

Optimization of gas mixtures of multiband excilamps

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The paper considers the problem of optimizing gas-discharge sources of multiband UV radiation — excilamps. A method is proposed for estimating the ratio of partial pressures of the components of a gas mixture, which makes it possible to simplify the process of developing a multiband excilamp with the desired power ratio of the emission bands. It is shown that the optimization of a multiband excilamp on a mixture of one noble gas with several halogen carriers can be reduced to the problem of optimizing an excilamp on a two-component mixture. If, in addition to the radiation of exciplex molecules, the radiation of excimer molecules is of importance, then the method makes it possible to control the relative contribution of the powers emitted by these molecules.

Keywords: gas discharge, sources of ultraviolet radiation, kinetics of plasma-chemical processes, excimer and exciplex molecules.

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Introduction

There is a wide variety of radiation sources [1] operating in different wavelength ranges and characterized by different operating principles and output parameters [1–21]. Among them, sources of ultraviolet (UV) and vacuum ultraviolet (VUV) radiation [1,6,9,10,12,14–18,21–40] are widely demanded in various areas of science, industry and consumption. Excilamps are becoming of special demand [1,6,9,10,12,14–18,22–25,27–32,34–36,41–43]. For example, excilamps based on KrCl* molecules provide better, compared to conventional low-pressure mercury lamps, inactivation of pathogens of viral sources [44,45].

Excilamps are sources of relatively narrow-band UV and VUV radiation. The emission half-maximum bandwidth of different types of exciplex molecules ranges from 2 to 15 nm, while for excimer molecules it can reach up to 30 nm. However, in practice, sometimes there is a need for a broadband or multiwave radiation source with a special spectral composition. Excilamps can also be used as such sources, since they are characterized by the possibility of simultaneous excitation of several types of exciplex molecules [46–50]. By choosing exciplex molecules emitting in the desired spectral region [51], one can obtain a radiation source with a spectral characteristic that is a combination of the spectral characteristics of the corresponding exciplex molecules. The most difficult task that has to be solved in the course of developing a multiband source of UV radiation with the desired spectral composition is to ensure the necessary ratio of the radiation powers of each band. When developing a multiband excilamp based on a multicomponent mixture, it is necessary to find the optimal

operating conditions for the excilamp. This is essentially an optimization problem for a function of many variables. In practice, its solution turns out to be a very costly and labor-consuming process.

There are quite a lot of various effective methods for optimizing a function of many variables (see, e.g., [52–54]), among them the method of steepest descent [55], the method of conjugate directions [56], etc., allowing quick determination of the extremum of the objective function. However, in practice, there are difficulties with the use of such methods, since the existing methods do not take into account the specifics of the operation of the experimental setup and, therefore, may require changing the conditions in a sequence that is very inconvenient from a practical point of view, which, in turn, can lead to such undesirable effects as, for example, excessive consumption of working gases. There are a number of other difficulties that significantly limit the use of existing mathematical methods for optimizing a function of many variables in practice. So, for each individual case, it may be necessary to develop its own optimization technique, taking into account the specifics of the operation of the experimental setup.

In practice, when optimizing multicomponent mixtures, the method of cyclic coordinate-wise descent, which is not very effective from a mathematical point of view, is very popular. The essence of the method is sequential one-dimensional optimization for each of the variables, while all the others remain fixed [52–54]. Optimization is carried out in several cycles until a change in each of the variables leads to a decrease in the value being optimized. It should be noted that although this method is very simple, it requires a large expenditure of working gases and time.

Moreover, according to the theory, the method of cyclic coordinate descent cannot always guarantee finding the global extremum of a function of many variables [52–54].

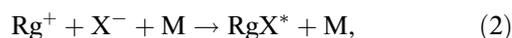
It is important to note that the greater complexity in this problem is inherent in the optimization of the working mixture, i.e., determination of the optimal partial pressures of the gas mixture components. Therefore, in connection with the existing problem, the development of techniques that allow accelerating the process of optimizing multiband excilamps based on a multicomponent mixture is of great practical value. To the best of our knowledge, no work has been carried out previously to develop techniques for optimizing the active media of gas-discharge radiation sources, in particular, multiband excilamps.

1. Radiation of exciplex molecules

It is well known that the formation of exciplex molecules is implemented mainly through two mechanisms [57–67]: the harpoon reaction



and the ion-ion recombination reaction



where Rg is an atom of an inert gas, X is a halogen atom, RgX* is an exciplex molecule, M is a third particle that carries away excess energy (as a rule, an atom of the working inert gas).

