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*UV–VUV EMITTERS*

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# Characteristics of a nanosecond-barrier-discharge-pumped multiwave UV–VUV lamp on a mixture of argon, krypton and vapours of freon

A.K. Shuaibov, A.I. Minya, R.V. Hrytsak, Z.T. Gomoki

*Abstract.* **We present the results of investigation of the characteristics of a nanosecond-barrier-discharge-pumped multiwave lamp based on a gas mixture of Ar–Kr–$CCl_4$, which emits in the spectral range of 170–260 nm. The main emission bands in the lamp spectrum are ArCl (B → X) near 175 nm, KrCl (B → X) near 222 nm and $Cl_2$ (D′→A′) near 258 nm. The lamp intensity with respect to pressure, working mixture composition and pump regime is optimised.**

***Keywords:*** *barrier discharge, lamp, chlorides of argon and krypton, chlorine and freon molecules, radiation intensity.*

Mastering of the near-VUV spectrum region (160–190 nm) of the total bactericidal wavelength range (160–280 nm) can be achieved through the development of a lamp emitting simultaneously at several wavelengths (multiwave lamp) on the basis of the bands of argon and krypton chlorides: ArCl (B → X) with $\lambda \approx 175$ nm and KrCl (B → X) with $\lambda \approx 222$ nm [1]. The results of research on the characteristics of emitters based on ArCl (B → X) pumped by a pulsed volume discharge are presented in [2, 3]. The VUV radiation of the band of argon chloride is usually accompanied by radiation of the band of chlorine molecule D′→A′ near $\lambda = 258$ nm, which is close to the wavelength of the resonance spectral line of mercury atom (253 nm) that is widely used in various optical technologies based on application of mercury vapour lamps. The perspective of using a lamp that emits near $\lambda = 258$ nm compared to a low-pressure mercury lamp ($\lambda = 253$ nm) for bactericidal applications is conditioned by complete coincidence of the main absorption maximum of the DNA molecule with the band of chlorine rather than with the spectral resonance line of mercury atom. The working environment of emitters based on argon chloride and chlorine is much cheaper than that of excimer xenon lamps ($\lambda \approx 172$ nm).

The exciplex lamps pumped by a volume discharge emit the bands ArCl (B → X) with $\lambda \approx 175$ nm and $Cl_2$ (D′→A′) with $\lambda \approx 258$ nm at a peak intensity up to 0.4 kW cm$^{-2}$. However, the service life of such emitters on a single working mixture is less than a few tens of hours. Another drawback is small working aperture of the emitter.

In work [4], the results of investigating the characteristics of multiwave UV lamps on the Ar–Kr–$Cl_2$ and Ar–Kr–$Br_2$ mixtures pumped by a pulsed barrier discharge are presented. However, the VUV radiation of the ArCl (B → X) and ArBr (B → X) bands has not been investigated in these experiments, which may lead to underestimation of the efficiency and radiation power of these multiwave emitters.

Radiation of the band of the KrCl molecule was observed in the experiments using single- and two-barrier lamps of nanosecond duration [5]. The peak intensity of the lamps was 0.5 kW cm$^{-2}$ at 4 ns. In contrast to work [5], Panchenko and Tarasenko [6] investigated the KrCl excilamp excited by a barrier discharge of sub-microsecond duration at a frequency $f = 50$ Hz (the lamp radiation power constituted more than 100 kW at the efficiency up to 10%).

To increase the pulse power and working resource of a multiwave lamp, which is promising for applications in photochemistry, photo-medicine and ecology, a repetitively pulsed nanosecond barrier discharge ignited in a mixture of Ar – $CCl_4$ – $H_2O$ was used in [7]. This lamp emits the bands with $\lambda = 175$ and 258 nm, and a band of OH (A → X) with $\lambda = 308$ nm at comparable intensities.

For a more uniform overlap of the bactericidal wavelength range and for an increase in the output power and selective impact on individual biomolecules, we need high-power UV–VUV emitters with a possibility of restructuring their emission spectrum, and sufficiently long working life in the gas-static regime, herewith a mixture of Kr–Ar–$CCl_4$ may serve as a working environment for those emitters. However, the output characteristics of such multiwave lamps pumped by a high-voltage barrier discharge of nanosecond duration are virtually unexplored.

