PACS numbers: 42.72.Bj; 52.80.-s DOI: 10.1070/QE2013v043n08ABEH015085

# **Optical characteristics of a HgBr excilamp**

A.A. Malinina, A.N. Malinin, A.K. Shuaibov

Abstract. Optical characteristics of a coaxial HgBr excilamp on multicomponent mercury dibromide vapour mixtures with helium, nitrogen and sulfur hexafluoride are investigated under pumping by a pulse-periodic barrier discharge. Stable excilamp operation was demonstrated at a pump pulse repetition rate of 3-9 kHz. The component composition of the working system was determined, which provides a maximal average and pulsed specific radiation power of 48.8 mW cm<sup>-3</sup> and 40.6 W cm<sup>-3</sup>, respectively, at the efficiency of 7.3% in the blue-green spectral range with the maximal radiation intensity at the wavelength of 502 nm. The reduction in the radiation power after  $2.5 \times 10^6$  shots is 5%. Interpretation is given for the results of optimisation of excilamp characteristics.

**Keywords:** excilamp, barrier discharge, gas-discharge plasma, mixture components, mercury dibromide, nitrogen, sulfur hexafluoride, helium.

## 1. Introduction

An interest in the exciplex sources of spontaneous emission in the visible, UV and VUV spectral ranges is related with promising practical applications. In many applications, it is sufficient to have the incoherent high-power source with a large luminous area (of more than 100 cm<sup>2</sup>) emitting in a relatively wide wavelength interval. Excilamps are simple in production and easily scalable in the dimension, pump power and geometry of active volume. Their employment is promising in various technologies, medicine and biotechnology [1–14]. In addition, excilamps of the blue-green spectral range emitting in the range of photoactive plants may be used for solving agrophysical problems on enhancing the efficiency of exciting chlorophyll molecules, thus increasing the growing rate in the conditions of greenhouse enterprises [15–17].

Presently, several types of exciplex sources of spontaneous visible-range emission are designed and studied, in which the working mixture is excited by a barrier discharge or other types of discharge [6,8-12,18,19].

In this work we present the results of experimentally studied influence of the component composition of a working mixture, energy stored in the dielectric capacitance, and pulse repetition rate on spectral, energy, and temporal characteristics of radiation of a coaxial HgBr excilamp.

A.A. Malinina, A.N. Malinin, A.K. Shuaibov Uzhgorod National University, ul. Pidgirna 46, 88000 Uzhhorod, Ukraine; e-mail: mal@univ.uzhgorod.ua

Received 16 December 2012; revision received 7 February 2013 *Kvantovaya Elektronika* **43** (8) 757–761 (2013) Translated by N.A. Raspopov

## 2. Investigation technique and methods

The radiation characteristics of excilamp were investigated with the working mixtures of mercury dibromide vapours with helium, nitrogen and sulfur hexafluoride. The working mixtures were pumped by a pulse-periodic barrier discharge.

The schematic diagram of the coaxial excilamp is shown in Fig. 1. The lamp was made of a quartz tube. The external diameter and length of the tube were 8.8 mm and 7 cm, respectively. The tube wall was 1-mm thick. Inside the tube along its axis, the molybdenum electrode with a diameter of 2 mm was placed. The discharge gap was 2.4-mm wide. The external perforated electrode had the length of 3 cm (with the transmission coefficient of ~0.7). The operating volume was ~1 cm<sup>3</sup>. The capillary 1.5 mm in diameter was placed at the end of the quartz tube. It was intended for reducing the evacuation of mercury dibromide vapours from the lamp to a vacuum gasmixing system.



Figure 1. Schematic diagram of the coaxial excilamp: (1) internal electrode; (2) external perforated electrode; (3) discharge zone; (4) quartz glass; (5) capillary; (6) valves of gas evacuation and feeding systems.