It should be noted that there are other mechanisms for the formation of exciplex molecules, such as [9,68–80], but they are less common and therefore are not considered in this paper.

As a rule, in most cases only one of the above reactions is predominant in the formation of exciplex molecules [1,17,18,81]. The efficiency of the formation of exciplex molecules due to the three-particle reaction of ion-ion recombination (2) is proportional to the probability of triple collision (approach to a distance within the interaction region) of the corresponding reagents [62,82–86], and hence their concentrations, which, respectively is realized at significant pressures of the working mixture. At relatively low pressures of the working mixture, the predominant contribution to the formation of exciplex molecules is made by the binary harpoon reaction (1) [1,17,18,87–93].

A review of the literature indicates that there is no consensus on the boundary values of the pressure of the working mixture, corresponding to the transition from the predominance of one mechanism for the formation of exciplex molecules to the predominance of another. The prevalence of one channel or another depends not only on the total pressure of the working mixture, but also on the pumping conditions and the type of halogen carrier. In particular, in the papers [1,17,18,81] it is noted that in the case of using Cl₂ molecules as a halogen carrier, the

formation of exciplex molecules in the mixture pressure up to an atmosphere is implemented mainly through the harpoon reaction. The authors of Ref. [94] came to a similar conclusion, having found that in the range of optimal pressures of a mixture of xenon and molecular chlorine (~200–250 Torr), the harpoon reaction is the main channel for the formation of XeCl* exciplex molecules.

In this paper, we assume that at optimal pressures of the working mixture, the contribution of reaction (2) to the formation of exciplex molecules can be neglected. This situation is more consistent with excilamps with a relatively low total pressure of the mixture, such as excilamps with excitation by glow and capacitive discharges, although Refs. [1,17,18,81,94] indicate a negligible contribution of the ion-ion recombination also in barrier discharge with a working mixture pressure of hundreds of Torr for mixtures containing Cl₂. Then, by analyzing the harpoon reaction (1), it is possible to conclude that the case of a mixture of one inert gas with several halogen carriers has a feature that simplifies the consideration of the kinetics of the exciplex molecules formation.

The theoretical description of laser and lamp sources is a complex problem. In such a description, a large number of problems are simultaneously solved that are directly related to the processes taking place in these systems, ranging from power source, the processes of introducing energy into the medium, the kinetic description of the processes occurring in the plasma, taking into account the concentration inhomogeneity of the components of the medium, till the processes of formation and extraction of radiation from these sources. Such a complete description is very costly and justified at the design stage of any large installations of exceptional importance. Note, for example, that the total number of plasma-chemical processes taken into account in modern models reaches several hundred. But often such a detailed description is not necessary. When using reasonable assumptions, as a rule, it is possible to describe the output characteristics of radiation sources without significant losses, while significantly reducing the complexity of modeling. One of such assumptions is the use of a uniform distribution of reagent concentrations in the active medium. Such models turned out to be able to adequately describe not only the output characteristics of radiation, but also the optimal values of various parameters from the concentrations of various reagents to the parameters of resonators in both lamp [1,17,18] and laser sources of radiation with different types of nonequilibrium in both plasma [58–67] and gas [95,96] lasers. Because of this, the distributions of plasma parameters will be assumed to be uniform in the following consideration. In addition, we will consider only a few of a huge number of reactions, that are of key importance for the issues considered in this paper.

Since the partial pressures of halogen carriers can be considered constant, and hence their concentrations are known, the only unknown (reagent) in the harpoon reaction of the formation of exciplex molecules is the concentration of excited atoms of the inert gas, which depends on the

excilamp excitation conditions. But, since the excited atoms of an inert gas in the case of a mixture of one inert gas with several halogen carriers are a common reagent for the formation of exciplex molecules of both one type and other types, this significantly simplifies the analysis of exciplex molecule formation kinetics and the excilamp optimization. It is quite obvious that in this case there should be a complete correlation of the rates of formation of all types of exciplex molecules due to harpoon reactions.

