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# Excilamps: efficient sources of spontaneous UV and VUV radiation

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## Contents



**Abstract.** **The results of research into high-power, high-efficiency noble-gas-halide excilamps using glow, capacitive, and barrier discharges for the excitation sources are presented. The maximum radiation powers and minimum consumption are achieved with glow discharge lamps. An excilamp with an average radiation power of 1.6 kW on KrCl* molecules ($\lambda = 222$ nm) and 1.1 kW on XeCl* molecules ($\lambda = 308$ nm) is developed, whose energy conversion efficiency exceeds 10%. The use of an electrodeless capacitive discharge leads to sealed off excilamps with a simple emitter design, which have a power of 1 to 10 W and a service life of about 2500 h and more. Barrier-discharge excilamps possess both high energy parameters (> 100 W m$^{-1}$) and a long service life. Excilamps can find wide practical applications as new powerful sources of UV and VUV radiation.**

## 1. Introduction

Excilamps (UV and VUV radiation sources) make up a class of spontaneous radiation sources which have evolved relatively recently and utilize the nonequilibrium radiation of excimer and exciplex molecules [1 – 9]. A specific feature of these molecules is their stability in the excited electronic state and the absence of a strong bond in the ground state. Several of these molecules have an intense B-X transition in the UV and VUV spectral ranges, making it possible to efficiently convert energy deposited to the medium to optical radiation. The main distinction between the excilamps and available luminescent as well as thermal sources of UV and VUV radiation lies with the emission spectrum. Up to 80% of the total output radiation power and even more can be concentrated in a relatively narrow (no more than 10 nm at half height) band of the corresponding molecule. In this case, the radiation power density exceeds the values inherent in low-pressure lamps that utilize resonance atomic transitions. Furthermore, in the excitation of multicomponent gas mixtures it is possible to simultaneously obtain radiation of comparable intensity from two or more molecules [10 – 13].

The merits of excilamps from the standpoint of their application are a high photon energy (3.5 – 10 eV), a narrow emission band, a relatively high radiation power density, and the possibility of scaling and selecting an arbitrary geometry of the emitting surface. The absence of mercury in the excilamps is worthy of special mention. This gives them an edge in the competition with widely implemented but ecologically insecure mercury-filled lamps. At present, excilamps are coming into use in photochemistry and microelectronics; for cleansing and modifying surface properties; for the polymerization of lacquers and paints; in technologies for the disinfection of industrial waist, water, and air, and in biology, medicine, etc. This has become possible due to

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considerable steps forward in the understanding of the processes occurring in the optical media of excilamps and to the development of pilot samples suitable for practical implementation [14, 15].

In this work, we outline new results obtained in the investigation and design of high-power and efficient exciplex lamps. Primary emphasis has been placed on rare-gas-halide excilamps with glow-, capacitive-, and barrier-discharge excitation.

## 2. Optical media and optical transitions harnessed in excilamps

In excilamps, use is made of the working media containing rare gases as well as rare gases mixed with halogens. These media are distinguished by the character of energy relaxation over the electronic states of excimer and exciplex molecules produced in the medium during its excitation and by the relatively high energy of a photon emitted in the molecular transition to the ground state [2]. The presence of ionized and excited states coupled to each other by numerous intersections of potential energy curves has the effect that the relaxation of the medium involves a radiativeless sequential (upper-to-lower) excited-state population. The subsequent transition from the lower excited states (B, C, D in Fig. 1) of exciplex molecules to the ground state (the repulsive state A and the weakly bound or repulsive state X) is radiative, the energy level spacing between the lower excited and ground states being quite large. This accounts, first, for the high efficiency of conversion of the energy deposited to the medium to radiation and, second, for the presence in the emission spectrum of only the above-specified radiative transitions pertaining to the UV and VUV ranges. In this case, the B-X transition is highest in intensity. The spectrum may include weaker D-X, D-A, and C-A transition bands as well as emission bands of halogen molecules (Tables 1 and 2).

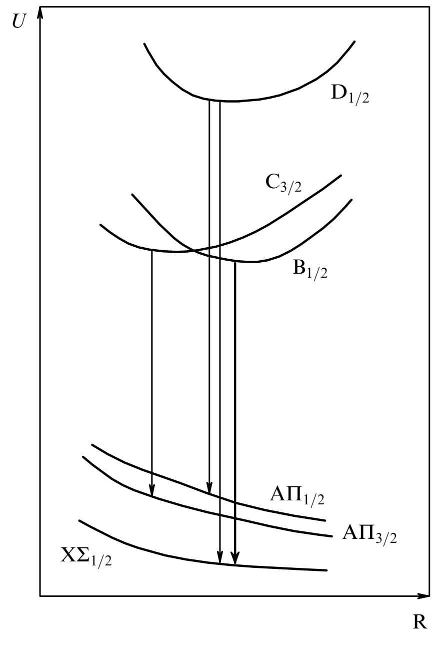


**Figure 1.** Lower terms of a molecule made up of a rare-gas atom and a halogen atom, and intense radiative transitions between them [2].

**Table 1.** Wavelengths of the principal radiative transitions in exciplex molecules [16].

| Rare-gas atom (R) | Halogen atom (Y) | Different transition wavelengths of the $RY^*$ molecule, nm | | | |
|---|---|---|---|---|---|
| | | $D_{1/2} \to X_{1/2}$ | $B_{1/2} \to X_{1/2}$ | $C_{3/2} \to A_{3/2}$ | $B_{1/2} \to A_{1/2}$ |
| Ne | F | 106 | 108 | 110 | 111 |
| Ar | F | 185 | 193 | 203 | 204 |
| Ar | Cl | | 175 | | 195 |
| Ar | Br | | 165 | 172 | 183 |
| Kr | F | 220 | 248 | 275 | 272 |
| Kr | Cl | 200 | 222 | 240 | 235 |
| Kr | Br | | 207 | 222 | 228 |
| Kr | I | | 190 | 195 | 225 |
| Xe | F | 264 | 351 | 460 | 410 |
| Xe | Cl | 236 | 308 | 345 | 340 |
| Xe | Br | 221 | 282 | 300 | 325 |
| Xe | I | 203 | 253 | 265 | 320 |

**Table 2.** Transition wavelengths of excimer and homonuclear halogen molecules [17].

| $R_2^*$, $Y_2^*$ | Wavelength, nm |
|---|---|
| $Ar_2^*$ | 126 |
| $Kr_2^*$ | 146 |
| $Xe_2^*$ | 172 |
| $F_2^*$ | 158 |
| $Cl_2^*$ | 259 |
| $Br_2^*$ | 289 |
| $I_2^*$ | 342 |

Excimer and exciplex molecules are produced in the working medium through different reaction channels [2]. Excimer molecules emerge by the association reaction

$$R^* + R + R \to R_2^* + R\,, \tag{1}$$

where $R^*$ and R are rare-gas atoms in the excited and ground states, respectively. The rate of reaction (1) is proportional to the square of the atomic number density in the ground state. This accounts for the strong pressure dependences of the radiation intensity and the shape of the rare-gas emission spectrum. Figure 2 shows the xenon emission spectrum excited by a barrier discharge at different pressures. The resonance line prevails at low pressure. As the pressure increases, one can observe two broad bands in the spectrum, which are conventionally termed the first and second

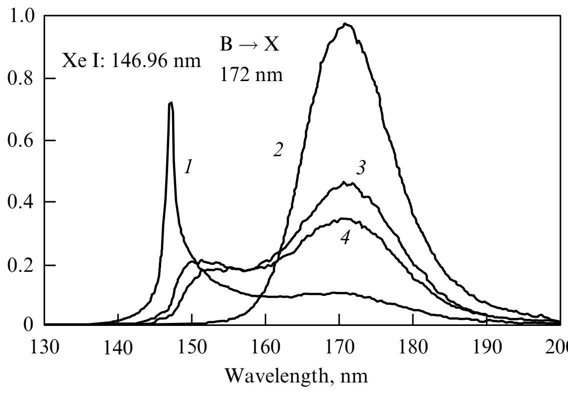


**Figure 2.** Xenon emission spectrum excited by a barrier discharge at different pressures [6]: 37 Torr (*1*), 510 Torr (*2*), 75 Torr (*3*), and 52 Torr (*4*).

continua. Grouped into the first are radiative transitions from high vibrational levels. This continuum is adjacent to the long-wavelength wing of the resonance line. The second continuum includes radiative transitions from the lower vibrational levels of the excited state and is accordingly located at the larger distance from the resonance line on the long-wavelength side. At a pressure of 100 Torr and above, the spectrum is dominated by the emission band of the second continuum.

Exciplex molecules are produced by the so-called harpoon reactions with participation of a rare-gas atom and a halogen-containing molecule:

$$R^* + TY(Y_2) \rightarrow RY^* + T(Y) \qquad (2)$$

or in the course of ion–ion recombination of a positive atomic or molecular rare-gas ion $(R^+, R_2^+)$ with a negative halogen ion $Y^-$:

$$R^+(R_2^+) + Y^- + M \rightarrow RY^* + M + (R)\,, \qquad (3)$$

where TY and $Y_2$ are halogen-containing molecules, Y is a halogen atom, and M is the third particle that carries away the excess energy. The function of particle M may be fulfilled by working or 'buffer' rare-gas atoms.

The most efficient optical media are xenon (the radiation efficiency $\eta$ of the $Xe_2^*$ molecules ranges up to 60%, the wavelength is $\lambda = 172$ nm [18, 19]), the mixtures Kr-$Cl_2$ and Xe-$Cl_2$ ($KrCl^*$ molecules, $\lambda = 222$ nm; $XeCl^*$, $\lambda = 308$ nm, $\eta \sim 25\%$ [16, 20, 21]), Xe-$Br_2$ ($XeBr^*$, $\lambda = 282$ nm, $\eta \sim 15\%$ [22, 23]), and Xe-$I_2$ ($XeI^*$, $\lambda = 253$ nm, $\eta \sim 20\%$ [24]). The radiation at these wavelengths is transmitted well by quartz, including the $\lambda = 172$ nm wavelength (the GE 021 SUPER type quartz), which allows us to design sealed-off radiators with a long service life and ensures their widespread use.

## 3. Types of discharges employed to excite excilamps

The working medium in excilamps can be excited in different ways. In stationary and quasi-stationary plasmadynamic light sources, special halogen-bearing additions are mixed with an equilibrium plasma flux to produce a plasma radiating on the transitions of exciplex molecules [25, 26]. In pulsed and pulse-periodic light sources, the excitation is effected by a broadband (thermal) plasma emission of a high-power discharge [27], an electron beam [8, 10], a discharge initiated or controlled by an external ionizer [28, 29], and also a self-sustained discharge [1, 4–6, 8, 9, 11–24, and so forth]. The last technique, the simplest and most accessible way, has received the widest acceptance, including use in the stationary excitation mode.

The operation of electric discharge excilamps relies on the excitation of the working medium contained in a quartz envelope of the lamp, achieved by passing an electric current. This is followed by the production of excimer or exciplex molecules in the discharge plasma, which radiate in the UV or VUV spectral range. The high radiation efficiency stems from the fact that the energy of the electric field is converted to optical radiation without significant losses by way of excitation and ionization of the particles, and the absorption by the components of the working medium and excited molecules and ions is of minor importance in spontaneous radiation sources as opposed to active laser media.