The excilamp was excited by a nanosecond-pulse generator (Fig. 2). A TGI 1-35/3 thyratron was used as a switch, and the storage capacitance was collected from KVI-3 low-inductance capacitors. This capacitance was recharged through a primary winding of the step-up transformer with the turn ratio of 1:4. The value of the capacitance was 1.36 nF. The generator provided the voltage pulses with an amplitude of 1-10 kV, duration of 400–600 ns, and repetition rate of 1-9 kHz.

The working mixtures were prepared directly in the emitting volume. The mercury dibromide powder (HgBr<sub>2</sub>) was evenly spilled in amounts of 60 mg into the excilamp bulb. In studying the excilamp radiation, the partial pressure of saturated mercury dibromide vapour was provided by the selfheating due to discharge energy dissipation and by heating the bulb with an external heater (if a partial pressure of mercury dibromide vapour above 0.1 kPa was needed). After charging the salt, the radiation source was dehydrated by heating it to 70 °C and evacuating for two hours through the capillary (5) (Fig. 1).



**Figure 2.** Electrical circuit of the pump generator: (Cm) commutator; (*C*) storage capacitor; (Tr) step-up transformer.

The partial pressure of mercury dibromide vapour was measured by the temperature of the most cold point on the lamp surface using interpolated reference data [20]. In the conditions of our experiment the pressure varied within the limits 0.1-2 kPa. The partial pressures of helium, sulfur hexafluoride and nitrogen were measured by a standard pressure gauge and vacuum indicator.

The excilamp radiation was detected in the direction normal to the lateral surface of the quartz tube and was analysed in the spectral range 380–600 nm. The radiation spectra were recorded by a grating monochromator (with the grating 600 grooves mm<sup>-1</sup>). The spectral resolution of the recording system was 2.4 nm. It was calibrated by using a SI 8-200 standard tungsten lamp operated at the filament temperature T = 2173 K.

The voltage and current pulses of the emitter were observed with a C1-72 oscilloscope which detected the signals from a voltage divider and integrated circuit of a calibrated Rogowski loop.

The average power of excilamp radiation was measured by a 'Kvarts-01' device. An optical signal after passing the diaphragm with the area of 0.25 cm<sup>2</sup> and the SZS-16 light filter with a maximum transmission wavelength of 500 nm entered the measuring head of the device. The radiation power from the whole surface of the radiation source was determined according to the method given in [21].

### 3. Investigation results and discussion

#### 3.1. Oscillograms of pump pulses

Immediately after applying the voltage pulses to excilamp electrodes, a set of the conical micro-discharges was observed in a working volume, which had the cone vertex on the metal electrode and base on the internal surface of the quartz emitting tube. The discharge colour (pink) at the initial stage (for first 30 seconds) was determined by helium buffer gas. Then for 30–60 s the discharge colour was blue-green. The discharge colour became more saturated with mixture self-heating. In this case the discharge was diffused and uniform; the brightness contrast was noticeably smoothed in a volume discharge and microdischarges.

Characteristic oscillograms of the pump pulses (voltage and current) are shown in Fig. 3. The oscillating structure of the current pulses is explained by a repeated discharge of the dielectric capacitance during the action of the voltage pulse with the amplitude sufficient for the breakdown of the discharge gap [22]. A distinction in the oscillating structure of current pulses at the leading and trailing edges of a voltage pump pulse is related with opposite directions of the current



**Figure 3.** Oscillograms of the voltage pulses across excilamp electrodes U and the discharge current I in the mixtures HgBr<sub>2</sub>: He = 0.1:117 at the total pressure of mixture p = 117.1 kPa (a); HgBr<sub>2</sub>: N<sub>2</sub>: He = 0.1:4:120 at p = 124.1 kPa (b); HgBr<sub>2</sub>:SF<sub>6</sub>: He = 0.1:0.07:117 at p = 117.17 kPa (c); and HgBr<sub>2</sub>:SF<sub>6</sub>: N<sub>2</sub>: He = 0.1:0.07:4:117.2 at p = 121.37 kPa (d). The pulse repetition rate is f = 6 kHz.

flowing through the gas-discharge separation (2.4 mm) and, hence, with unequal conditions of discharge decomposition on the internal surface of dielectric under the single-barrier discharge used in our experiment.