Let us proceed to a simplified consideration of the kinetics of the key processes relevant to the case. As an example, we will consider a ternary mixture of xenon inert gas with molecular chlorine and bromine (Xe–Cl₂–Br₂). It should be noted that this analysis is general and remains valid in the case of mixtures with another inert gas or with another combination of halogen carriers. As halogen carriers, mainly homonuclear diatomic halogen molecules are assumed. For the case under consideration, the kinetic equations describing the time dependences of the concentration of XeCl*, XeBr* exciplex molecules formed and Xe* excited xenon atoms will have the following form:

$$\begin{cases} \frac{d[\text{XeCl}^*]}{dt} = k_1[\text{Cl}_2][\text{Xe}^*] - \frac{[\text{XeCl}^*]}{\tau(\text{XeCl}^*)} \\ \frac{d[\text{XeBr}^*]}{dt} = k_2[\text{Br}_2][\text{Xe}^*] - \frac{[\text{XeBr}^*]}{\tau(\text{XeBr}^*)} \\ \frac{d[\text{Xe}^*]}{dt} = \Gamma - k_1[\text{Cl}_2][\text{Xe}^*] - k_2[\text{Br}_2][\text{Xe}^*] - F[\text{Xe}^*] \end{cases}, \quad (3)$$

where k_1 and k_2 are the rate constants for the formation of XeCl* and XeBr* exciplex molecules due to the harpoon reaction (1), respectively, $\tau(\text{XeCl}^*)$ and $\tau(\text{XeBr}^*)$ are the effective lifetimes of exciplex molecules XeCl* and XeBr*, respectively, Γ is the function describing the formation of excited xenon atoms, F is the function considering all other exit channels of excited xenon atoms (ionization, quenching, etc.).

In system (3), the channels of formation and escape of excited xenon atoms are replaced by functions Γ and F , which are not specified (see also system (14)). It may seem that in such a description we gain nothing, since these functions are unknown to us. Actually this is not true. First, as a result, we understand the nature of the evolution of the concentrations of the reagents and powers of interest (see (7)–(10), (15)–(18)). Second, these functions drop out of the relations based on which the optimization is proposed (see (11)–(12), (19)). Note that the processes of quenching of radiating states are also present, and they should also be taken into account, but we do not do this for the reasons described in Section 4 of this paper.

The average radiation powers of XeCl* and XeBr* exciplex molecules, which are ultimately of our interest, are expressed as

$$P(\text{XeCl}^*) = \frac{V}{T} \int_0^T \frac{[\text{XeCl}^*]hv(\text{XeCl}^*)}{\tau(\text{XeCl}^*)} dt,$$

$$P(\text{XeBr}^*) = \frac{V}{T} \int_0^T \frac{[\text{XeBr}^*]hv(\text{XeBr}^*)}{\tau(\text{XeBr}^*)} dt, \quad (4)$$

where V is the volume of the active (radiating) medium, T is the period of exciting pulses, $hv(\text{XeCl}^*)$ and $hv(\text{XeBr}^*)$ are the energies of photons emitted by the XeCl* and XeBr* exciplex molecule, respectively.

If there is no mixture degradation, then upon reaching the steady-state regime

$$\int_0^T \frac{d[\text{XeCl}^*]}{dt} dt = 0, \quad \int_0^T \frac{d[\text{XeBr}^*]}{dt} dt = 0, \quad (5)$$

and Eq. (4) can be rewritten in the form

$$\begin{aligned} P(\text{XeCl}^*) &= \frac{V}{T} \int_0^T k_1[\text{Cl}_2][\text{Xe}^*]hv(\text{XeCl}^*) dt, \\ P(\text{XeBr}^*) &= \frac{V}{T} \int_0^T k_2[\text{Br}_2][\text{Xe}^*]hv(\text{XeBr}^*) dt, \end{aligned} \quad (6)$$

and under optimal operating conditions, when exit channels of excited xenon atoms are insignificant, we can rewrite (6) as a branch ratio:

$$\begin{aligned} P(\text{XeCl}^*) &= \frac{k_1[\text{Cl}_2][\text{Xe}^*]hv(\text{XeCl}^*)}{k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F} fV, \\ P(\text{XeBr}^*) &= \frac{k_2[\text{Br}_2][\text{Xe}^*]hv(\text{XeBr}^*)}{k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F} fV, \end{aligned} \quad (7)$$

where $f = 1/T$ is the repetition rate of the excilamp excitation pulses, $[\text{Xe}^*]'$ is the concentration of excited xenon atoms formed during the pump pulse

$$[\text{Xe}^*]' = \int_0^T [\text{Xe}^*](k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F) dt. \quad (8)$$