In this paper we present the results of an experimental study on characteristics of a nanosecond-barrier-discharge-pumped UV–VUV lamp on a mixture of Ar–Kr–$CCl_4$.

A high-voltage nanosecond discharge with two barriers made of quartz was ignited in a cylindrical bulb made of KU-1 grade quartz. The working length of the bulb of the multiwave UV–VUV emitter constituted 20 cm, with a diameter of the inner quartz tube equal to 14 mm. The distance between the inner and outer barriers was about 4.5 mm. The inner electrode of the lamp was made of a solid aluminium cylinder and installed into the inner quartz tube. The external electrode was a spiral nickel wire with a transparency of 80%. The effective surface area of the emitter was 360 cm$^2$.

A pulsed barrier discharge was ignited in the no-filamentation regime by means of a source of high voltage pulses with resonant overcharging of an accumulative capacitance made of KVI-3 capacitors. The total capacitance of the modulator was 1.54 nF, and a TGI-I-1000/25 hydrogen thyratron was used as a switch. The amplitude of the voltage pulses at the modulator output increased approximately three-fold with

**A.K. Shuaibov, A.I. Minya, R.V. Hrytsak, Z.T. Gomoki** Uzhgorod National University, Podgornaya ul. 46, 88000 Uzhgorod, Ukraine, e-mail: roksolanija@ukr.net

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the use of an impulse cable transformer, and reached 40 – 50 kV with duration of about 20 ns for a single voltage surge. The amplitude of the main maximum of the current pulse reached 50 A with its duration of 20–30 ns. The voltage pulse repetition rate could be varied in the range of 35–1000 Hz. The current and voltage pulses were recorded using a Rogowski coil, low-inductance voltage divider and a high-speed 6-LOR oscilloscope. The absolute power of the lamp was measured with a 'Kvarts-01' device.

Plasma emission was analysed in the spectral range of 140–310 nm using a vacuum monochromator and a FEU-142 photomultiplier with a window made of lithium fluoride. Figure 1 shows the emission spectrum of the multiwave UV–VUV lamp on a mixture of argon, krypton and vapours of $CCl_4$. The lamp radiation overlaps the spectral range of 170–260 nm and consists of most intense bands of the chlorine molecule as well as of chlorides of argon and krypton. At a low partial pressure of krypton (0.3–1.3 kPa), the ArCl (B → X) bands with $\lambda \approx 175$ nm, KrCl (D → X) with $\lambda \approx 199$ nm, KrCl (B → X) with $\lambda \approx 222$ nm and Cl2 (D′→ A′) with $\lambda \approx 258$ nm are observed in the emission spectrum of the lamp. These bands have previously been observed in the spectra of UV–VUV emitters based on a longitudinal low-pressure glow discharge and in the spectrum of the elevated-pressure barrier discharge in a mixture of Ar–Kr–$Cl_2$ [4]. The intensity distribution in the lamp spectrum is the most sensitive to the partial pressure of krypton in the mixture, because the energy of the lower metastable energetic levels is the lowest in the atom of krypton, and the 'harpoon' reaction and step ionisation of atoms of heavy inert gases in the ion–ion channel of formation of exciplex molecules play an important role in the formation of molecules of halides of heavy inert gases.