The distinction was also observed in the current pulses in the mixture comprising nitrogen (Fig. 3b) and without it (Fig. 3a). In the mixture with nitrogen, at the leading edge of a pump pulse one can see the characteristic pulse with higher amplitude and longer duration then in the mixture without nitrogen. In addition, at  $t \simeq 400$  ns its amplitude is also greater than that in the mixture without nitrogen, which may be related with a greater charge of the dielectric surface as compared to that in the mixture HgBr<sub>2</sub>-He. If sulfur hexafluoride was added to the mixture (Fig. 3c), the pulse intensities in the structure were redistributed in the negative half-cycle in favour of a single pulse, and in the positive half-cycle the intensities of the pulses following the first one were increased. Oscillograms in the mixture comprising sulfur hexafluoride and nitrogen (Fig. 3d) are specific in that the oscillating character of the current pulse is less pronounced than in the mixtures without nitrogen and sulfur hexafluoride.

#### 3.2. Spectral and integral characteristics of radiation

Spectral and integral characteristics of the HgBr excilamp radiation were studied with various compositions of the working mixtures  $HgBr_2-He$ ,  $HgBr_2-N_2(SF_6)-He$ , and  $HgBr_2-SF_6-N_2-He$ , at the partial pressures of helium 80–160 kPa, nitrogen 1–10 kPa, sulfur hexafluoride up to 200 Pa and mercury dibromide to 2 kPa. The maximal amplitudes of voltage and current pulses, and the repetition rate were 9–10 kV, 9–11 A, and 3–9 kHz, respectively.

Characteristic spectra of excilamp radiation in the visible spectral range are shown in Fig. 4 for the working mixtures HgBr<sub>2</sub>-He and HgBr<sub>2</sub>-SF<sub>6</sub>-He. In these mixtures, a spectral band with a maximum at the wavelength of 502 nm was observed. The band was of sparse oscillating character and corresponded to the electron-vibration transition  $B^2\Sigma_{1/2}^+ \rightarrow X^2\Sigma_{1/2}^+$  of mercury monobromide (HgBr\*) [23]. The main part of the radiation intensity was concentrated in the wavelength range 450-512 nm. An abrupt intensity fall was observed at longer wavelengths and smooth reduction in the range of



**Figure 4.** Excilamp radiation spectra in the mixtures  $HgBr_2:SF_6:He = 0.1:0.07:117$  at p = 117.17 kPa (1) and  $HgBr_2:He = 0.1:117$  at p = 117.1 kPa (2). The pump pulse rate and amplitude is f = 6 kHz and  $U_a = 9$  kV, respectively.

shorter wavelengths. The shape of the spectral band and its half-height width (15-16 nm) are similar to those for the spectral bands corresponding to the transition  $B \rightarrow X$  in mercury mono-halogenides [6, 8, 11, 18].

Results of investigations of integral characteristics of excilamp radiation are presented in Fig. 5. In all the mixtures, under an elevating pressure of their components, the radiation power increased to a maximal value and then reduced. Note that all the mixtures under study comprised helium. Its partial pressure was taken 118–120 kPa, in which case a maximal radiation power was observed in the mixture HgBr<sub>2</sub>–He (Fig. 5a).

The character of the dependences of the excilamp radiation power on the pressures of helium, nitrogen, sulfur hexafluoride and mercury dibromide vapour (Fig. 5) is determined by a higher electron concentration at enhanced pressures of He, N<sub>2</sub> and SF<sub>6</sub> in the mixture, by a variation of the part of energy spent to mixture heating, by variations of the average electron energy and rate of excitation of HgBr\* molecule subject to parameter E/N (E is the intensity of the electric field applied to the gap space, N is the total concentration of atoms and molecules in the working mixture), and by the process of quenching the  $B^2\Sigma_{1/2}^+$  state of HgBr<sup>\*</sup> molecules in their collisions with atoms of helium, nitrogen, and molecules of sulfur hexafluoride and mercury dibromide [24, 25]. The considerable increase in the lamp radiation power in the mixtures of mercury dibromide vapour with sulfur hexafluoride and helium (Fig. 5c) as well as with added nitrogen (Fig. 5d) is related with the processes of quenching the higher energy states of mercury mono-bromide, which lead to an additional population of its  $B^2\Sigma_{1/2}^+$  state [26]:

$$HgBr_{2} + e \rightarrow HgBr_{2}(D) \rightarrow HgBr(C^{2}\Pi_{1/2}, D^{2}\Pi_{3/2}) + SF_{6}$$
$$\rightarrow HgBr(B^{2}\Sigma_{1/2}^{+}) + SF_{6} + \Delta E, \qquad (1)$$

 $HgBr_2 + e \rightarrow HgBr_2(D) \rightarrow HgBr(C^2\Pi_{1/2}, D^2\Pi_{3/2}) + SF_6 + N_2$ 

$$\rightarrow \text{HgBr}(\text{B}^2\Sigma_{1/2}^+) + \text{SF}_6 + \Delta E, \qquad (2)$$

where  $\Delta E$  is the difference of excitation energy for states C, D, and B of mercury monobromide molecule [27, 28].

The time dependence of the pulsed power of HgBr-excilamp radiation is shown in Fig. 6 for the mixture HgBr<sub>2</sub>– SF<sub>6</sub>–N<sub>2</sub>–He, which is most optimal with respect to average power. Maxima of the radiation power in the time scale coincide (within the measurement accuracy of ~10%) with the current maxima (Fig. 3d), and the amplitude of third pulse is greater than that of first and second pulses.

The highest average radiation power of 48.8 mW was observed in the blue-green spectral range in the mixture  $HgBr_2-SF_6-N_2-He$  in the self-heating operation regime. The pulsed power  $P_p$  was 40.6 W and was determined from known average power  $P_{av}$ , pulse duration  $\Delta \tau$  and pulse repetition rate f using the expression

$$P_{\rm av} = P_{\rm p} f \Delta \tau.$$

The efficiency in this case was 7.3%.Under external heating of the mixture, the average power reaches a maximal value of 480 mW at the partial pressure of mercury dibromide vapours  $\sim$ 0.7 kPa.

Investigation results for the dependences of excilamp radiation power on the energy stored in the dielectric capacitance (7 pF, quartz glass), on the repetition rate of pump pulses,



Figure 5. Average power  $P_{\rm av}$  of HgBr-excilamp radiation vs. the partial pressure of helium in the mixture HgBr<sub>2</sub>-He (a), nitrogen in the mixture HgBr<sub>2</sub>-N<sub>2</sub>-He (b), sulfur hexafluoride in the mixture HgBr<sub>2</sub>-SF<sub>6</sub>-He (c), nitrogen in the mixture HgBr<sub>2</sub>-SF<sub>6</sub>-N<sub>2</sub>-He (d), mercury dibromide vapour in the mixtures HgBr<sub>2</sub>-N<sub>2</sub>-He (1), HgBr<sub>2</sub>-SF<sub>6</sub>-He (2), and HgBr<sub>2</sub>-SF<sub>6</sub>-N<sub>2</sub>-He (3) (e).



Figure 6. Time dependence of the pulsed power  $P_p$  of HgBr-excilamp radiation in the mixture HgBr<sub>2</sub>:SF<sub>6</sub>:N<sub>2</sub>:He = 0.1:0.07:4:117.2 at p = 191.3 kPa.