When deriving Eq. (7), similar to Eq. (6), it is taken into account that on reaching the steady-state regime

$$\int_0^T \frac{d[\text{Xe}^*]}{dt} dt = 0$$

and

$$\int_0^T \Gamma dt = \int_0^T (k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F)[\text{Xe}^*] dt,$$

respectively,

$$\begin{aligned} \int_0^T k_1[\text{Cl}_2][\text{Xe}^*] dt &\cong k_1[\text{Cl}_2] \int_0^T \frac{(k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F)[\text{Xe}^*] dt}{k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F} \\ &\cong \frac{k_1[\text{Cl}_2]}{k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F} \int_0^T (k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F)[\text{Xe}^*] dt \\ &= \frac{k_1[\text{Cl}_2][\text{Xe}^*]'}{k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F}. \end{aligned}$$

Note also that in the case of steady-state pumping (for example, pumping by a glow discharge or an electron beam), the radiation powers of XeCl^* and XeBr^* exciplex molecules will be equal to

$$\begin{aligned} P(\text{XeCl}^*) &= k_1[\text{Cl}_2][\text{Xe}^*]''h\nu(\text{XeCl}^*)V, \\ P(\text{XeBr}^*) &= k_2[\text{Br}_2][\text{Xe}^*]''h\nu(\text{XeBr}^*)V, \end{aligned} \quad (9)$$

where $[\text{Xe}^*]''$ is the stationary concentration of excited xenon atoms,

$$[\text{Xe}^*]'' = \frac{\Gamma}{k_1[\text{Cl}_2] + k_2[\text{Br}_2] + F}, \quad (10)$$

where Γ is the rate of formation of excited xenon atoms (see formula (3), for the case of steady-state pumping $\Gamma = \text{const}$).

Thus, the ratio of the average radiation powers of XeCl^* and XeBr^* exciplex molecules will be estimated by the simple expression

$$\frac{P(\text{XeCl}^*)}{P(\text{XeBr}^*)} = \frac{k_1[\text{Cl}_2]h\nu(\text{XeCl}^*)}{k_2[\text{Br}_2]h\nu(\text{XeBr}^*)}, \quad (11)$$

and the ratio of partial pressures of halogen carriers necessary to achieve the desired ratio of the average radiation powers of exciplex molecules XeCl^* and XeBr^*

$$\frac{p(\text{Cl}_2)}{p(\text{Br}_2)} = \frac{k_2}{k_1} \frac{h\nu(\text{XeBr}^*)}{h\nu(\text{XeCl}^*)} \frac{P(\text{XeCl}^*)}{P(\text{XeBr}^*)}, \quad (12)$$

where $p(\text{Cl}_2)$ and $p(\text{Br}_2)$ are the partial pressures of molecular chlorine and bromine, respectively.

The paper [49] presents the results of an experimental study of a barrier discharge excilamp based on the three-component mixture $\text{Kr}-\text{Cl}_2-\text{Br}_2$. We use the data of this paper to test the obtained relations. According to expression (12), the ratio of partial pressures of halogen carriers in the $\text{Kr}-\text{Cl}_2-\text{Br}_2$ mixture, ensuring commensurate average radiation powers of KrCl^* and KrBr^* exciplex molecules, is

$$\frac{p(\text{Br}_2)}{p(\text{Cl}_2)} = \frac{k(\text{KrCl}^*)}{k(\text{KrBr}^*)} \frac{h\nu(\text{KrCl}^*)}{h\nu(\text{KrBr}^*)} \approx 2.5,$$

where the rate constants of harpoon reactions are $k(\text{KrCl}^*) = 7.3 \cdot 10^{-10} \text{ cm}^3/\text{s}$ [97] and $k(\text{KrBr}^*) = 2.7 \cdot 10^{-10} \text{ cm}^3/\text{s}$ [98].

According to the results of experiment [49], the optimal composition of the working mixture, at which the commensurate radiation of the KrCl^* and KrBr^* exciplex molecules is achieved, is as follows: $\text{Kr}/\text{Cl}_2/\text{Br}_2 = 200/0.3/0.7$ Torr, i.e. the optimal ratio of partial pressures of molecular bromine and chlorine is $p(\text{Br}_2)/p(\text{Cl}_2) = 2.3$.

As we can see, the ratio of partial pressures of halogen carriers, estimated from expression (12), is in good agreement with the value determined experimentally. It should be noted that in Ref. [49], a barrier discharge was used and the total pressure of the working mixture was relatively high ($\sim 200-300$ Torr), which once again confirms the

correctness of the assumption of a negligible contribution from ion-ion recombination reactions. The insignificant contribution of this channel to the formation of exciplex molecules is likely due to the insufficient formation of ion-ion recombination reagents, especially negative atomic halogen ions, rather than to the total pressure of the working mixture.