There are several types of self-sustained discharges employed to excite excilamps. Among these primarily are capacitive [30–32] and barrier discharges [1, 4–6, and others], a high-pressure UV-preionized volume discharge [33–35, and others], normal and subnormal low-pressure glow discharges [21, 36–43], distributed spark [44, 45] and microwave [46–49] discharges, and also a microchannel discharge [50, 51]. We note that the barrier discharge is a version of the capacitive discharge. For excilamps utilizing rare-gas–halogen mixtures as the working media, of significance from the viewpoint of service life is the point of whether the working medium is in contact with the metal electrode surfaces. The service life is limited owing to the electrode metal–halogen interaction resulting in the degradation of the working medium. To prolong the service life, as a rule advantage is taken of electrodeless discharges — capacitive, barrier, and microwave discharges. In this case, the working medium is enclosed in a quartz cavity whose shape depends on the type of discharge. The discharge plasma does not come into contact with the electrodes, which ensures a longer service life of the mixture. Potentially, there is also demand for an inductive discharge as one of electrodeless discharge types. At present, however, it is not widely used to excite excilamps because of its higher working voltage and a more complex design of the excitation generator.

Discharges with the presence of electrodes in the discharge region — glow, pulsed high-pressure UV-preionized volume, and spark discharges — are also employed for the excitation of excilamps despite the limitation of service life. Typical of the first discharge is the simplicity of the power supply [use is made of a direct or an alternating (industrial) current with a frequency of 50–60 Hz], the possibility of exciting large volumes of the working medium, and accordingly the feasibility of achieving high energy output parameters of the device. A volume UV-preionized discharge was found to be capable of exciting a medium at high pressure, which allows us to obtain the highest energy density parameters — the power density and the specific radiant energy.

## 4. Experimental equipment and techniques

Experiments were conducted on excilamps fabricated of quartz tubes (the GE 214 SUPER type quartz) with a wall thickness of 1.5–2.5 mm. Prior to being filled with the working medium, the radiators were annealed in a muffle furnace, evacuated, and passivated by the halogen employed subsequently. Figure 3 depicts the most commonly encountered designs of the radiators in capacitive-, glow-, and barrier-discharge excilamps. In the case of a capacitive discharge, the function of electrodes was fulfilled by metal cylinders located at the ends of the quartz tube and tightly adjoined its outer surface. As a rule, in the long-term excilamp operation recourse was made to forced-air cooling. The barrier-discharge excilamps of coaxial design were cooled by passing water through the inner tube. In this case, the electrodes resided on the external surface of the outer tube and on the interior surface of the inner tube. One or both electrodes were made of a metallic mesh to transmit light in the right direction. Barrier- and capacitive-discharge excilamps were excited by uni- or bipolar voltage pulses of variable duration (0.2–2 μs) with a repetition rate of 10–100 kHz. The generator power ranged from 10 to 1500 W. Glow-discharge excilamps could be coaxial [21] or cylindrical (see Fig. 3). An advantage of the former design is the

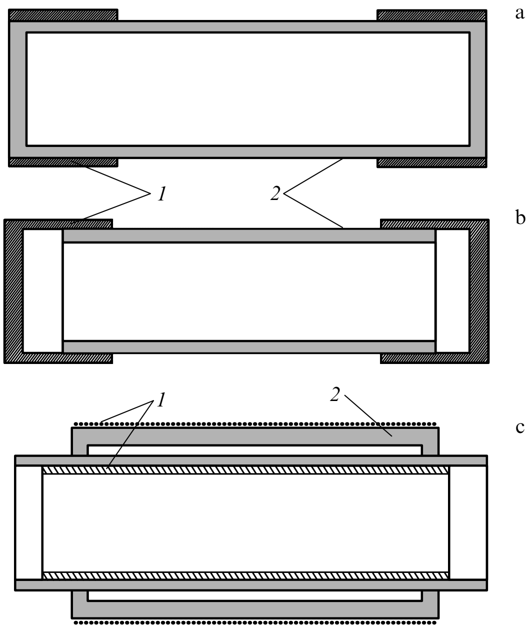


**Figure 3.** Design of capacitive- (a), glow- (b), and barrier- (c) discharge excilamps: electrodes (*1*), walls of lamp envelopes (*2*).\

possibility of efficient lamp cooling by the water flow through the inner tube. Meanwhile, the cylindrical lamp is the most simple in construction and allows an easy increase in structure length and, accordingly, in output radiation power. A glow-discharge excilamp of precisely this design was employed in our work to obtain a radiation power at a level of 1.5 kW. Air cooling was used for this excilamp. Replacing the air cooling with water cooling reduces the excilamp dimensions but involves a significant complication of the radiator design. The power supply comprised a step-up line-operated transformer with a power up to 7 kW and a voltage up to 15 kV, plus a voltage regulator. This radiator design and power supply are supposedly the most simple for attaining an average radiation power of 1 kW and over.

The radiation power was measured with a calibrated FEK-22 SPU photodetector by a traditional technique [52]. The current and voltage pulses as well as the time-resolved output radiation pulses were recorded with the aid of current shunts, voltage dividers, and an FEK-22 SPU photodetector, plus a TDS-224 oscilloscope, respectively. The discharge glow was detected with a digital camera. The working mixtures, which were made up of a working rare gas (krypton or xenon), a halogen ($Cl_2$, or $Br_2$, or $I_2$), and a buffer rare gas (helium or neon) in the case when it was used, were prepared immediately in the excilamp radiator or in a special mixer.

## 5. Determination of excitation parameters in excilamps

To get an adequate idea of the dynamics of different processes occurring in the gas-discharge plasma of the excilamps, one has to measure the pump power and the voltage across the discharge gap. When using a glow or pulsed volume discharge, this can be done in a relatively simple way — from oscilloscope traces of current and voltage. In this case, the voltage drop $U$ across the excilamp electrodes and the experimentally examined external current $I$ coincide respectively with the voltage $U_a$ across the discharge plasma and the conduction current $I_a$. However, during the excitation by electrodeless (barrier and capacitive) discharges, measuring the voltage across the electrodes and the external current would not suffice to determine $I_a$ and $U_a$. To reveal the optimal excitation conditions and the reason why the radiation efficiency depends on the excitation mode, it is therefore vital to determine the excitation parameters like the active constituents of current $I_a$ and voltage drop $U_a$ across the gas discharge gap, the instantaneous value of active power $P_a$, and the energy deposited to the system by the current point in time. Furthermore, estimates made for a barrier discharge show that the displacement current has a pronounced effect due to capacitance $C_g$ of a gas discharge gap. We note that the magnitude of $C_g$ may amount to as much as 10% of the dielectric barrier capacitances and more. In this case, the inductance of the gas discharge gap is of little significance in the case of both barrier and capacitive discharges. The algorithm for calculating the magnitudes of $P_a$, $U_a$, and $I_a$ for a barrier discharge is given in paper [53]:

$$P_a = I_a U_a \,. \tag{4}$$

The voltage $U_a$ can be found by the second Kirchhoff rule with the use of experimentally determined time dependences of the voltage $U$ across the discharge cell and the voltage drop $U_d$ across the dielectric capacitor:

$$U_a = U - U_d \,. \tag{5}$$

The voltage $U_d$ can be calculated knowing the charge $Q$ transferred and the capacitance $C_d$ of the dielectric barriers:

$$U_d = \frac{Q}{C_d} \,. \tag{6}$$

The charge transferred in the circuit can be determined by integrating the current subject to the initial conditions $Q(t=0) = Q_0$:

$$Q(t) = \int_0^t I(t')\,dt' + Q_0 \,. \tag{7}$$

The active current component $I_a$ can be obtained with the use of measured total current $I$:

$$I_a = I - C_g \frac{\partial U_a}{\partial t} \,. \tag{8}$$

The magnitudes of $C_d$ and $C_g$ are determined from the cell dimensions and the permittivity of the dielectric involved or from the slope of the corresponding portions of the volt–coulomb loops [54–56]. The energy $E(t)$ deposited to the plasma as a function of time is found by integrating expression (4):

$$E(t) = \int_0^t P(t')\,dt' \,. \tag{9}$$

For capacitive-discharge excilamps one can put $I_a \approx I$ owing to the smallness of $C_g$. The inclusion of the capacitance effect of the gas discharge gap is most pronounced under short-pulse excitation and makes it possible to obtain more comprehensive information about the parameters of an individual pulse and the average excitation power.

## 6. Excilamps with glow-discharge excitation

The classic glow discharge enjoys wide use in spontaneous radiation sources with low pressures of the working medium. In this case, the electrodes reside in the working volume. A line-operated transformer or a transformer and a rectifier can fulfil the function of a power supply. A voltage regulator is commonly placed ahead of the transformer in the experiment. The glow-discharge excilamp radiator consists of a quartz tube and electrodes located at its ends or of two coaxial quartz tubes, the inner one normally being cooled with water [40, 41]. Glow-discharge excilamps most easily allow an increase in the volume of the working medium and accordingly the average output radiation power.

### 6.1 Optimization of the composition and pressure of KrCl- and XeCl-excilamp mixtures

The pulsed glow discharge was first employed to excite excilamps in work [36]. In this case, the radiation efficiency did not exceed 1%. In Refs [37, 38], the radiation power was $\sim 8$ W for an efficiency up to $\sim 12\%$ under stationary glow-discharge excitation of Xe(Kr)-$Cl_2$ mixtures in a 28-cm long tube 14 mm in diameter. Two discharge stages were found to occur: the low-current stage characterized by current values ranging up to a few milliamperes, a high voltage, a weak uniform glow, and a radiation efficiency up to 10–15%, and the high-current stage, when the discharge passed, with increasing current, into a phase (normal glow discharge) with a significantly higher conductivity and radiation intensity. The characteristics of Xe(Kr)Cl excilamps excited by a low-current glow discharge were studied in greater detail in Ref. [43], and a radiation efficiency of 30% and a radiation power density of 50 mW $cm^{-3}$ were obtained. So high an efficiency is exhibited owing to a relatively low voltage drop across the cathode layer of the low-current glow discharge due to the high plasma resistance of the positive column. Furthermore, in a low-current glow discharge the photoemission induced by discharge radiation lowers the cathode drop. The low-current glow discharge is easier to obtain in low-diameter tubes or in coaxial radiators with a small spacing between the outer and inner tubes.

Unlike the low-current glow discharge which uniformly fills the entire tube surface, the high-current discharge (normal glow discharge) in optimal regimes occupies only a part of the tube cross section. The increase in total pressure or the content of a carrier of halogen has the effect that the discharge contracts into a narrow bright channel. Conversely, lowering the total pressure or the partial pressure of the halogen-bearer makes the discharge progressively more uniform and makes it occupy increasingly more space. The highest output radiation power was obtained (in relation to the excitation conditions) in the pressure range from a fraction of a torr to ten torrs using a mixture with a rare gas/halogen component pressure ratio between $1/3$ and $1/10$. The working mixture can also comprise a buffer gas, whose function can be fulfilled by light rare gases — helium or neon. This ensures, first, a lowering of the breakdown voltage and, accordingly, allows the use of power supplies with a lower output voltage. Second, raising the excitation power density in a ternary mixture improves efficiency and increases radiation power [57]. The highest radiation efficiency of exciplex molecules excited by a glow discharge is realized at a pressure of 0.5–2 Torr. Figure 4 shows the radiation efficiency of $XeCl^*$ molecules in a normal glow discharge as a function of excitation power. An estimate of excitation power density, whereby the efficiency attains its maximum, yields a value of $\sim 0.3$ W $(cm^3\ Torr)^{-1}$, which corresponds to $\sim 7 \times 10^{-18}$ W per particle. It is pertinent to note that the efficiency of about 25% is obtained only in the positive column of a high-current glow discharge without taking into account the voltage drop across the cathode layer, which may amount to half the total voltage across the excilamp electrodes [21]. In papers [21, 41, 42] we reported the development of coaxial and cylindrical KrCl and XeCl excilamps with an average output radiation power of 100–200 W in the UV spectral range, which were excited by a stationary glow discharge.