and duration of working mixture operation are presented in Fig. 7. In increasing the specific energy from 0.1 to 0.8 mJ cm<sup>-3</sup>, the average radiation power also increases (Fig. 7a). Its rate of growth was different, which is explained by different losses of the discharge power to elastic and inelastic processes occurring in plasma of various mixtures [24]. A maximal average power was obtained in the four-component mixture HgBr<sub>2</sub>- $SF_6-N_2$ -He. With increasing pulse repetition rate from 3 to 9 kHz, the average radiation power linearly increased (Fig. 7b). Such a type of the dependence is related with the linear growth of the number of photons which arrive at a photodetector with increasing pump pulse repetition rate. The growth of the average radiation power (Fig. 7c) at a greater number of pulses was more intensive for the mixtures HgBr<sub>2</sub>-SF<sub>6</sub>-He and HgBr<sub>2</sub>-SF<sub>6</sub>-N<sub>2</sub>-He [curves (1,2)] than for the mixture  $HgBr_2 - N_2 - He$  [curve (3)]. Such a behaviour is caused by the different rates of growth of HgBr<sub>2</sub> vapour concentration in mixtures with various compositions, which, in turn, depend on the part of power deposited to elastic processes of collisions of plasma electrons with respect to the power deposited to inelastic scattering of electrons on plasma components [24]. In the four-component composition HgBr<sub>2</sub>-SF<sub>6</sub>-N<sub>2</sub>-He, the rate of growth of mercury dibromide vapour concentration was minimal, which is related with higher discharge power losses to the inelastic processes with participation of sulfur hexafluoride and nitrogen. This resulted in a slower rise of the mixture temperature and, hence, slower elevation of the partial pressure of mercury dibromide vapour and finally, led to a slower increase in the radiation power with increasing number of pulses N up to  $1.5 \times 10^6$ . Starting with this value of N, the sharp increase in the rate of radiation power was observed, which was caused by the concurrent processes of the increase in the concentration of mercury dibromide vapour and the heat transfer to the internal surface of excilamp. In increasing the number of pulses, the concentration of mercury dibromide vapour raised faster than the heat losses through the dielectric (the excilamp case). At  $N = 1 \times 10^6$ ,  $1.5 \times 10^6$  and  $3 \times 10^6$ (Fig. 7c), the equilibrium was observed between the rates of rising concentration of mercury dibromide molecules and heat losses, which led to the saturation of the radiation power dependence on the number of pulses.

## 4. Conclusions

Investigations of spectral, integral and temporal characteristics of the radiation of the HgBr excilamp pumped by a nanosecond



**Figure 7.** Average power  $P_{av}$  of HgBr-excilamp radiation vs. the energy *W* stored in the dielectric capacitance (a), the pump pulse repetition rate *f* (b), and the number of pulses *N* (c) in the mixtures HgBr<sub>2</sub>-SF<sub>6</sub>-N<sub>2</sub>-He (1), HgBr<sub>2</sub>-SF<sub>6</sub>-He (2), and HgBr<sub>2</sub>-N<sub>2</sub>-He (3).

barrier discharge showed that its surface emits uniformly in the blue-green spectral range with a maximum at the wavelength of 502 nm. The highest values of average and pulsed power are observed in the four-component mixture HgBr<sub>2</sub>– SF<sub>6</sub>–N<sub>2</sub>–He at the total pressure of 121.4 kPa, which in the self-heating regime are 48.8 mW cm<sup>-3</sup> and 40.6 W cm<sup>-3</sup>, respectively, at the efficiency of 7.3%. The external heating of the lamp with this mixture composition increased the average radiation power to ~480 mW. Stable operation of the HgBrexcilamp was demonstrated at the pump pulse repetition rate of up to 9 kHz.