It should be noted that expression (12) is also suitable for estimating the ratio of partial pressures of halogen carriers in the case of an excilamp based on an inert gas mixture with three or more halogen carriers. In this case, the final expressions (11) and (12) do not undergo any changes. It is easy to verify this if in the expressions involving the function F we single out the channel for the escape of excited xenon atoms to form third-class exciplex molecules, i.e., represent F as $k[\text{X}_2] + F$, where $[\text{X}_2]$ is the third-class halogen carrier concentration and k is the rate constant for the formation of third-class exciplex molecules due to the harpoon reaction with X_2 . Thus, consistently applying the expression (12) for other halogen carriers, it is possible to estimate the ratio of partial pressures of all halogen carriers in the working mixture.

Determining the ratio of partial pressures of halogen carriers can significantly reduce the gas consumption and the duration of the process of optimizing a multiband excilamp on a mixture of one inert gas and several halogen carriers. In this case, optimization is substantially simplified and, in principle, reduces to the case of optimizing an excilamp on a two-component mixture, in which the percentage of inert gas and the total pressure of the working mixture are found.

When making estimates in practice, the absence of reliable values of the rate constants of harpoon reactions can cause difficulties. But to make an estimate, it is sufficient to have the ratio of reaction rate constants, which can be determined from the same ratio (12) by substituting the experimentally measured ratio of the radiation powers of exciplex molecules. In this case, it should be kept in mind that it is better to carry out measurements at relatively low pressures and pump power in order to level out the quenching processes and the contribution from the three-particle ion-ion recombination reaction.

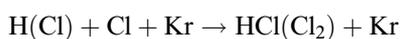
It should be noted that the question of the actual concentrations of halogen carriers in the mixture has not yet been fully clarified. The point is that some time after the onset of the excilamp operation, the concentrations of halogen carriers may change. It depends on the type of halogen-containing molecule, the material of the lamp, the procedure for its passivation, the method of preparing the mixture, the presence of impurities that arise due to various reasons. As an example, we recall the well-known facts related to mixtures containing hydrogen chloride and hydrogen bromide, although these halogen-containing molecules are not considered in this paper. For example, in mixtures with Cl_2 molecules at high currents in glow discharge tubes, the radiation efficiency, as noted in the papers [89,99], significantly exceeds the radiation efficiency

compared to mixtures containing HCl molecules (Note that the best characteristics of XeCl, KrCl exciplex lasers were obtained with the HCl molecule [59–61,67]).

In Ref. [89] it is noted that this is due to the fact that the rates of harpoon reactions with Cl₂ molecules significantly exceed the rates of reactions with the participation of HCl molecules. But it turns out that in the regime of low glow discharge currents, the emission efficiencies of mixtures with these two donors of chlorine molecules are approximately the same [99,100]. This can no longer be explained by the difference in reaction rates, since then the same difference in efficiencies would also exist at low glow discharge currents. On the other hand, if the rates of harpoon reactions are equal, the differences in radiation efficiencies could be attributed to the presence of impurities in the mixtures, but then, again, these impurities should lead to the same consequences for high and low glow discharge currents.

Perhaps this is due to the difference in the rate constants of attachment to these molecules with the formation of negatively charged ions, including atomic chlorine ions in different energy ranges of the discharge electrons, but most likely, this is due to the partial dissociation of HCl molecules. When the discharge is switched on, the initial HCl concentration may not correspond to the actual HCl concentration in the glow discharge plasma. In the plasma, the processes of both HCl decay (due to harpoon reactions of the formation of exciplex molecules) into H and Cl atoms and the association of these atoms into HCl and Cl₂ molecules are permanently occurring. Thus, if the initial mixture contains only HCl molecules, then Cl₂ molecules also accumulate during excilamp operation, and the efficiency of the excilamps can become equal. However, it is not clear why this occurs only at low currents. Partial confirmation of this point of view is contained in Refs. [101,102]. When hydrogen molecules are added to the active medium of a CuBr laser, the same changes occur in the temporal behavior of the lasing characteristics of this laser as when hydrogen bromide molecules are added.