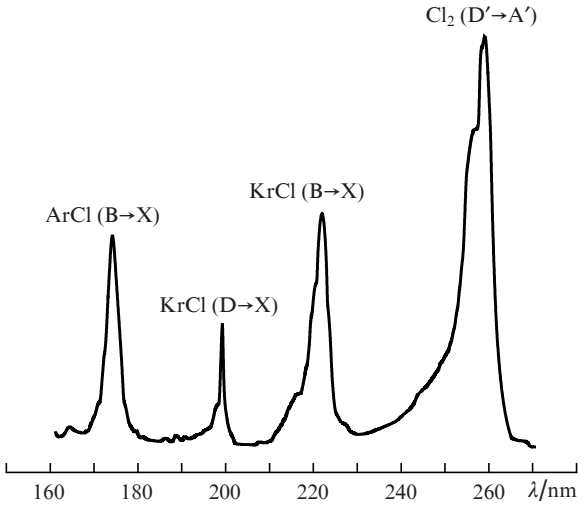


**Figure 1.** Emission spectrum of the multiwave UV–VUV lamp on the mixtures Ar:Kr:$CCl_4$ = 6.6:1.33:0.13 kPa at a repetition rate of current pulses $f$ = 80 Hz.

The increase in partial pressure of krypton up to 6.6 kPa in a nanosecond barrier discharge in the Ar–Kr–$CCl_4$ mixture resulted in the dominance of the bands of chloride krypton in the emission spectrum of the lamp.

The peculiarity of the use of freon molecules ($CCl_4$) as a chlorine-carrier [compared to more simple molecules (HCl, $Cl_2$)] in the lamps based on chlorides of heavy inert gases [8] is the intensity increase in the emission band of $Cl_2$ (D′ → A′) with $\lambda \approx 258$ nm compared to the bands of ArCl (B → X) with $\lambda \approx 175$ nm, KrCl (D → X) with $\lambda \approx 199$ nm and KrCl (B → X) with $\lambda \approx 222$ nm. Introducing krypton into the double mixture of Ar–$CCl_4$ (at $p_{Kr} \approx 1.3$ kPa) results not in a strong change in the total intensity of UV–VUV molecular bands, but only in a redistribution of their intensities among themselves.

The main process that controls the relation between the intensities of the emission bands of ArCl (B → X) with $\lambda \approx 175$ nm and KrCl (B → X) with $\lambda = 222$ nm in the barrier discharge is the reaction of substitution of atoms of argon by atoms of krypton in the formation of corresponding exciplex molecules:

$$\text{ArCl (B)} + \text{Kr} \rightarrow \text{KrCl (B)} + \text{Ar}, \qquad (1)$$

the mechanism of which is associated with the energy transfer from the metastable atoms of argon to the atoms of krypton. These reactions are characterised by rather large rate constants (for example, in the reaction of substitution of the atoms of krypton by the atoms of xenon in formation of XeCl (B) molecule, this constant constitutes $7\times10^{-10}$ $cm^3$ $s^{-1}$ [9]).

Figure 2 shows electrical characteristics of the lamp. With account for the voltage drop on the dielectric barriers, the voltage pulse amplitude reached 22 kV at the duration of a separate voltage surge of about 8–10 ns. The amplitude of the main current pulse peak was 25–50 ns for the pulse duration of 10–30 ns. The main contribution of energy into the plasma of a nanosecond barrier discharge in a ternary mixture lasted within ~50 ns. The maximum pulse power introduced into the discharge reached 0.15–0.20 MW. Due to mismatch of the pulse voltage generator output with the load (lamp), three peaks of energy contribution were observed in the course of 75 ns.

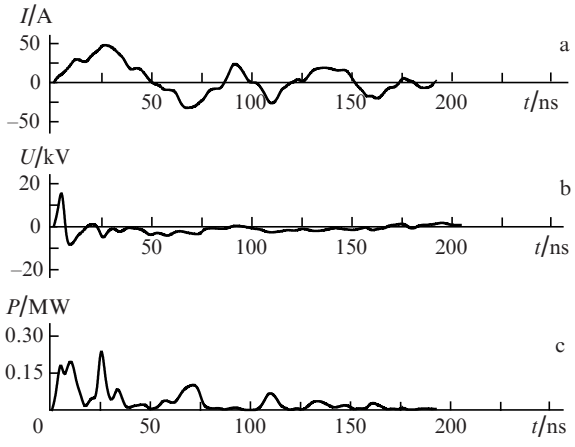


**Figure 2.** Oscillograms of (a) the current pulses, (b) voltage and (c) power supplied into the nanosecond barrier discharge in a mixture of Ar:Kr: $CCl_4$ = 6.6:1.33:0.13 kPa at the working capacitor voltage of 13 kV ($f$ = 80 Hz).