#### References

- 1. Kumagai H., Obara M. Appl. Phys. Lett., 54, 2619 (1989).
- 2. Kumagai H., Obara M. Appl. Phys. Lett., 55, 1583 (1989).
- Kumagai H., Toyoda K. *Appl. Phys. Lett.*, **59**, 2811 (1991).
  Vizir' V.A., Skakun V.S., Smorudov G.V., Sosnin E.A.,
- Tarasenko V.F., Fomin E.A., Chervyakov V.V. Kvantovaya Elektron., 22, 519 (1995) [Quantum Electron., 25, 494 (1995)].
- Furusawa H., Okada S., Obara M. Appl. Phys. Lett., 66, 1877 (1995).
- 6. Malinin A.N. Laser Phys., 7, 1032 (1997).
- Borisov V.M., Vodchits V.A., El'tsov A.V., Khristoforov O.V. Kvantovaya Elektron., 25, 308 (1998) [Quantum Electron., 28, 297 (1998)].
- Malinin A.N., Shimon L.L., Guivan N.N., Polyak A.V. Opt. Atmos. Okeana, 12, 1024 (1999) [Atmos. Ocean. Opt., 12, 977 (1999)].
- Malinin A.N., Polyak A.V., Guivan N.N., Zubrilin N.G., Shimon L.L. Kvantovaya Elektron., 32, 155 (2002) [Quantum Electron., 32, 155 (2002)].
- 10. Kogelschatz U. Plasma Chem. Plasma Process., 23, 1 (2003).
- 11. Malinin A.N. *Kvantovaya Elektron.*, **35**, 243 (2005) [*Quantum Electron.*, **35**, 243 (2005)].
- Malinin A.N., Shuaibov A.K., Shimon L.L., Grabovaya I.A., Polyak A.V. Prikl. Fiz., (1), 27 (2006).
- Guivan N.N., Malinin A.N. Opt. Spektrosk., 101, 399 (2006) [Opt. Spectrosc., 101, 375 (2006)].
- Guivan N.N., Malinin A.N. *Teplofiz. Vys. Temp.*, 44, 362 (2006).
  Posudin Yu.I. *Lazernava fotobiologiya* (Laser Photobiology)
- 15. Posudin Yu.I. *Lazernaya fotobiologiya* (Laser Photobiology) (Kiev: Vysshaya shkola, 1989) p. 248.
- Romanenko V.D. *Biotekhnologiya kul'tivirovaniya gydrobiontov* (Biotechnology of Production of Hydrobionts) (Kiev: Izd. Inst. Gydrobiologii NAN Ukrainy, 1999) p. 264.
   Sosnin E.A., Oppenlander T., Tarasenko V.F. *J. Photochem.*
- Sosnin E.A., Oppenlander T., Tarasenko V.F. J. Photochem Photobiol. C, 7, 145 (2006).
- Guivan M.M., Malinina A.A., Brablec A. J. Phys. D: Appl. Phys., 44, 1 (2011).
- Boichenko A.M., Lomaev M.I., Panchenko A.N. et al. Ul'trafioletovye i vakuumno-ul'trafioletovye eksilampy: fizika, tekhnika i primenenie (UV and VUV Excilamps: Physics, Technique and Application) (Tomsk: STT, 2011) p. 512.
- Efimov A.I., Belorukova L.P., Vasil'kova I.V., Chechev V.P. Svoistva neorganicheskikh soedinenii (Spravochnik) (Handbook of Properties of Inorganic Compounds) (Leningrad: Khimiya, 1983) p. 392.
- 21. Sapozhnikov R.A. *Teoreticheskaya fotometriya* (Theoretical Photometry) (Moscow: Energy, 1977) p.264.
- Akishev Yu.S., Dem'yanov A.V., Karal'nik V.B. et al. *Fiz. Plazmy*, 27, 176 (2001) [*Plasma Phys. Rep.*, 27, 164 (2001)].
- Pearse R.W., Gaydon A.G. *The Identification of Molecular Spectra* (London: Chapman & Hall, 1963) p. 347.
- 24. Raizer Yu.P. *Gas Discharge Physics* (Berlin: Springer, 1991; Moscow: Nauka, 1987).
- McDaniel E.W., Nighan W.L. Gas Lasers (New York: Acad. Press, 1962).
- 26. Nighan W.L., Brown R.T. J. Appl. Phys., 53, 7201 (1982).
- 27. Chang R.S.T., Burnham R. Appl. Phys. Lett., 36, 397 (1980).
- 28. Wadt W.R. Appl. Phys. Lett., 34, 658 (1979).