If the dissociation of molecules is significant, then the actual concentration of HCl, Cl₂ will always be less than the initial concentration of HCl, Cl₂ in the mixture. The association rate resulting from bulk reactions

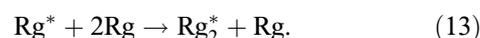


is small, especially at low pressures of the mixture. It is possible that the reduction of HCl and Cl₂ molecules occurs mainly on the walls of the discharge tube, but these data are not yet known. Thus, we do not know the actual degree of dissociation of HCl, Cl₂ molecules in the discharge and, therefore, we cannot assume that the concentration of halogen carriers during operation of the excilamp is the same as at the moment of gas puffing when preparing the mixture for work. If the difference between the behavior of excilamp output powers

and the concentrations of halogen carriers depends on the processes of restoring their concentrations on the tube walls, then it should be expected that the actual concentration of halogen donors during excilamp operation will depend not only on the type of halogen carrier, but also on the design of the excilamp, in particular, on the material of its walls and its geometry, which affects the rate of diffusion of halogen-carrier atoms to the walls of the excilamp. Thus, the concentrations of halogen carriers should be controlled during the operation of the excilamp. This is more expensive than the determination of concentrations before starting its work, but in principle it is possible. For example, the built-in reversible generator HBr [95,102] made it possible not only to supply HBr to the laser active medium, but also to pump it back to the generator. This made it possible to maintain the pressure of hydrogen bromide constant at the level required in the experiment.

2. Radiation of exciplex molecules

An electric discharge in mixtures containing an inert gas (Rg) can lead to the formation of excimer molecules due to a three-body association reaction [12,15,57,59–61,67,103]:



In this connection, the issue of the radiation power of excimer molecules formed in an excilamp together with exciplex molecules may be of interest. As in the previous case, the only unknown (reagent) in the reaction of the formation of excimer molecules (13) is the concentration of excited atoms of the inert gas (since we transfer the results of optimization of two-component mixtures to multicomponent mixtures, it is assumed that we know the optimal concentrations of the reagents of a two-component mixture), which depends on the excilamp pumping conditions. However, since excited atoms of an inert gas are a common reagent for the formation of both excimer and exciplex molecules, this makes it possible to simplify the assessment of the radiative characteristics of excilamps.

Let us proceed to a simplified consideration of the kinetics of the key processes relevant to the case. As an example, we will also consider a ternary mixture of xenon inert gas with molecular chlorine and bromine (Xe–Cl₂–Br₂). The kinetic equations describing the time dependences of the concentration of the formed XeCl*, XeBr* exciplex molecules, Xe₂* excimer molecules, and

excited xenon atoms Xe^* will have the following form:

$$\begin{cases} \frac{d[XeCl^*]}{dt} = k_1[Cl_2][Xe^*] - \frac{[XeCl^*]}{\tau(XeCl^*)} \\ \frac{d[XeBr^*]}{dt} = k_2[Br_2][Xe^*] - \frac{[XeBr^*]}{\tau(XeBr^*)} \\ \frac{d[Xe_2^*]}{dt} = k_3[Xe][Xe][Xe^*] - \frac{[Xe_2^*]}{\tau(Xe_2^*)} \\ \frac{d[Xe^*]}{dt} = \Gamma - (k_1[Cl_2] + k_2[Br_2] + k_3[Xe]^2 + F)[Xe^*], \end{cases} \quad (14)$$

where k_1, k_2 and k_3 are, respectively, the rate constants for the formation of exciplex molecules $XeCl^*$, $XeBr^*$ due to the harpoon reaction (1) and excimer molecules Xe_2^* due to the association reaction (13), $\tau(XeCl^*)$, $\tau(XeBr^*)$ and $\tau(Xe_2^*)$ are, respectively, the effective lifetimes of $XeCl$ exciplex molecules*, $XeBr^*$ and Xe_2^* excimer molecules, Γ is the function describing the formation of excited xenon atoms, F is the function that takes into account all other exit channels of excited xenon atoms (ionization, quenching, etc.).

