *Dependence of the intensity of the hydroxyl radical emission bands on the partial pressure of water vapor; He/H_2O mixture, p(He) = 2.6 kPa*

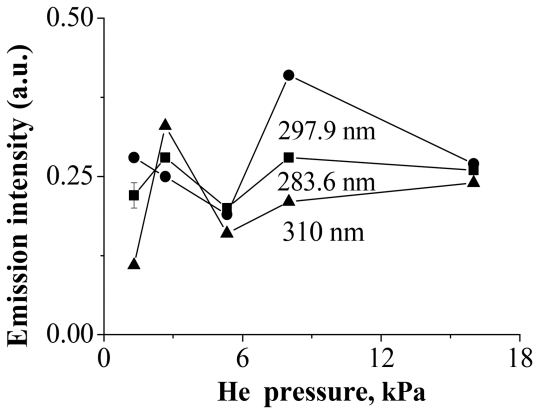

Fig. 6 *Influence of the He partial pressure on the intensity of the OH (X–A) emission band; He/H_2O mixture, p(H_2O) = 130 Pa*

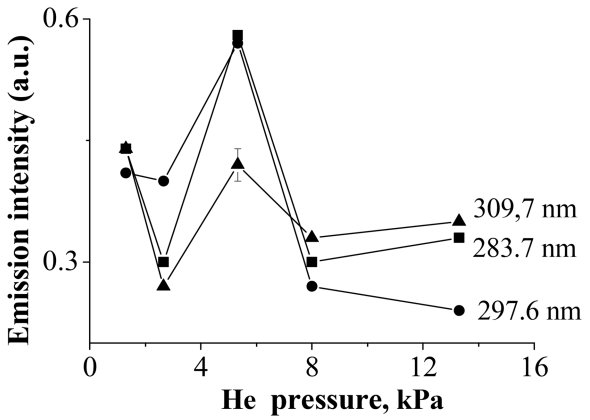

Fig. 7 *Influence of the He partial pressure on the intensity of the emission bands of the hydroxyl radical; He/D_2O mixture, p(D_2O) = 130 Pa*

The optimum partial He pressure for the lamp working on the He/D_2O mixture in homogeneous and constricted regimes of capacitive discharge are 1.3 and 5.3 kPa, respectively (Fig. 7).

The optimal pressure of D_2O vapors was in the range 100–130 Pa. The decrease of the optimum pressure of water vapor during the transition from normal to 'heavy' water is probably due to the more efficient quenching of A, C and B states of the hydroxyl molecules by D_2O. The transition from He/H_2O to He/D_2O leads to the decrease of the optimum helium pressure. The results of optimisation of the intensity of the emission bands in the spectral range $\Delta\lambda$ = 176.0–183.9 and 184.5–198.0 nm for p(H_2O) = 106.6 Pa have shown that the optimal helium pressure for the homogeneous and constricted discharge is 1 and 11 kPa, respectively.

The results of the optimisation of the optical characteristics of the nanosecond capacitive discharge lamp working on the Ar/H_2O mixture are shown in Figs. 8 and 9. Fig. 8 shows the results of optimisation of the VUV intensity of radiation of OH bands depending on the argon partial pressure for the fixed p(H_2O) and the partial pressure of water vapor for the fixed pressure of argon. We found that to obtain maximum intensity of the VUV emission, the optimal argon pressure must be in the range of 0.1–0.5 kPa (Fig. 8*a*).

The dependence of the intensity of OH (X–C) bands of the lamp on the partial pressure of water vapor (Fig. 8*b*) shows that their optimal pressure is in the range 0.06–0.16 kPa. The decrease in intensity of emission bands of hydroxyl radical for increasing pressures of argon and water vapor can be associated with the quenching of OH (A, C) and the decrease in the electron temperature in the plasma.

We also tried to optimise the intensity of the 308.9 nm band. We obtained the maximum intensity for the water vapor pressure in the range 13–0.20 kPa (Fig. 9*a*). To obtain the maximum intensity of the band 308.9 nm, argon pressure must be ∼1.33 kPa for the homogeneous discharge mode and ∼12 kPa for the stratified