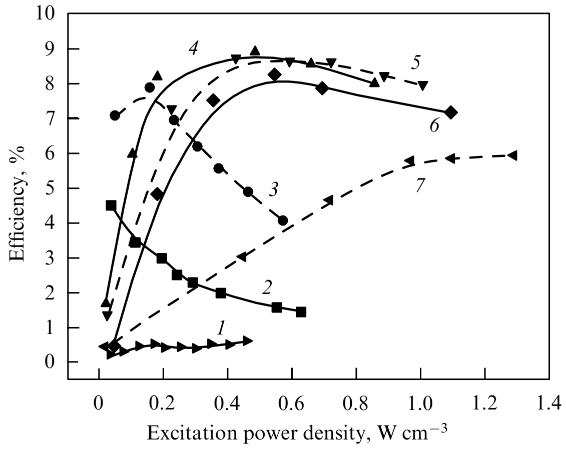


**Figure 4.** Radiation efficiency of $XeCl^*$ molecules as a function of excitation power for a $Xe/Cl_2 = 3/1$ mixture at pressures of less than 0.25 Torr (*1*), 0.25 Torr (*2*), 0.5 Torr (*3*), 1 Torr (*4*), 2 Torr (*5*), 4 Torr (*6*), and 6 Torr (*7*).

### 6.2 Efficiency of a glow-discharge excilamp

An analysis of the processes responsible for the production of exciplex molecules in a high-current glow discharge shows [42] that of the two possible channels of their production — ion–ion recombination and the harpoon reaction — the latter is the principal one [see reaction (2)]. The production rate, say, for the $XeCl^*$ molecules under conditions of the quasi-stationary regime for the working medium of a Xe-$Cl_2$ excilamp can be estimated as follows. The electron number density $N_e$ is found from the relation $j = -eN_e V_{dr}$ [30] using the experimental values of discharge current, the discharge channel diameter, and the voltage drop across the electrodes of the discharge tube. The electron drift velocity $V_{dr}$ is determined by the magnitude of the reduced electric field strength $E/N$ ($E$ is the electric field intensity, and $N$ the number density of the working mixture) at a given point in time [58]. For the electron number density we obtain $N_e \sim 10^{11}$ $cm^{-3}$. The concentration of metastable xenon atoms $N_{Xe^*}$ is estimated from the relation

$$N_{Xe^*} \sim \frac{K_{ex} N_e N_{Xe}}{K_h N_{Cl_2}} \sim 5 \times 10^{10}\ cm^{-3}\,, \tag{10}$$

where $N_{Xe}$ and $N_{Cl_2}$ are the xenon and chlorine concentrations, and $K_{ex}$ and $K_h$ are the rate constants of excitation and harpoon reactions, respectively. Under these conditions, the rate of $XeCl^*$ molecule production by the harpoon reaction is estimated as

$$\frac{\mathrm{d}[\mathrm{XeCl}^*]}{\mathrm{d}t} = K_{\mathrm{h}} N_{\mathrm{Xe}^*} N_{\mathrm{Cl}_2} \sim 10^{18}\ \mathrm{cm}^{-3}\,. \qquad (11)$$

On the other hand, the radiation power density in this case amounts to $\sim 1$ W cm$^{-3}$, which corresponds to a luminescence of $\sim 10^{18}$ molecules in 1 s per 1 cm$^3$ of plasma. This signifies that the rate of radiativeless decay of the XeCl* molecules in the glow-discharge plasma is low. Hence, the high radiation efficiency in these conditions can be attributed to the high efficiency of exciplex molecule production by the harpoon reaction and to the low rate of their radiativeless decay.

### 6.3 Glow-discharge excilamp with a radiation power of over 1.0 kW

The design simplicity of a glow-discharge excilamp and the power supply allow for easy scaling up of the device and obtaining a high output radiation power. We created a cylindrical excilamp with a radiation power of 1.6 kW utilizing KrCl* molecules, and 1.1 kW utilizing XeCl* molecules. The multisectional excilamp radiator comprised three paralleled branches made up of four separate sections connected in series (Fig. 5). The total branch length was $\sim 4$ m. The external diameter of the quartz tubes with a wall thickness of $\sim 2$ mm, used in the excilamp fabrication, was equal to 56 mm for two of the branches, and to 52 mm for the third one. The working mixture was prepared in a mixer and was then admitted to the lamp cavity at the requisite pressure. Three independent line-operated sources of alternating voltage regulated from 0 to 15 kV were utilized for the excitation. An investigation was made of the operation of individual sections, separate excilamp branches, and the excilamp as a whole. For tubes 52 – 56 mm in diameter, the optimal pressure for a voltage up to 15 kV was equal to 0.3 Torr. Figure 6 shows the dependences of output radiation power on the discharge current in the operation of each branch of the KrCl excilamp as well as the total radiation power. A higher output power was attained with the use of quartz tubes with an external diameter of 56 mm. In trials, a separate four-section branch yielded an average radiation power of 0.7 kW at a wavelength of $\sim 222$ nm for an efficiency of $\sim 12\%$ (the tube diameters were equal to 56 mm). On simultaneous actuation of all three branches, the average output radiation power was equal to 1.6 kW, and the efficiency to more than 10%.

Replacing krypton with xenon increased the breakdown voltage and the resistance of the gas discharge plasma, which lowered the average excitation power. This took place owing to a relatively low maximum voltage of the power supplies. That is why the average output radiation power with XeCl* molecules was lower than with KrCl* molecules and amounted to 1.1 kW, while the average output power of one branch with quartz tubes 56 mm in diameter was equal to 0.4 kW, and with quartz tubes 52 mm in diameter to 0.3 kW. The highest radiation efficiency was attained for relatively low discharge currents both in binary and ternary mixtures and was equal to $\sim 20\%$ for the KrCl excilamp, and to $\sim 15\%$ for the XeCl excilamp. The working mixture pressures of the given excilamp were lower than those employed in Refs [21, 42]. The reason lies with lower power-supply voltages per unit length of the radiator. At a higher discharge current density, the maximum average radiation powers with either of the molecules were attained when employing ternary mixtures with the addition of light rare gases [57]. The parameter values obtained for this radiator are not the limiting ones and can be improved by taking advantage of higher-power and higher-voltage power supplies, by increasing the working mixture pressure, and by cooling the quartz tubes and electrodes more intensely. Notice that the cathode and anode drops were recorded only at the ends of separate branches in extremely positioned sections, while in central sections observations were made of the positive column of the glow discharge.

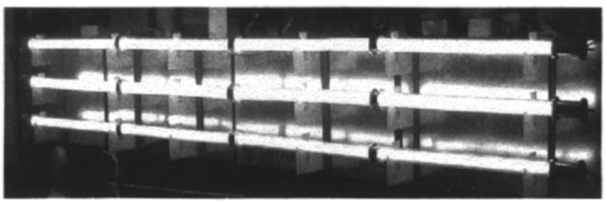

**Figure 5.** Exterior view of the glow-discharge excilamp with an output radiation power of 1.5 kW.

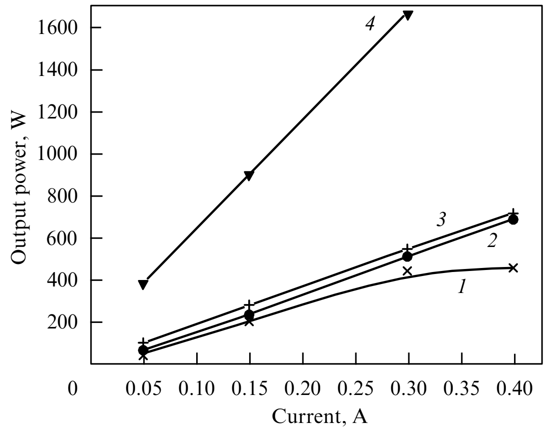


**Figure 6.** Dependences of the average output radiation power of KrCl* molecules on the discharge current for excilamp branches made of quartz tubes 52 mm (*1*) and 56 mm (*2*, *3*) in diameter, as well as on actuation of the entire radiator (*4*). A He/Kr/$Cl_2$ = 2/5/1 mixture at a pressure of 0.3 Torr is utilized.

Figure 7 gives the oscilloscope traces of the output radiation power, the current and voltage for one branch, and also the calculated excitation power curve. As already noted, the excitation was effected by an alternating voltage from a line-operated transformer. One can see that the radiation pulses correspond to current pulses in duration, are independent of the current pulse polarity, and practically replicate the temporal run of the excitation power curve. The emission spectrum of the excilamp is typical of excilamps excited by a glow discharge. The output radiation power is concentrated primarily in the B-X band (more than 70% of the output power).

## 7. Excilamps with capacitive-discharge excitation

In the general case, a high-frequency discharge of the capacitive type may involve electrodes or not, depending on whether the discharge plasma makes contact with the electrode surface or is separated from them by dielectric

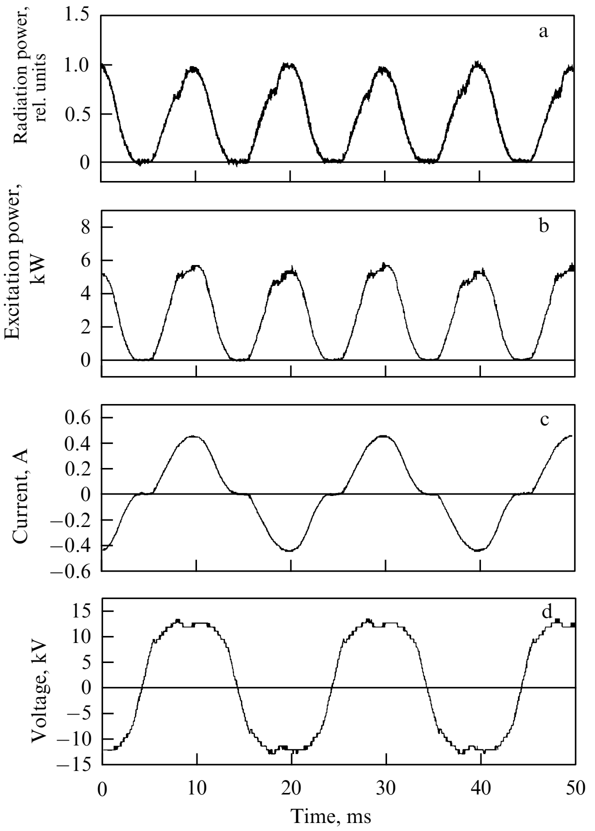


**Figure 7.** Typical oscilloscope traces of the output radiation power of $KrCl^*$ molecules (a), the discharge current (c), and the voltage (d); and the excitation power curve calculated for one branch of the excilamp (b). The mixture comprises $He/Kr/Cl_2 = 2/5/1$ at a pressure of 0.3 Torr.