The main results of optimisation of the radiation intensity of the multiwave UV–VUV lamp with respect to the pressure and partial composition of the working mixture are shown in Figs 3 and 4.

The study of emission of the bands of argon and krypton chlorides has shown that the optimum partial vapour pressure of freon ($CCl_4$) lies in the range of 130–180 Pa, whilst the

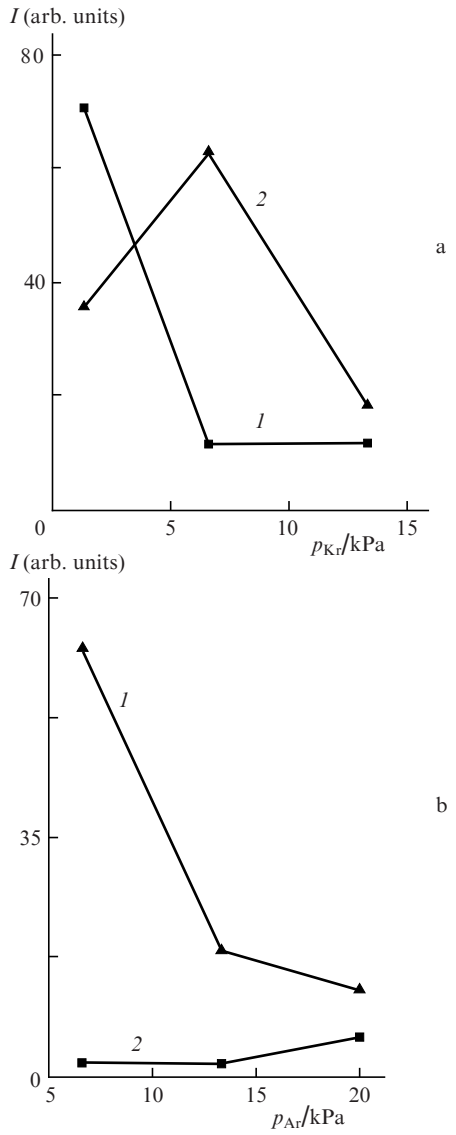


**Figure 3.** Radiation intensity of the bands of KrCl (B → X) with $\lambda = 222$ nm (*1*), ArC (BOX) with $\lambda = 175$ nm (*2*) as a function of (a) the partial pressure of krypton in the lamp on the mixture $Ar-Kr-CCl_4$ at $p_{CCl_4} = 130$ Pa and $p_{Ar} = 6.6$ kPa and (b) of the partial pressure of argon in the lamp on the same mixture at $p_{CCl_4} = 130$ Pa and $p_{Kr} = 6.6$ kPa.

band intensity of chlorine molecule increases linearly with increasing partial pressure of freon in the range of 30–300 kPa.

An increase in the partial pressure of argon in a ternary mixture (at constant $p_{CCl_4}$ and $p_{Kr}$) in the range of 6.5–13.5 kPa leads to a strong decrease in intensity of the emission bands of KrCl (B → X) and ArCl (B → X). It has virtually no effect on the intensity of the $Cl_2$ (D′ → A′) band with $\lambda \approx 258$ nm, which is mainly due to a decrease in the parameter $E/p$. This causes a decrease in the concentration of metastable atoms and positive ions of heavy inert gases, which largely determine the effectiveness of formation of halides of inert gases as a result of 'harpoon' reactions and ion–ion recombination.