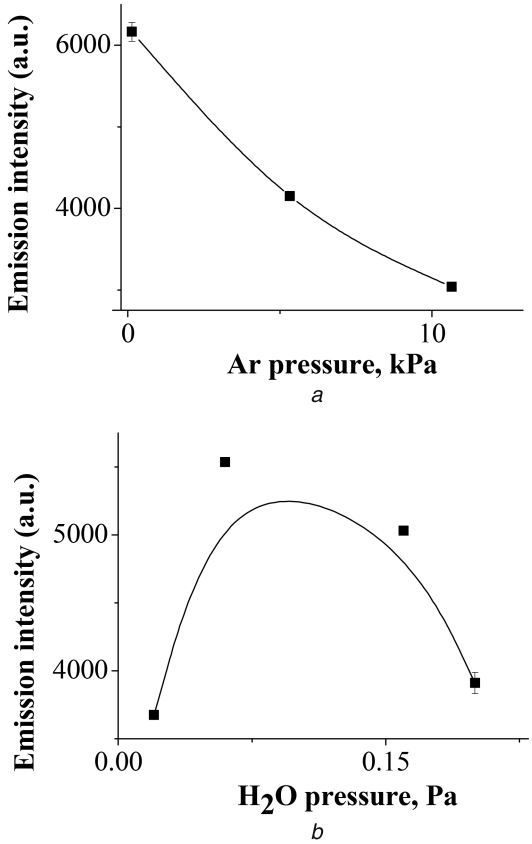

Fig. 8 *Dependence of the intensity of the emission bands in the range 150–200 nm on*
(a) Ar partial pressure at p(H_2O) = 0.13 kPa, ***(b)*** Water partial pressure at p(Ar) = 2.66 kPa; Ar/H_2O mixture

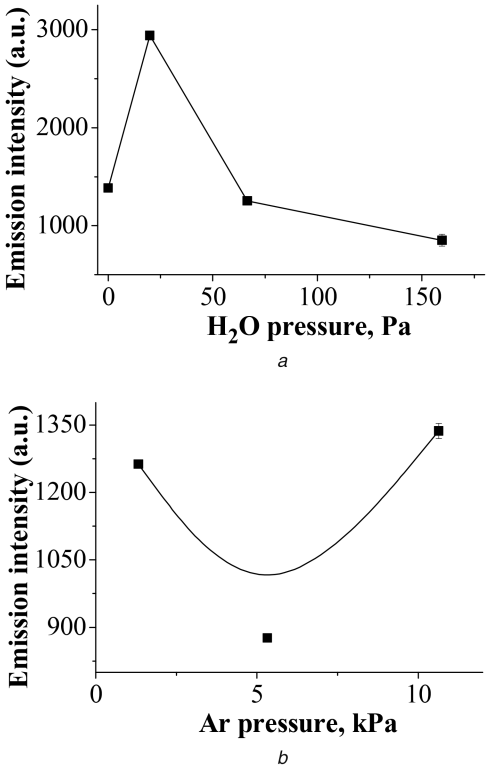

Fig. 9 *Dependence of the intensity of the emission band 308.9 nm of OH radical on*

(a) Partial pressure of water vapour at p(Ar) = 2.66 kPa, ***(b)*** Partial pressure of argon at p(H_2O) = 0.133 kPa; Ar/H_2O mixture

discharge mode (Fig. 9*b*). The main cause of foliation of the discharge of low pressure in the medium of electro-negative molecules (water molecules) is forming 'ion–ion' plasma in the central part of the discharge tube. Normal 'electron-ion' plasma is formed at the periphery of the discharge near inner surface of the glass tube. This is due to the large difference in the electron drift velocity and negative ions along the radius of the discharge tube (electrons fast leaves the center of the tube, where there are only slow negative ions) [9].

The physical mechanism of ignition of capacitive discharge similar to mechanism of ignition of stationary longitudinal glow discharge of low pressure. In the capacitive discharge lamp on the mixtures of water vapor with helium basic mechanisms of formation of excited hydroxyl radicals is reaction [7]:

$$e + H_2O \rightarrow H + OH(A) + e\,. \quad (1)$$

The generation of OH radicals (A) occurs in the next process

$$e + OH \rightarrow OH(A) + e\,. \quad (2)$$

In mixtures, where one component is argon, the transfer of energy from metastable atoms Ar to molecules of water [6] is important. Also, this mechanism occurs in the barrier discharge, when a pressure of argon is high.

Our experiments have shown that the intensity of the characteristic hydroxyl radical bands is the linear function of the repetition rate of the voltage pulses and of the voltage amplitude. We did not obtain the saturation of intensity. The maximum absolute intensity of UV–VUV radiation during the short turning on of the lamp (1 kHz repetition frequency) reached 1 W.

4 Conclusion

The study of the optical characteristics of the UV–VUV hydroxyl lamp pumped by the nanosecond capacitive discharge showed that the efficiency of the lamp working on the 'heavy' water (D_2O) vapors is ~1.5 times larger than that working on the ordinary water (H_2O) vapor. We obtained that the discharge remains spatially homogeneous for the helium and argon pressures in the range 0.1–3 kPa. For higher pressure of He and Ar (8–15 kPa), the central part of the discharge was also a rather effective source of the shortwave radiation. The optimal partial water vapor pressure for the VUV radiation was in the range 10–20 Pa while the optimal pressure for the emission of 309 nm OH (X–A) band was in the range 130–150 Pa. The power of the lamp emission did not exceed 1 W.

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