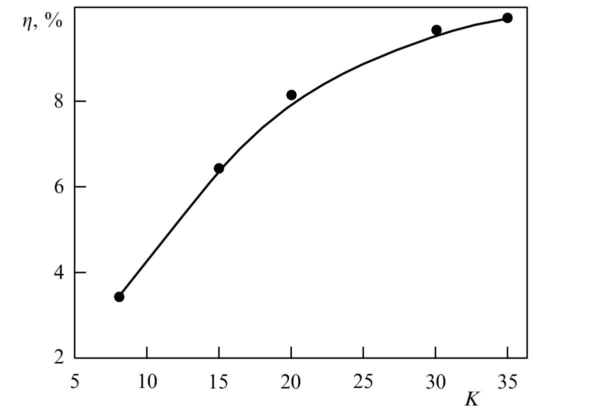


**Figure 8.** Radiation efficiency $\eta$ of a capacitive-discharge excilamp as a function of krypton-to-chlorine partial pressure ratio $K = p_{Kr}/p_{Cl_2}$. The total pressure mixture is 7 Torr, and the maximum output radiation power is 7 W.

layers. From the standpoint of the application of this discharge type to the excitation of excilamps, it is expedient to resort to an electrodeless capacitive discharge, thereby ensuring a longer service life of the working medium in this case. The main features which distinguish the capacitive discharge from the barrier one (the excilamps with barrier-discharge excitation are discussed in the next section) are a relatively low pressure of the working mixture (no higher than tens of torrs) and a long electrode spacing (tens of centimeters). The electrodeless capacitive discharge was first employed to excite excilamps in work [32]. An immanent feature of electrodeless capacitive and barrier discharges is the existence of a dielectric barrier and, hence, the limitation of the energy deposited to the plasma during an individual excitation pulse. The equivalent electrical circuits of the electrodeless capacitive and barrier discharges are similar, with the exception of the fact that in the latter case, as noted above, a significant part in the flow of current may be played by the capacitance $C_g$ of the electrode gap. However, the characteristics of the plasma in capacitive-discharge excilamps are closer to those in glow-discharge excilamps. Therefore, several properties typical of both barrier- and glow-discharge excilamps are inherent in excilamps with capacitive-discharge excitation.

### 7.1 Optimization of the working mixtures of XeCl, XeBr, and KrCl excilamps

For capacitive-discharge excilamps, the working medium was optimized for Kr(Xe)-$Cl_2$ and Xe-$Br_2$ binary mixtures. For all the above mixtures, the total optimal pressure did not exceed $\sim$ 15 Torr for a typical electrode spacing of $\sim$ 20 cm and a rare gas/halogen component pressure ratio between 7/1 and 35/1. In reducing the pressure and/or further lowering the halogen content, the discharge became progressively more uniform (diffusive). However, the radiation power decreases in this case. Figure 8 shows the radiation efficiency of the KrCl excilamp at a total pressure of 7 Torr and an output radiation power up to 7 W as a function of Kr-to-$Cl_2$ partial pressure ratio. One can see from this figure that the optimal content of $Cl_2$ in the mixture is equal to $\sim$ 3%. Further lowering of the $Cl_2$ fraction does not entail a substantial improvement of the radiation efficiency. Furthermore, in this case the service life of the working mixture is significantly shortened. That is why in the fabrication of sealed-off excilamps it is more expedient to utilize mixtures with a chlorine concentration exceeding the optimal one by 10–20%. We note that small (below 30%) additions of the He(Ne) buffer gas improved the output power and the radiation efficiency, as in the case of glow-discharge excilamps.

### 7.2 Effect of the excitation mode

The excitation mode, which is defined by the build-up and fall-off rates of the voltage across the device electrodes where discharges with dielectric-barrier current limitation are involved, affects not only the radiation power and efficiency, but the visual appearance of the discharge as well. When a capacitive-discharge excilamp was excited by a generator with steep leading and trailing pulse edges (no longer than 200–250 ns), there formed a narrow, brightly glowing current channel. The excilamp radiation efficiency and output power were below the maximum values. The best results were obtained when using bipolar excitation pulses with leading- and trailing-edge times of $\sim$ 1–1.5 μs and a repetition rate up to $\sim$ 100 kHz. In this case, the current channel became diffusive and expanded to a characteristic diameter of $\sim$ 1–1.5 cm. In outward appearance it resembled the positive column of a glow discharge. As in the case of barrier and glow discharges, the radiation efficiency depended on specific excitation power to attain a value of $\sim$ 15% for $XeBr^*$ molecules.

Figure 9 shows typical oscilloscope traces of the radiation pulses, the current, the voltage across the electrodes (as well as

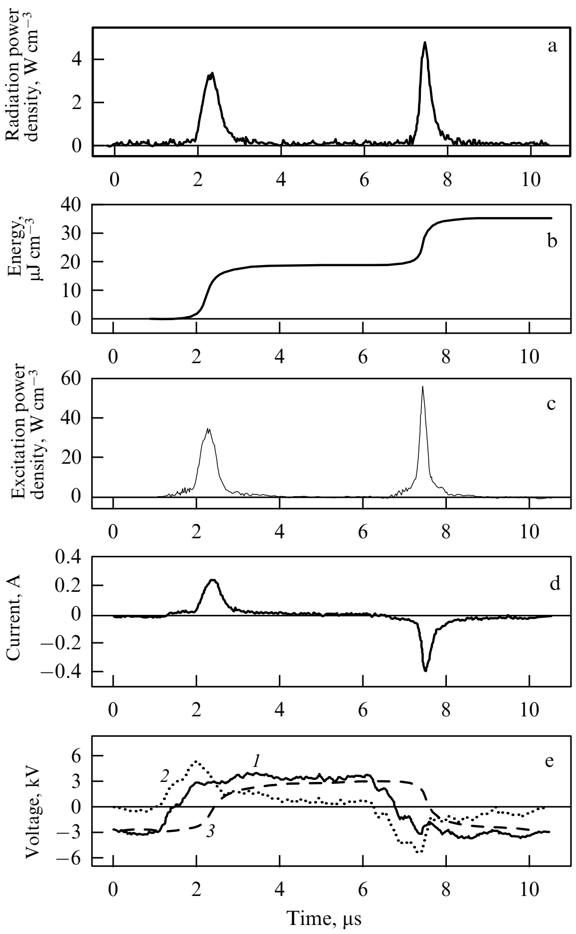


**Figure 9.** Oscilloscope traces of output radiation power density ($\lambda = 282$ nm) (a), the current (d), the voltage drop (e) (*1* — across the electrodes, *2* — as calculated across the gas discharge gap, *3* — the same across the capacitor formed by the dielectric barriers), the excitation power (c) and energy (b) for a capacitive-discharge XeBr excilamp. Use is made of an $Xe/Br_2 = 3/1$ working mixture at a total pressure of 3.5 Torr.

the calculated curves of the voltage across the gas discharge gap and the capacitor formed by the dielectric barriers), and also the power and energy of excitation for the XeBr excilamp. For excitation purposes, advantage was taken of a bipolar pulse generator with a repetition rate of $\sim 100$ kHz and with leading- and trailing-edge times of the voltage pulses of $\sim 1$ μs. Referring to this figure, the radiation pulse correlates with the excitation power curve and the radiation efficiency is approximately the same for positive and negative semiperiods of the excitation pulse. The plasma resistance during the current flow lies in the range between 10 and 100 kΩ, which is close to the plasma resistance in glow-discharge excilamps and is significantly higher than in the case of barrier-discharge excitation. For moderate dimensions of the electrodes and a long electrode spacing, the capacitance of the excilamp interelectrode gap does not exceed $\sim 1.5$ pF. Accordingly, the magnitude of a total current recorded is practically equal to the active constituent of the current traversing the gas discharge gap.

An estimate of the electron concentration $N_e$ in the discharge plasma column by the relation $j = -eN_e V_{dr}$ with neglect of the ion current in comparison with the electron one yields the following result. If the current density $j$ is approximately 0.3 A cm$^{-2}$, and the drift velocity $V_{dr} \sim 7 \times 10^6$ cm s$^{-1}$, the electron number density $N_e$ is equal to about $2.7 \times 10^{11}$ cm$^{-3}$, close to observations in the glow discharge conditions.

## 8. Excilamps with barrier-discharge excitation

A discharge wherein the current flow is limited by at least one dielectric layer and the characteristic electrode dimensions far exceed the electrode spacing is referred to as a barrier discharge. This predetermines the necessity of employing a power supply (generator) with a time-variable voltage $U$: on the strength of the law of conservation of the sum of conduction and displacement currents throughout the whole circuit, the conduction current $I_a$ carried by the gas discharge plasma is defined by the displacement current $I_{dis,b}$ through the dielectric barrier and we have

$$I_a = I_{dis,b} - I_{dis,g}\,, \tag{12}$$

where $I_{dis,g}$ is the displacement current through the capacitor formed by the gas discharge gap. The magnitude of $I_{dis,b}$ is in turn proportional to the rate of change of the voltage across the dielectric barrier:

$$I_{dis,b} = \frac{\partial C_d U}{\partial t} = C_d \frac{\partial U}{\partial t} + U \frac{\partial C_d}{\partial t}\,. \tag{13}$$

The authors of Ref. [4] were among the first to take advantage of the barrier discharge to produce rare-gas radiation continua. Remarkable progress in the investigation and production of $Xe_2$ excilamps was made in works [18, 19, 59–61, and some others]. Xenon barrier excilamps underlie the development of a whole family of spontaneous radiation sources in the VUV spectral range as well as mercury-free linear and plane luminescent lamps.

A barrier-discharge plasma exhibits several characteristic properties, namely:

— a high pressure (several hundred torrs and higher);

— spatial nonuniformity and short duration of different physicochemical processes. Along with the volume discharge, randomly located microdischarges (filaments) are, as a rule, observed in the plasma, wherein the duration of the current pulse does not exceed 10–50 ns;

— a large magnitude of departure from equilibrium plasma state. In this case, the average electron temperatures may range up to several electron-volts, while the gas temperature remains close to the temperature of dielectric barriers;

— the possibility of varying the reduced electric field strength $E/p$ ($E$ is the electric field strength, and $p$ is the pressure) in the plasma by changing the external parameters like the supply voltage, the pressure of the working medium, and the thickness of the gas discharge gap (as a rule, this thickness does not exceed several-to-tens of millimeters);

— the possibility of scaling and predefining arbitrary geometry for the lamp radiating surface.

All this, along with the absence of contact between the working medium and the metal surface and, accordingly, a longer service life of the gaseous mixture, predetermined the widespread use of the barrier discharge for the excitation of excilamps.

One of the parameters which affect the operation efficiency and service life of the excilamp is its temperature mode. Numerous experiments suggest that the increase in temperature of the lamp envelope is responsible for the reduction of both its efficiency and service life. This generates a need for forced cooling of the envelope. Quite often used in practice is one- or two-sided water cooling. In this case, for KrCl and XeCl excilamps it is possible to raise the time- and volume-averaged excitation power density to approximately 1 W $cm^{-3}$ for an output radiation power density up to 0.1 W $cm^{-3}$.