A similar consideration (see Section 2 for details) of equations (14) leads to the average powers

$$\begin{aligned} P(XeCl^*) &= \frac{k_1[Cl_2][Xe^*]'hv(XeCl^*)}{k_1[Cl_2] + k_2[Br_2] + k_3[Xe]^2 + F} fV, \\ P(XeBr^*) &= \frac{k_2[Br_2][Xe^*]'hv(XeBr^*)}{k_1[Cl_2] + k_2[Br_2] + k_3[Xe]^2 + F} fV, \\ P(Xe_2^*) &= \frac{k_3[Xe]^2[Xe^*]'hv(Xe_2^*)}{k_1[Cl_2] + k_2[Br_2] + k_3[Xe]^2 + F} fV, \end{aligned} \quad (15)$$

where $f = 1/T$ is the repetition rate of the excilamp excitation pulses, $[Xe^*]'$ is the concentration of excited xenon atoms produced during the pump pulse

$$[Xe^*]' = \int_0^T [Xe^*](k_1[Cl_2] + k_2[Br_2] + k_3[Xe]^2 + F) dt. \quad (16)$$

Note also that in the case of steady-state pumping (for example, pumping by a glow discharge or an electron beam), the expressions describing the radiation powers of exciplex molecules $XeCl^*$, $XeBr^*$ and excimer molecules Xe_2^* have the following form:

$$\begin{aligned} P(XeCl^*) &= k_1[Cl_2][Xe^*]''hv(XeCl^*)V, \\ P(XeBr^*) &= k_2[Br_2][Xe^*]''hv(XeBr^*)V, \\ P(Xe_2^*) &= k_3[Xe]^2[Xe^*]''hv(Xe_2^*)V, \end{aligned} \quad (17)$$

where $[Xe^*]''$ is the stationary concentration of excited xenon atoms,

$$[Xe^*]'' = \frac{\Gamma}{k_1[Cl_2] + k_2[Br_2] + k_3[Xe]^2 + F}, \quad (18)$$

where Γ is the rate of formation of excited xenon atoms ($\Gamma = \text{const}$ for steady-state pumping).

Thus, the ratio of the average radiation powers of excimer molecules Xe_2^* and exciplex molecules $XeCl^*$ can be estimated by the simple expression

$$\frac{P(Xe_2^*)}{P(XeCl^*)} = \frac{k_3[Xe]^2hv(Xe_2^*)}{k_1[Cl_2]hv(XeCl^*)}. \quad (19)$$

Since excimer molecules emit in the VUV spectral region, their application is significantly limited by the strong absorption of VUV radiation in the atmosphere. In practice, radiation from exciplex molecules is predominantly used, and the radiation from excimer molecules accompanying the excilamp is not taken into account. If the excilamp is optimized with respect to the total power of excimer and exciplex molecules, and excimer radiation is not used, then this can lead, due to reaction (13), to a significant decrease in the efficiency of the excilamp at exciplex radiation wavelengths. In this case, to increase the efficiency of the excilamp, it is better to focus on the conditions when the radiation of excimer molecules is suppressed in comparison with the radiation of exciplex molecules. Relation (19) can be useful in this case and when optimizing single-band excilamps on binary mixtures. By setting the radiation power of excimer molecules to be significantly lower than the radiation power of exciplex molecules, one can estimate the relative content of the inert gas.

3. Method of optimizing the gas mixture of a multiband excilamp

When optimizing a two-component working mixture, the following approach is often used in practice. The excilamp is filled with gases with a trial ratio of the mixture components and a slight excess in total pressure. Next, the total pressure of the mixture is gradually reduced by pumping out the excilamp with simultaneous recording of the radiation power of the exciplex molecules. After running one ratio of the mixture components, another is run. Thus, the optimal total pressure and the ratio of the components of the working mixture are found, at which the radiation power or efficiency is maximum. However, in the case of a multicomponent gas mixture, this procedure becomes more complicated due to the large number of variables.

If it is only needed to achieve the desired ratio of the radiation powers of exciplex molecules, and the radiation of excimer molecules is insignificant, then the process of optimizing such an excilamp is relatively simple. Since the ratio of partial pressures of halogen carriers can be estimated from expression (12), it remains to experimentally determine only the relative content of the inert gas and the total pressure of the mixture. And this problem is similar to the optimization of an excilamp on a two-component mixture, despite the possible use of a large number of different halogen carriers. As can be seen in Eq. (12), the ratio of the radiation powers of exciplex molecules at a constant ratio of partial pressures of halogen carriers does not depend on the total pressure of the

mixture (within the limits where the contribution of the ion-ion recombination reaction and quenching processes is insignificant). Therefore, the aforementioned technique for optimizing an excilamp by means of pumping out can be successfully used in conjunction with making estimates using relation (12).