An increase in the partial pressure of krypton from 1.2 to 6.6 kPa (at $p_{Ar}$ = const) resulted in a significant decrease in the intensity of the band with $\lambda \approx 175$ nm due to reaction of substitution of argon atoms by krypton atoms in the formation of chlorides of argon and krypton in plasma. Herewith, the radiation intensity of the band of chlorine molecule also

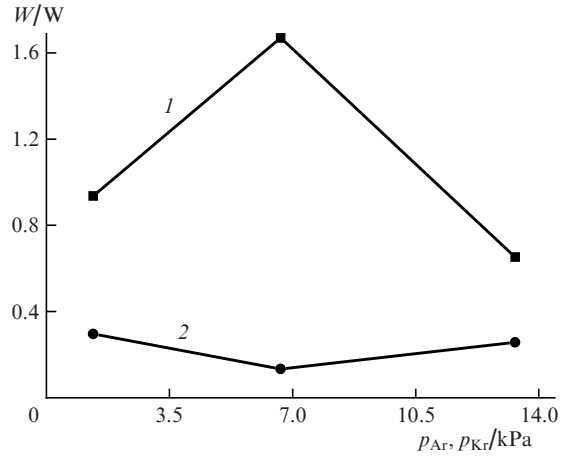


**Figure 4.** Average output power of the multiwave UV–VUV lamp on a mixture $Ar-Kr-CCl_4$ as a function of the partial pressure of krypton (at $p_{Ar} = 1.3$ kPa) (*1*) and argon (at $p_{Kr} = 6.6$ kPa) (*2*) for $p_{CCl_4} = 130$ Pa.

decreased by about an order. To obtain the maximum intensity of the emission bands of KrCl (B → X, D → X), the optimal partial pressure of krypton in the ternary mixture should lie in the range of 6–8 kPa.

The maximum average radiation power of the lamp reached 1.4–1.7 W (Fig. 4) at the optimum partial pressure of krypton $p_{Kr}$ = 6–8 kPa and fixed argon pressure $p_{Ar}$ = 1.3 kPa. Further increase in $p_{Kr}$ in the ternary mixture resulted in a decrease in the average output power of the UV–VUV lamp. At $p_{Ar} = 1.3$ kPa and optimal $p_{Kr}$, the average radiation power reached the first peak, and at a higher partial pressure of argon its second maximum was observed. In this case, the emission spectrum changed significantly – it consisted mainly of the bands of chlorine molecule and krypton chloride. Optimisation of the dependences of the output power on the partial pressure of argon shows that the output power of a lamp on the basis of a ternary mixture is twofold higher compared to the output power of a lamp on the $Ar-CCl_4$ mixture.

When cooling the lamp bulb by the air flow from a ventilator, the lamp worked stable for a long time at a current pulse repetition rate of 40–150 Hz. The intensity of the main emission bands of the lamp increases with increasing number $N$ of discharge pulses in this frequency range up to $(3-5)\times10^5$, and then does not change up to $N = 10^6$ (experiments with a larger number of pulses were not performed).

At a higher frequency, only a short-duration (5 – 10 min) switching on of the lamp is possible. In this case, the lifetime dramatically decreases, presumably due to the low efficiency of the air cooling system. In the range of 40–400 Hz, frequency dependences of intensity of the emission bands of chlorine and chlorides of inert gases are nonlinear. With a frequency increase up to 1000 Hz, these dependences become linear with no signs of saturation. Such a character of the frequency dependences of the intensity may be associated with the contribution of the dissociation products of freon molecules in the formation of exciplex molecules and molecules of $Cl_2$ (D′).

Thus, the study on the output characteristics of the multiwave UV–VUV lamp based on the $Ar-Kr-Cl_2$ gas mixture has shown that the lamp emits the bands of chlorides of krypton and argon with $\lambda \approx 175$ and 222 nm, respectively, and a band of chlorine molecule with $\lambda \approx 258$ nm. For the emission bands B → X of ArCl and KrCl molecules, the optimum par-

tial pressure of freon vapours lies in the range of 130–180 Pa, and for the band of $Cl_2$ with $\lambda \approx 258$ nm constitutes 300 Pa; the optimum partial pressure of krypton lies in the range of 6–8 kPa and the optimum pressure of argon is 1.3 kPa. The average output power of the lamp reaches 1.7 W at a pulse repetition rate of 80 Hz, and the working lifetime of the lamp in the gas-static regime within the frequency range of 40–150 Hz amounts to $10^6$ pulses, which is about two orders higher than that for the corresponding lamps on the basis of a transverse volume discharge [3, 10]. Varying the pressure and partial composition of the gas mixture allows controlling the lamp emission spectrum in the wavelength range of 170–260 nm. This can be used for simultaneous and selective destruction of bonds in various bases of nucleic acids: in particular, the absorption maximum of adenine is located at $\lambda \approx 260$ nm (which corresponds to the emission band of chlorine molecule), whilst that of cytosine – at $\lambda \approx 220$ nm (which corresponds to the emission band of krypton chloride) [11].