### 8.1 Effect of excitation pulse shape

Generators with a sinusoidal voltage pulse shape are traditionally employed as excitation sources for barrier-discharge excilamps. In this case, typical efficiencies of conversion of the power added to the working medium to optical radiation amount to 10–15% [5, 16]. Noted in paper [62] earlier was the advantage of using a sine-shaped voltage pulse over a short, high-voltage, 50–100-ns long pulse for the excitation of $Xe_2$, $Kr_2$, KrCl, and XeCl excilamps by a barrier discharge. The main cause of the low radiation efficiency when effecting the excitation with short high-voltage pulses produced in a TGI 10000/25-thyratron-based generator is, first, the lowering of the fraction of energy deposited to the gas discharge plasma in comparison with that stored in the energy storage device. Second, in this case there occur significant overvoltages across the gas discharge gap, resulting in values of reduced electric field strength $E/p$ in the gas discharge plasma, which are nonoptimal from the standpoint of production of exciplex molecules. Meanwhile, an increase in energy conversion efficiency of a barrier-discharge $Xe_2$ excilamp under short-pulse (voltage pulse rise time $\sim$ 250–750 ns) excitation in comparison with sine-shaped pulse excitation was demonstrated recently in Refs [18, 19]. In the view of the authors of Ref. [18], the observed rise in radiation efficiency is due to the formation of the optimal electron energy distribution function and the minimization of elastic and inelastic electron energy losses in the processes which do not result in the production of $Xe_2^*$ excimer molecules. In Ref. [19], it is noted that improving the efficiency of the $Xe_2$ excilamp necessitates producing a uniform (diffusive) discharge with an electron density significantly lower than that characteristic of filaments. The discharge uniformity in this case is ensured due to the steep edge of the voltage pulse.

Since exciplex and excimer molecules are produced in dissimilar ways, it is conceivable that the conditions corresponding to the attainment of highest radiation efficiencies of, for instance, $Xe_2$ and KrCl excilamps may be different.

To verify the effect of excitation pulse shape on the excilamp parameters, use was made of two generators, one of them providing sine-shaped voltage with a frequency of 17 kHz and the other delivering uni- or bipolar voltage pulses with a total pulse duration of 2 μs, leading- and trailing-edge times of about 250 ns or $\sim$ 1 μs for a pulse repetition rate of 10–100 kHz. The pressure and composition of the mixture containing Kr and $Cl_2$ was preliminarily optimized in the experiments. The best results were obtained with the $Kr/Cl_2 = 200/(1-0.5)$ mixture for a total pressure of approximately 200 Torr. As the total pressure or the $Cl_2$ content in the mixture are lowered, a progressively more uniform volume discharge is observed, but the output radiation power diminishes in this case. Increasing the pressure or the $Cl_2$ content in the mixture results in discharge contraction, with a consequential reduction in radiation power.

Figure 10 shows the lamp operation efficiency and the output radiation power density as functions of specific excitation power for the optimal working mixture and sine-shaped excitation pulses. The absolute values of the efficiency sought and the pattern of its dependence on the excitation power agree well with the data of Zhang and Boyd [16]. A rather uniform discharge with diffuse filaments is visible during lamp operation. Figure 11 depicts the oscilloscope traces of the current pulses, the voltage across the lamp electrodes, the pulse of radiation on the B-X transition of the KrCl* molecule, the calculated excitation power curves,

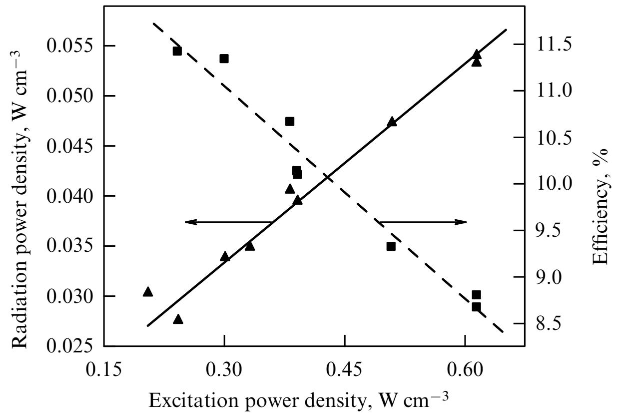


**Figure 10.** Operation efficiency of a barrier-discharge excilamp and output radiation power density as functions of excitation power density for a sinusoidal shape of excitation pulses.

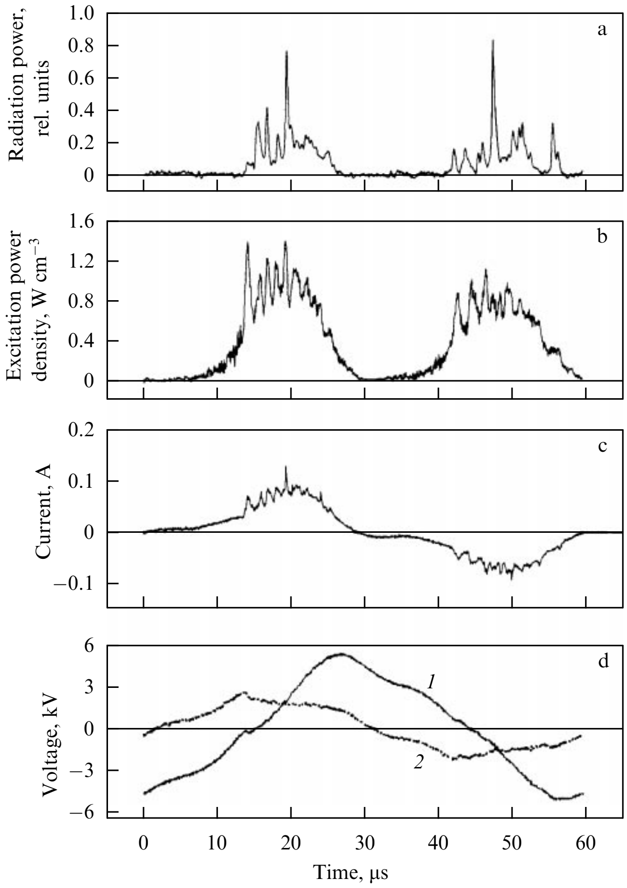


**Figure 11.** Oscilloscope traces of the radiation power (a) and current (c) pulses, the voltage across the electrodes of a barrier-discharge excilamp (d, curve *1*), calculated curves of excitation power density (b), and voltage drop across the gas discharge gap (d, curve *2*) for a sinusoidal shape of excitation pulses.

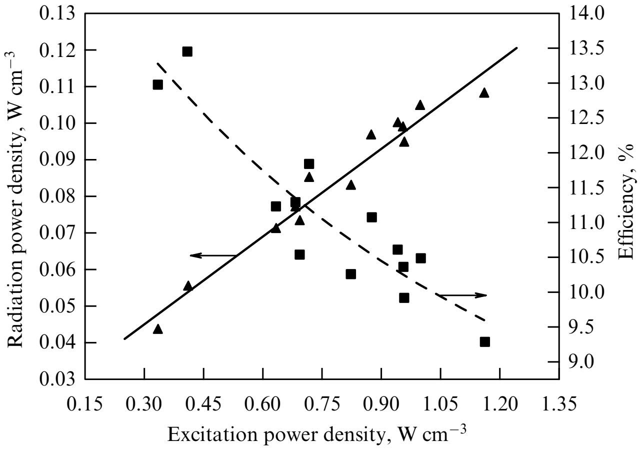


**Figure 12.** Lamp operation efficiency and radiation power density as functions of excitation power density for a frequency of 93 kHz and a unipolar shape of excitation pulses.

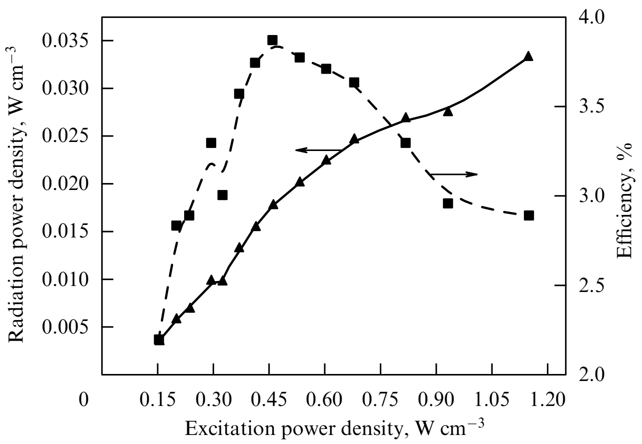


**Figure 13.** Efficiency and radiation power density as functions of excitation power density in a discharge void of visually detected filaments under short-pulse excitation ($Kr/Cl_2 = 200/0.5$ mixture under a total pressure of 100 Torr).

and the calculated voltage drop across the gas discharge gap that correspond to the conditions exhibited in Fig. 10. One can see that the emission takes place during most of the active phase of discharge (after a sharp decrease in the voltage across the discharge gap). Also noteworthy are the modulation of radiation intensity, the radiation pulse delay ($\sim 2-2.5$ μs) relative to the onset of the excitation pulse, and the radiation – excitation power correlation at later points in time. Characteristically, the voltage drop across the discharge plasma is relatively constant.

When using uni- and bipolar excitation pulses with a total pulse duration of 2 μs for repetition rates of 17, 33, 60, and 93 kHz, a more nonuniform discharge glow with filaments clearly manifested in the glow intensity is visually observed. Figure 12 illustrates the radiation efficiency and the output radiation power density as functions of excitation power density for unipolar excitation pulses at a repetition rate of 93 kHz. For the other repetition rates and bipolar excitation pulses, the data are close to those given in Fig. 12. It is noteworthy that the energy density characteristics were determined taking into account the entire discharge volume, though it is evident that both the excitation and the emission exhibit a strong spatial nonuniformity in the presence of clearly defined filaments. That is why the above characteristics in the presence of filaments should be treated as though averaged over the discharge volume. Comparing Figs 10 and 12 allows the conclusions that, first, increasing the input power in both cases results in a lowering of radiation efficiency, which may be attributed to the overexcitation and heating of the medium. Second, short-pulse excitation is an advantage over the other excitation modes (by $\sim 20\%$ for similar levels of excitation power density).

### 8.2 Effect of mixture pressure

Gas pressure is one of the major factors determining the nature of discharge. As noted above, a progressively more uniform discharge is observed with lowering the mixture pressure. To verify the possible effect of the degree of discharge homogeneity on the radiation efficiency of the KrCl excilamp, an experiment was conducted in the absence of visually detected filaments in the gas discharge plasma, which was realized in a $Kr/Cl_2 = 200/0.5$ mixture at a total pressure of 100 Torr under short-pulse excitation. The test data are given in Fig. 13. The main distinction from the previously discussed data is, first, the nonmonotonic run of the radiation efficiency curve with a peak for an excitation power density of about 0.45 W cm$^{-3}$. Second, the peak efficiency in this case proves to be $2-3$ times lower than those obtained under the conditions of Fig. 12 for the same excitation power densities. Furthermore, a radiation pulse delay ($\sim 0.5$ μs) relative to the onset of the excitation pulse is observed in the absence of filaments, like in the case of excitation by sine-shaped pulses. On the other hand, in the presence of filaments this delay is practically absent. The principal cause of this distinction is supposedly due to the high excitation power density in the volume occupied by the filaments. The difference may be as high as two orders of magnitude and more, since the volume occupied by the filaments is estimated to be smaller, by the same factor, than the total gas discharge volume taken into account in the determination of excitation power density. Comparing the reduced electric field strength $E/p$, the excitation power per unit volume and per particle, and the radiation efficiency in the conditions of the experiment conducted and of the glow discharge [41] suggests the following conclusions. First, in the case of comparable efficiencies of about 10% in the glow discharge and the barrier discharge with the presence of filaments, there occur close values of $E/p \sim 10$ V (cm Torr)$^{-1}$ and excitation power densities of about tens-to-hundreds of W cm$^{-3}$ per unit volume and of the order of $(1-50) \times 10^{-17}$ W per particle. In this case, the volume occupied by the filaments is taken into account for a barrier discharge. Second, for a significantly lower efficiency ($2-4\%$) pertinent to the homogeneous barrier discharge without visually identified filaments, the excitation power densities are equal to $\sim 10-15$ W cm$^{-3}$ and $\sim (2-20) \times 10^{-18}$ W per particle. The reduced field strength $E/p$ in this case amounts to $\geqslant 15$ V (cm Torr)$^{-1}$.

The amount of energy $W_{\mathrm{vol}}$ deposited to a unit volume of the barrier-discharge plasma in one period is defined by the specific capacitance $C_{\mathrm{d}}$ of lamp dielectric layers (the typical values are $C_{\mathrm{d}} \sim 1-1.5$ pF cm$^{-2}$), the discharge gap $d$, and the amplitude $U_{\max}$ of the supply voltage:

$$W_{\mathrm{vol}} \leqslant \frac{C_{\mathrm{d}} U_{\max}^2}{d} \, . \tag{14}$$

If $U_{max} \sim 5$ kV and $d \sim 5$ mm, then $W_{vol} \approx 20-40$ μJ cm$^{-3}$. When the duration of current flow is about 50 ns, the average power during an individual excitation pulse is of order 400–800 W cm$^{-3}$ under conditions when the deposited energy is uniformly distributed over the volume. However, in the presence of filaments, which is characteristic of the barrier discharge, the energy is deposited primarily to the volume occupied by the filaments. Accordingly, the excitation power density in the filament region may be as high as several-to-tens of kilowatts per cubic centimeter. We believe that in the volume occupied by filaments there occur excitation conditions close to those which take place under excitation by a volume transverse discharge.

The amount of energy deposited to a unit volume of the barrier-discharge plasma in one period, including the homogeneous discharge mode, can be increased by raising the supply voltage and employing a dielectric material with a high permittivity value. In doing this, however, there emerge evident technical complications, like the inadequate electric strength of the dielectric material, difficulties associated with high-voltage operation, etc.

Therefore, it is believed that the presence of filaments — the spatial regions with a high excitation power density — in the barrier discharge is the necessary condition for the attainment of a high-efficiency operation of a barrier-discharge KrCl excilamp. The uniform volume distribution of the same input excitation power under conditions of a homogeneous discharge results in a reduction of radiation efficiency.

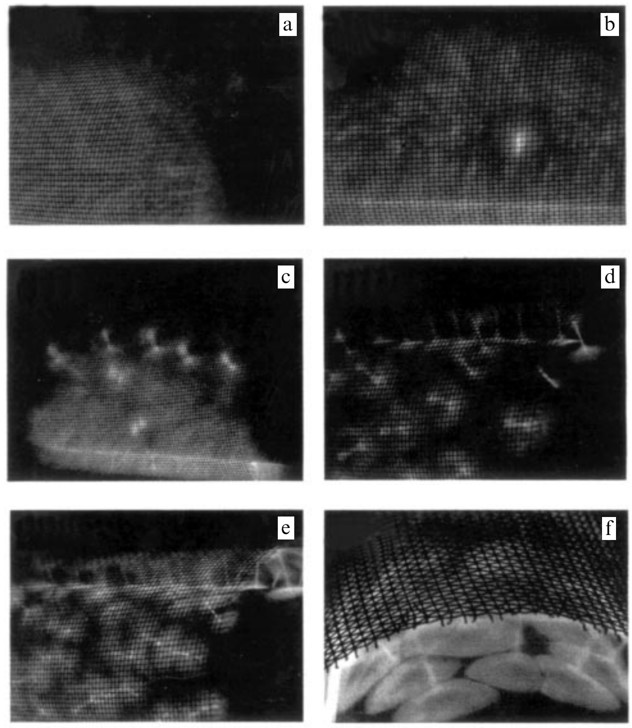


**Figure 14.** Exterior view of the excilamp barrier discharge for excitation pulse repetition rates of 500 Hz (a), 1.1 kHz (b), 1.3 kHz (c), 5 kHz (d), 20 kHz (e), and 200 kHz (f).

### 8.3 Effect of excitation pulse repetition rate on the radiation efficiency and formation of discharge

Among the factors which affect the conditions of producing an individual filament, the discharge as a whole, and the radiation efficiency and output power of an excilamp is the pulse repetition rate $\nu$. To elucidate the effect of this factor, the unipolar-pulse repetition rate was varied from 10 Hz to 200 kHz in the excitation of an XeCl excilamp. Figure 14 displays the photographs of discharge in the excilamp for different $\nu$. One can see from this figure that clearly defined filaments emerge in the discharge plasma at an excitation pulse repetition rate $\nu_0 \sim 1$ kHz. For $\nu < \nu_0$, diffuse channels with a relatively low glow brightness and random distribution over the volume (frame a in Fig. 14) are observable in the gas discharge plasma. For $\nu \geqslant \nu_0$, the filament density, the filament foot dimensions and their brightness build up with increasing $\nu$. In this case, the excitation power and, accordingly, the output radiation power increase. An interesting feature is the absence of overlapping of the filament feet, which is most pronounced for $\nu = 200$ kHz. The effect of residual electron concentration is supposedly the cause of formation of a brightly glowing individual filament for $\nu_0 \sim 1$ kHz, which normally stays put (frame b in Fig. 14). At this frequency and with a temporal interval of about 1 ms between the excitation pulses, the residual electron concentration in the filament region is high enough for the next gas gap breakdown followed by filament production to occur again in this region.

The change of exterior appearance, the filament density, and the glow intensity of an individual filament, as well as the variation of excitation power density under variations of $\nu$, can affect the excilamp operation efficiency. Figure 15 shows the excitation power, output radiation power, and radiation efficiency of the XeCl excilamp as functions of $\nu$. An

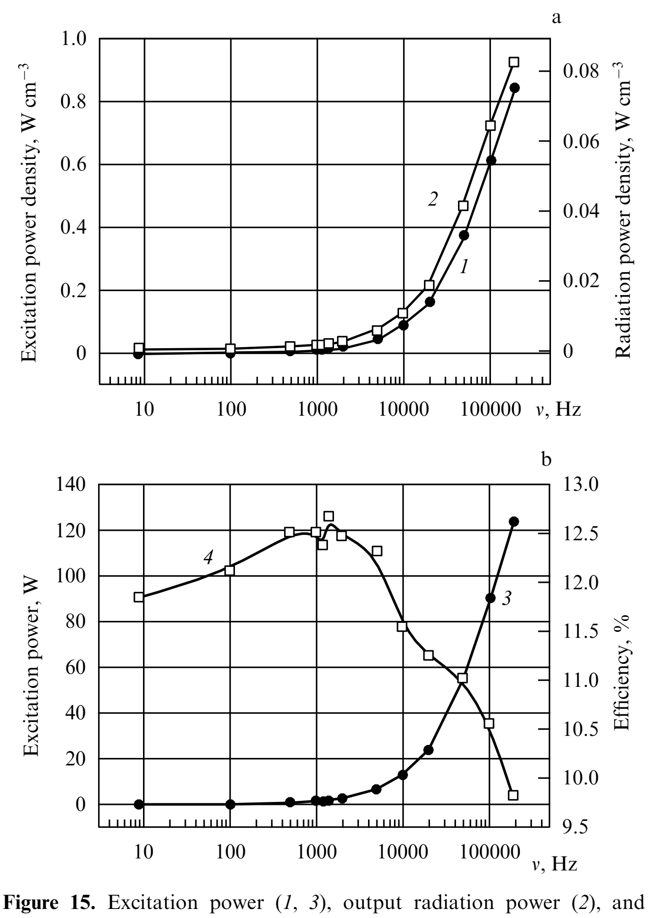


**Figure 15.** Excitation power (*1*, *3*), output radiation power (*2*), and radiation efficiency (*4*) as functions of the excitation pulse repetition rate $\nu$ for a barrier-discharge XeCl excilamp.

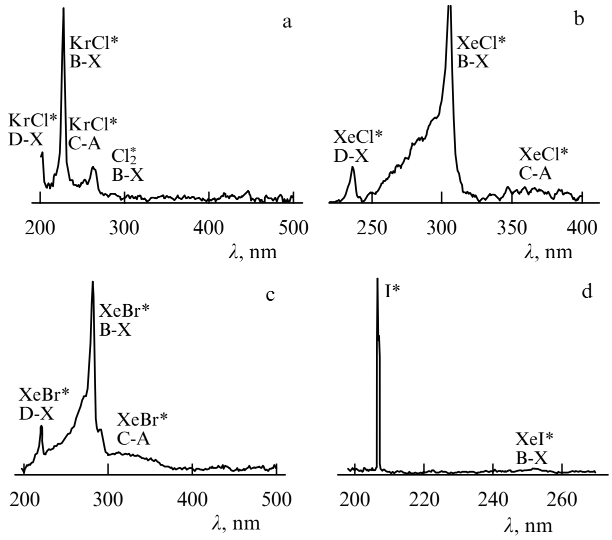


**Figure 16.** Emission spectra of capacitive-discharge excilamps for the following mixtures: Kr-$Cl_2$ at a pressure of 6 Torr (a), Xe-$Cl_2$ at a pressure of 6 Torr (b), Xe-$Br_2$ at a pressure of 4.4 Torr (c), and Xe-$I_2$ at a pressure of 1.5 Torr (d).

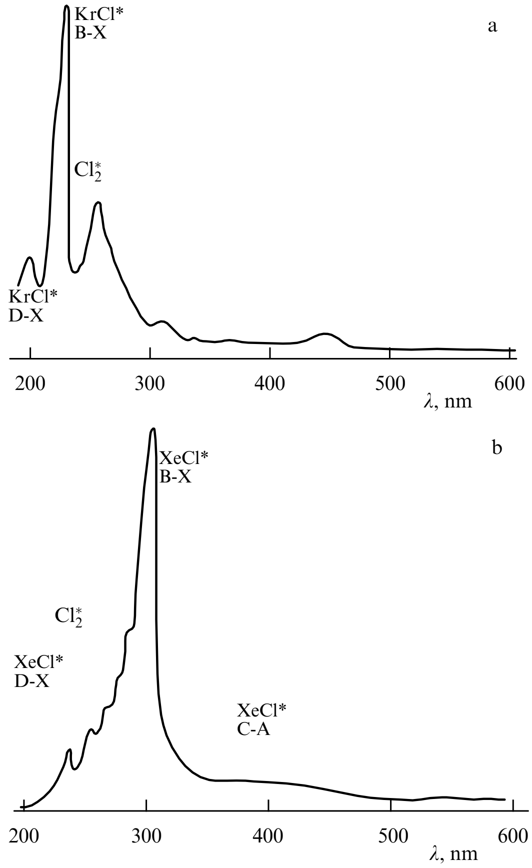


**Figure 17.** Emission spectra of glow-discharge excilamps for a Kr-$Cl_2$ mixture at a pressure of 6 Torr (a), and a Xe-$Cl_2$ mixture at a pressure of 6 Torr (b).

important fact is the retention of radiation efficiency for $v < v_0$ at a level close to the highest one. A discharge with diffuse channels randomly located in the discharge gap is observed in this range of excitation pulse repetition rates (see Fig. 14). It is characteristic that no filament feet are visually observed. This may be an indication of the fact that the excitation level required for the efficient production of exciplex molecules may be attainable both in the case of formation of clearly defined filaments and in a discharge with diffuse channels at low excitation pulse repetition rates. However, in the latter case the average output radiation power of an excilamp is low.