## References


1. Boichenko A.M., Lomaev M.M., Panchenko A.N., Sosnin E.A., Tarasenko V.F. *Ultrafioletovye i vakuumno-ultrafioletovye eksilampy: fizika, tekhnika i primeneniya* (Ultraviolet and Vacuum-Ultraviolet Excilamps: Physics, Technology and Applications) (Tomsk: 'SST', 2011).
2. Shuaibov A.K., Dashchenko A.I. *Kvantovaya Elektron.*, **30** (3), 279 (2000) [*Quantum Electron.*, **30** (3), 279 (2000)].
3. Panchenko A.N., Tarasenko V.F. *Kvantovaya Elektron.*, **36** (2), 169 (2006) [*Quantum Electron.*, **36** (2), 169 (2006).
4. Avdeev S.M., Sosnin E.A., Smirnov A.A., Tarasenko V.F. *Izv. Tomskogo Polytekh. Univ., Ser. Energ.*, **317**, 161 (2010).
5. Erofeev M.V., Tarasenko V.F. *J. Phys. D: Appl. Phys.*, **39**, 3609 (2006).
6. Panchenko A.N., Tarasenko V.F. *Kvantovaya Elektron.*, **38** (1), 88 (2008) [*Quantum Electron.*, **38** (1), 88 (2008)].
7. Shuaibov A.K., Minya A.I., Gomoki Z.T., Gritsak R.V. *Opt. Spektrosk.*, **114**, 30 (2013).
8. Gritsak R.V., Shuaibov A.K., Minya A.I., Gomoki Z.T., Varga T.I. *Vestn. Uzhgorodskogo Natsional'nogo Univ., Ser. Fiz.*, **34**, 5 (2014).
9. Shuaibov A.K., Shevera V.S. *Opt. Spektrosk.*, **47** (8), 409 (1979).
10. Shuaibov A.K., Chigin' V.I., Shimon L.L., Shevera I.V., Gorun P.P., et al. *Zh. Tekh. Fiz.*, **80** (5), 87 (2010).
11. Shuaibov A.K., Shevera I.V., Shimon L.L., Sosnin E.A. *Suchasni djerela ultrafioletovogo viprominyuvannya: rozrobka ta zastosuvannya* (Modern Sources of Ultraviolet Radiation: Development and Applications) (Uzgorod–Tomsk: 'Goverla', 2006).


***PERSONALIA***

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# Vasily Ivanovich Shveikin

The editorial council, editorial board and editorial stuff of 'Quantum Electronics', collective and directorate of OJSC 'M.F. Stel'makh Polyus Research Institute', colleagues and disciples congratulate prominent Russian scientist, pioneer in the development of semiconductor lasers, doctor of technical sciences, professor, laureate of the Lenin Prize of the USSR, former deputy director of the Polyus Research Institute Vasily Ivanovich Shveikin on his 80th birthday and wish him sound health, creative longevity, happiness and success in all his endeavours.

Promising developments in semiconductor lasers and devices on their basis, which have been supervised by V.I. Shveikin, are even today very important. Many of these devices are widely used in the national economy. Research and production base, which ensures their civil and special applications, was established under V.I. Shveikin's leadership at the institute.

For his work in the field of semiconductor lasers Vasily Ivanovich Shveikin was decorated with the Order of the Red Banner of Labour and the Jubilee Medal 'For Valiant Labour – In commemoration of the 100th Anniversary of the Birth of Vladimir Ilych Lenin'. He holds the title of the 'Best Designer of the Industry' and was awarded the lapel badge 'Honorary Worker of the Electronics Industry'.