## 9. Emission spectra of excilamps

As noted above, along with the service life and the energy parameters the emission spectrum is one of the key characteristics of a radiation source. An immanent feature of the excilamps from this viewpoint is the presence of only separate narrow UV or VUV bands in the emission spectrum, which belong to the corresponding molecules. Figures 16–18 show the emission spectra of capacitive-, glow-, and barrier-discharge excilamps under the conditions specified in Table 3. Referring to Fig. 16, when the excitation is effected by a capacitive discharge, the spectral halfwidth of the most intense B-X transition in KrCl* ($\lambda \sim 222$ nm), XeCl* ($\lambda \sim 308$ nm), XeBr* ($\lambda \sim 282$ nm), and XeI* ($\lambda \sim 206$ nm) is $\sim 4$, 5, 8, and 7 nm, respectively. Characteristic of the spectra under these conditions is, first, the presence of clearly defined D-X and C-A transitions of the above molecules. Second, a broad and intense blue wing of the B-X transition band is observed in the case of XeBr* and XeCl* molecules. In the XeI-excilamp spectrum at a low mixture pressure (1.5 Torr and less), the atomic iodine $\lambda = 206$ nm line proved to be highest in intensity (Fig. 16d). In this case, the intensity of the B-X transition band of the XeI* molecule ($\lambda \sim 253$ nm) is significantly lower. As the pressure increases, the intensity ratio between the indicated line and band changes, and at a pressure of 10 Torr they become comparable in amplitude.

Figure 17 gives the KrCl- and XeCl-excilamp emission spectra under glow-discharge excitation. A comparison of Figs 16 and 17 allows us to draw the conclusion that the spectral distribution of radiation is hardly different in both cases.

A characteristic feature of excilamps excited by a barrier discharge is a short gap and relatively high pressures of the

**Table 3.** Characteristics of capacitive-, barrier-, and glow-discharge excilamps for which spectra were taken.

| Characteristic | Discharge type | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | Capacitive | | | | Barrier | | Glow | |
| Working mixture | Xe-$Cl_2$ | Kr-$Cl_2$ | Xe-$Br_2$ | Xe-$I_2$ | Xe-$Cl_2$ | Kr-$Cl_2$ | Xe-$Cl_2$ | Kr-$Cl_2$ |
| Working mixture pressure, Torr | 6 | 6 | 4.5 | 1.5 | 120 | 200 | 6–9 | 6–9 |
| Interelectrode gap, cm | 20 | 20 | 20 | 50 | 0.8 | 0.8 | 20–80 | 20–80 |
| External tube diameter, cm | 3.8 | 4.2 | 4.2 | 3.5 | 6.5 | 6.5 | 1–6 | 1–6 |
| Wavelength of spectral distribution peak, nm | 308 | 222 | 282 | 206 | 308 | 222 | 308 | 222 |

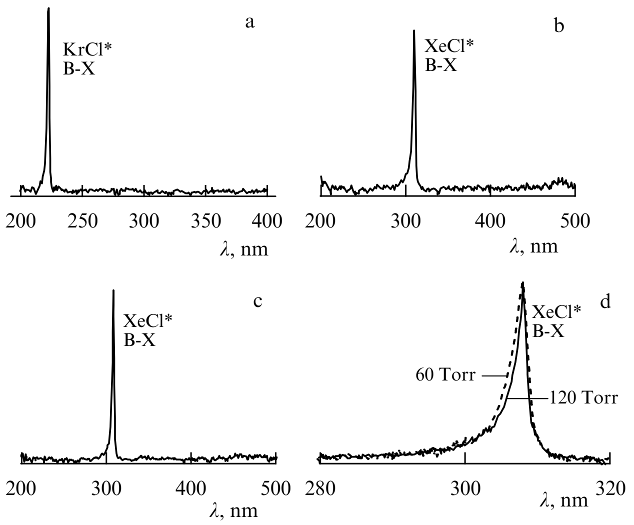


**Figure 18.** Emission spectra of the barrier-discharge KrCl excilamp for a mixture pressure of 200 Torr (a), and the barrier-discharge XeCl excilamp for a mixture pressure of 60 Torr (b) and 120 Torr (c). The spectra (b) and (c) in the vicinity of the peak are superposed in the last figure (d).

working medium. Figure 18 depicts the KrCl- and XeCl-excilamp emission spectra taken under barrier-discharge excitation. One can see from this figure that only the B-X transition bands are observed in the emission spectrum under these conditions. The spectral halfwidth of the B-X transition in $KrCl^*$ molecules is about 2 nm. The D-X and C-A transition bands of the $KrCl^*$ molecule, as well as the $Cl_2^*$ molecular band, which were present in the emission spectrum obtained under capacitive- and glow-discharge excitations, are practically missing from the barrier-discharge spectrum. The emission-spectrum stability was checked for sealed-off capacitive-discharge excilamps, whose service life exceeds 1000 h [32]. The spectral distributions of the KrCl excilamp after 250- and 850-h operation were found to be hardly different.

## 10. Lifetime of the working mixture and service life of excilamps

Service life is among the critical performance parameters of radiation sources, including excilamps. The major factor which limits the duration of UV exciplex lamp operation is the degradation of the working medium due to the chemical reaction of a halogen with the electrodes and/or with the quartz of the lamp envelope. The reaction rates rise with temperature of the working medium, and therefore advantage is taken of air or water cooling with increasing excitation power density. The service life of the remaining elements ordinarily ranges into the tens – hundreds of thousands of hours.

The shortest service life of the working mixture, which may be as short as several minutes, is exhibited by glow-discharge excilamps. This is their main drawback. The limited service life of glow-discharge excilamps is due to exposure of the metal electrodes to the plasma. But the service life can be prolonged if advantage is taken of electrodes made of weakly corroding materials, a buffer volume, and continuous circulation and regeneration of the working medium, which complicates the design and raises the cost of the excilamp. Recourse to a more intense cooling

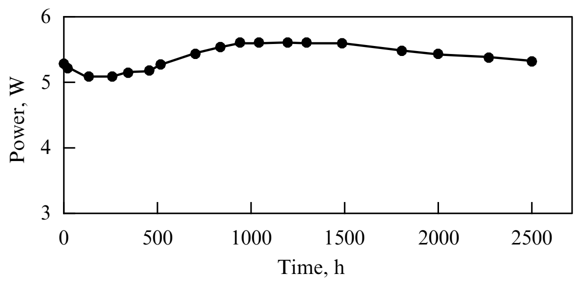


**Figure 19.** Average power of a capacitive-discharge KrCl excilamp as a function of operation time.

can also reduce the rate of output power decay in the course of operation.

In excilamps with barrier-discharge excitation, the plasma does not usually make contact with the metal electrodes, which facilitates the development of sealed-off excilamps and prolongs the service life of the working mixture. To prolong the mixture service life, advantage is taken of deep degassing on heating in a muffle furnace with subsequent passivation of the interior lamp-envelope surface by the halogen to be used. This technology was employed to fabricate a KrCl excilamp with the following dimensions: a length of 60 cm, an outer diameter of 65 mm, and a discharge gap thickness of 10 mm. The lamp was cooled by water flow through the internal cavity of the inner tube and also by external air ventilation. The excilamp excitation was effected with a generator of bipolar voltage pulses with a total duration of about 2 μs, a repetition rate of 93 kHz, and a power up to 1.5 kW. The highest output radiation power of the excilamp was $\sim$ 115 W, and the output radiation power densities were at a level of about 100 mW $cm^{-2}$ and 0.2 W $cm^{-3}$. The radiation efficiency in this mode amounted to $\sim$ 15%. In 100 h, the output radiation power of the excilamp fell off by less than 10%.

Excilamps excited by an electrodeless capacitive discharge also offer certain advantages over barrier-discharge excilamps, since the excitation power density and, accordingly, the thermal load in this case are lower, thus resulting in a longer service life of the radiator. Figure 19 shows the life test data for a KrCl excilamp. The pressure and the mixture component ratio were 11 Torr and 5/1, respectively. In this case, the average output radiation power was no less than 5 W, and the radiation efficiency measured approximately 7%. The service life of this radiator exceeded 2500 h, which is acceptable for many applications.

## 11. Specific features of excilamps which distinguish them from other UV and VUV radiation sources

Excilamps can be compared with other radiation sources as to several physical, technical, and operating parameters. These are the emission spectrum and its stability; the energy characteristics — the total and specific radiation powers; the character of operation; the service life; the fields of application; the typical dimensions of the emitting surface, which affect the conditions and possibilities of radiation transfer; the design and technological features; the cost, etc. We make a comparison as regards the critical parameters from among these.

Apart from excilamps, a wide diversity of spontaneous and stimulated (lasers) radiation sources is presently available

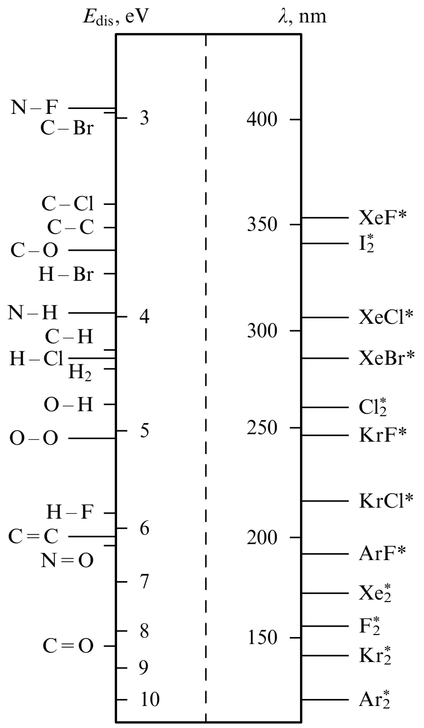


**Figure 20.** Dissociation energies of several chemical bonds and B-X transition wavelengths of excimer and exciplex molecules, as well as halogen dimers.

in the UV and VUV spectral ranges. Employed most extensively are mercury (high- and low-pressure, with and without additions of other gases), hydrogen, deuterium, metal-halide, and xenon (pulsed and continuously operated) lamps. Among UV and VUV lasers, mention should be made primarily of exciplex and excimer lasers — presently the highest powered and most efficient pulsed sources of stimulated radiation in the above-mentioned wavelength ranges. Furthermore, also available are nitrogen ($N_2$), helium – cadmium (He-Cd), fluorine ($F_2$), argon ion (Ar II), and other lasers.

The employment of stimulated radiation sources is favored in those practical applications which necessitate a high radiation power density (from several kilowatts to several gigawatts per square centimeter) or a high spatial resolution during irradiation (of the order of the laser radiation wavelength) [63]. As a rule, the laser irradiation of a material at treatment is local (ablation, cutting, welding, strengthening, marking of articles, etc.). UV and VUV lasers are used to advantage to deposit thin films in the production of microelectronic components, in the processing of semiconductor materials, including UV and VUV microlithography, in medicine, and in numerous other applications [63 – 65].

However, many applications require large volumes or large-area surfaces to be uniformly irradiated with a rather high power density in the UV or VUV spectral range. This provides a means for the excitation or breaking of chemical bonds and ensures the initiation of photoinduced chemical reactions that are requisite for a specific application. Figure 20 depicts the dissociation energies of several chemical bonds and the corresponding (as regards the photon energy) B-X transition wavelengths of excimer and exciplex molecules, as well as halogen dimers. These requirements are largely met by excilamps, since they afford a higher level of radiation power in the above wavelength range in comparison with, for instance, mercury and xenon arc continuous or pulsed lamps, as well as deuterium lamps, which exhibit a continuous or line spectrum. Furthermore, in some cases excilamps maximize the efficiency of a photochemical process owing to the coincidence of their narrow-band emission spectrum with the absorption spectrum of the material under irradiation.

At present, excilamps enjoy wide use as linear fluorescent lamps in copiers and scanners; the radiation of a microcell barrier discharge in rare-gas mixtures has found application to the fabrication of plasma displays. Moreover, excilamps are used validly in the technologies of photoinduced surface cleaning, thin-film deposition, and the fabrication of coatings with the requisite properties. Owing to the high excilamp-radiation power density (from 1 to 50 mW $cm^{-2}$), the rates of photochemical processes in this case prove to be higher than with the use of, for instance, low-pressure mercury lamps. It is also significant that these processes take place at relatively low temperatures (250 – 400 °C) and are not attended with factors responsible for defects and damage, which ensures high optical and electrical parameters of the films and coatings being formed [66 – 73].

Excilamps as narrow-band UV radiation sources offer a significant advantage over traditional UV radiation sources (medium- and high-pressure mercury lamps) in the cure of thermosensitive films and coatings [66, 67]. In the event when a lamp has a broadband emission spectrum, only a minor part of the radiation is utilized to entail solidification. The remaining part of the radiation is wasted on the useless heating of the surface under irradiation. In this case, the cure effect shows itself under substantially different levels of radiation energy density (accordingly several millijoules or joules per square centimeter), depending on whether use is made of excilamps or conventional broadband radiation sources. Furthermore, in excilamps, unlike in many other lamps where the working media are steam – gas mixtures, the operating mode sets in immediately after switching on, and the start-up and radiative excilamp characteristics are noncritical to the operation temperature.

It is also possible to point to other fields which have seen the pioneering application of excilamps: photobiology, photomedicine, photochemistry of solutions, decomposition of deleterious waste in solid and liquid phases, disinfection and purification of drinking water, and so forth. For instance, a recent paper [74] reports on comparison studies of the photolysis of aqueous solutions of phenol and *n*-chlorophenol under KrCl-laser and KrCl-excilamp excitations. It was shown that the photo-induced transformations proceeded much more effectively when the solutions were exposed to the 222-nm excilamp radiation with a pulse duration of $\sim$ 1 μs in comparison with exciplex laser irradiation at the same wavelength and a pulse duration of 10 ns.

During irradiation the light energy goes not only into the excitation of chemical bonds, but also into the production of radicals and bond breakage, which was used in Ref. [75] for the oxidation of solutions containing toxic organic compounds. A xenon barrier-discharge excimer lamp ($\lambda \sim 172$ nm) was employed for the mineralization of phenol and alcohol in a photochemical reactor in aqueous solutions. In combination with distillation, purified water was obtained, and it was suggested that the process itself be termed photoreactive distillation.

At present UV disinfection systems primarily employ mercury lamps. It is not improbable that mercury-free excilamps will replace them in the future. The first experience of employing XeBr, XeCl, and KrCl capacitive-discharge excilamps for disinfection is reported in Refs [76, 77]. The merits of excilamps as relatively safe narrow-band UV radiation sources may also be used for the purpose of UV photohemotherapy and photoimmunotherapy, in the development and employment of new photosensitizers for the therapy of skin diseases, and in other fields of medicine [78].

One characteristic feature of the excilamps that affects the operating conditions and the selection of the fields of application are the large dimensions and volume of the gas discharge plasma which is the radiation source. For several applications this is an excilamp virtue, for instance, where uniform illumination of large areas or volumes is required. Meanwhile, technical problems arising in the transfer of excilamp radiation over a distance are also evident, as they are in focusing or concentrating the radiation on a surface or a volume element. In this respect, excilamps are radically different from lamps with small dimensions of an emitting volume, as well as from lasers whose radiation is much easier to transfer over a distance and to concentrate. To concentrate the light flux of the excilamps whose design is diagrammed in Fig. 3, there is good reason to employ a specular parabolic–cylindrical reflector [79] for an individual excilamp or their combination for several excilamps. Furthermore, there exist barrier-discharge excilamp designs with a flat emitting surface, which are expedient from the viewpoint of obtaining and employing a uniform plane radiation flux [70]. Two of them are depicted in Fig. 21. However, the coaxial design (Fig. 3c) is most frequently employed, which makes it possible to obtain an axially symmetric radiation flux directed outwards or inwards when using an inner or outer specular electrode. The outer and/or inner tubes can be cooled by liquid or air flow. The solution under irradiation can also serve as the cooling liquid.

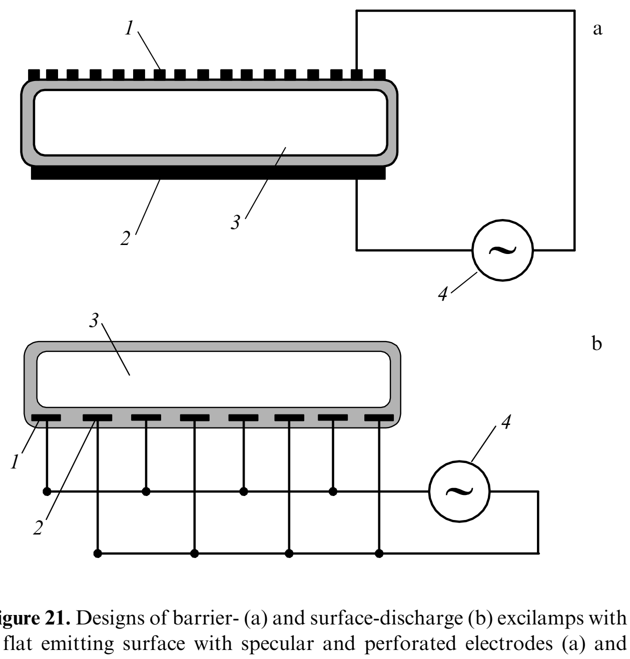


**Figure 21.** Designs of barrier- (a) and surface-discharge (b) excilamps with a flat emitting surface with specular and perforated electrodes (a) and sectionalized electrodes located in one plane (b): electrodes (*1*, *2*), gas discharge volume (*3*), and power supply (*4*).

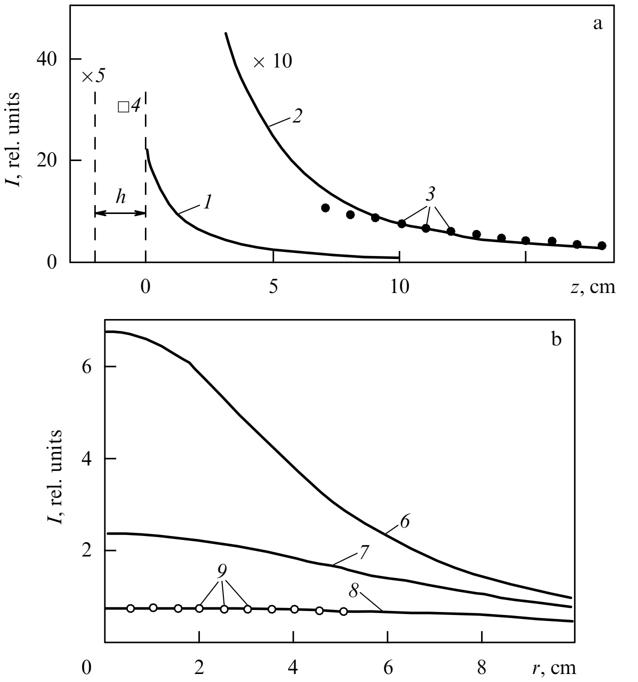


**Figure 22.** Spatial intensity distribution of excilamp radiation on the cylinder axis $z$ (a) and along the radius $r$ in the plane perpendicular to the $z$-axis (b):calculation (*1*, *2*, *4–8*), experiment (*3*, *9*); the experimental and calculated values for curves *2* and *3* were taken to be equal for $z = 10$ cm; intensities on the cylinder axis at the center of the radiating layer for $h = 2$ cm (the point □) and 4 cm (the point ×) (*4*, *5*); radial intensity distributions for $z = 2$ cm (curve *6*), 5 cm (*7*), and 10 cm (*8*, *9*) (curves *1* and *2* differ in scale).

Boichenko et al. [8, 80, 81] were concerned with the theoretical and experimental investigation of the spatial characteristics of the light beam emanating from cylindrical and planar excilamps. In both cases, the intensity was observed to decline fast with increasing distance to the radiating surface. The radiation intensity distribution on the $z$-axis of a planar excilamp and along the radius $r$ in the plane perpendicular to the $z$-axis is plotted in Fig. 22 [80]. The radiating domain had the shape of a cylinder with a height $h = 2$ cm and a radius $R = 4$ cm. To improve the radial uniformity of the power density, the distance from the radiating surface to the surface being irradiated should be increased. However, the power density declines rapidly in this case. For a planar excilamp, the thickness of the plasma layer should be selected so that it is approximately equal to the distance the exit excilamp window plane from the surface under irradiation.

## 12. Conclusions

The foregoing suggests that excilamps nowadays are the most efficient narrow-band spontaneous radiation sources in the UV and VUV spectral ranges. The emission spectrum of exciplex molecules consists primarily of a single intense B-X transition band. These sources will find wide practical application, from replacing traditionally employed mercury lamps to harnessing the selective absorption of narrow-band UV and VUV radiation.

The highest average output radiation powers at the lowest consumptions are attained with the use of glow-discharge excitation. We have created an excilamp with an average output radiation power of 1.6 kW utilizing $KrCl^*$ molecules, and a $XeCl^*$-molecule excilamp with an average output power of 1.1 kW for a radiation efficiency above 10%.

The employment of a capacitive electrodeless discharge makes it possible to construct excilamp radiators of simple design with an output radiation power of 1 – 10 W at wavelengths of $\sim 222$ nm ($KrCl^*$), $\sim 308$ nm ($XeCl^*$), $\sim 282$ nm ($XeBr^*$), $\sim 253$ nm ($XeI^*$), and $\sim 206$ nm ($XeI^*$) for a radiation efficiency of $\sim 10\%$ and higher. In this case, the longest service life of the working mixture is attained with sealed-off models (over 2500 h). The emission spectrum of glow- and capacitive-discharge excilamps comprises, apart from the B-X transition band with a halfwidth of no more than 10 nm, the D-X and C-A transition bands, as well as the bands of halogen molecules.

Excilamps excited with a barrier discharge exhibit both high energy parameters (over 100 W per 1 m of length) and a long service life. We have developed a 60-cm long sealed-off KrCl excilamp with an external diameter of 65 mm and an average output radiation power of 116 W. The radiation of exciplex molecules under barrier-discharge excitation is concentrated primarily in the B-X transition band and has a minimal half width of $\sim 2$ nm.

Iodine-based lamps hold considerable promise for practical use, making it possible to obtain high-power radiation at a wavelength of 206 nm, radiation simultaneously at 206 and 253 nm, and also radiation in the VUV spectral range.

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