Let us consider the case when it is necessary to provide the desired ratio of radiation powers not only for exciplex molecules, but also for excimer ones. Since the ratio of the components of the gas mixture can be estimated from expressions (12) and (19), it is necessary to select experimentally only the total pressure of the mixture. However, in this case, the pumping technique becomes more complicated, since the ratio of the radiation powers of excimer to exciplex molecules, according to expression (19), will decrease linearly as the excilamp is pumped out. Based on expression (19), it is not difficult to conclude that if the total pressure of the mixture during pumping decreases by n times, then in order for the ratio of the radiation powers of excimer and exciplex molecules to remain unchanged, it is necessary to increase the content in the working mixture of inert gas in \sqrt{n} times. Therefore, the total pressure of the mixture upon evacuation by $\Delta p_{\text{mixture}}$ must be increased by puffing in inert gas up to the value of p'_{mixture} equal to

$$p'_{\text{mixture}} = \frac{p_{\text{mixture}} - \Delta p_{\text{mixture}}}{p_{\text{mixture}}} \times \left(p(\text{Cl}_2) + p(\text{Br}_2) + p(\text{Xe}) \sqrt{\frac{p_{\text{mixture}}}{p_{\text{mixture}} - \Delta p_{\text{mixture}}}} \right), \quad (20)$$

where p_{mixture} is the mixture pressure before pumping, $\Delta p_{\text{mixture}}$ is the mixture pressure change by pumping, p'_{mixture} is the mixture pressure after addition of inert gas.

The recalculation of the partial pressures of the mixture components with the subsequent calculation of the amount of the necessary addition of an inert gas after each pumping step requires the use of computers, which can cause inconvenience in the process of optimizing the excilamp. But in practice, these recalculations can in principle be avoided. Based on the readings of the measuring equipment after each step of pumping out the mixture, it is necessary to restore the previous ratio of the radiation powers of the exciplex and excimer molecules due to the puffing of the inert gas.

4. Influence of quenching processes

If we also take into account the processes of quenching of excimer and exciplex molecules, then the powers of their radiation will be determined by the following expressions:

$$P(\text{XeCl}^*) = \frac{k_1[\text{Cl}_2][\text{Xe}^*]hv(\text{XeCl}^*)}{k_1[\text{Cl}_2] + k_2[\text{Br}_2] + k_3[\text{Xe}]^2 + F} \frac{fV}{Q(\text{XeCl}^*)},$$

$$P(\text{XeBr}^*) = \frac{k_2[\text{Br}_2][\text{Xe}^*]hv(\text{XeBr}^*)}{k_1[\text{Cl}_2] + k_2[\text{Br}_2] + k_3[\text{Xe}]^2 + F} \frac{fV}{Q(\text{XeBr}^*)},$$

$$P(\text{Xe}_2^*) = \frac{k_3[\text{Xe}]^2[\text{Xe}^*]hv(\text{Xe}_2^*)}{k_1[\text{Cl}_2] + k_2[\text{Br}_2] + k_3[\text{Xe}]^2 + F} \frac{fV}{Q(\text{Xe}_2^*)}, \quad (21)$$

where

$$Q(Y^*) = \tau(Y^*) \left(k'[\text{Cl}_2] + k''[\text{Br}_2] + k'''[\text{Xe}] + k''''n_e + \frac{1}{\tau(Y^*)} \right)$$

is a function taking into account the processes of quenching of the corresponding exciplex/excimer molecules; (Y^* is the exciplex/excimer molecule; k', k'', k''', k'''' are the quenching rate constant of the exciplex/excimer molecule Y^* by chlorine molecules, bromine molecules, xenon atoms and electrons, respectively).

If the inverse lifetime of molecules $\frac{1}{\tau(Y^*)}$ is much greater than the quenching rate

$$(k'[\text{Cl}_2] + k''[\text{Br}_2] + k'''[\text{Xe}] + k''''n_e) \ll \frac{1}{\tau(Y^*)}, \quad (22)$$

then the function $Q(Y^*) \approx 1$, which means that the quenching processes can be neglected. However, at significant concentrations of quenching particles, the quenching processes become significant and affect the radiative characteristics of the excilamp, and relations (12) and (19) are changed, but it can be expected that the quenching processes have a significant effect outside the optimal operating conditions of the excilamp. The grounds for believing this are based on the fact that, under optimal operating conditions, the quenching of exciplex molecules should be insignificant, since it leads to a decrease in the efficiency of the excilamp, which means that the operating conditions of the excilamp under which quenching is significant will not be optimal. Nevertheless, quenching processes can manifest themselves and interfere with the optimization of the excilamp, especially when the conditions are far from optimal. Inequality (22) can be used as an additional condition for estimating the absolute concentrations of the excilamp gas mixture components in order to achieve its maximum efficiency.

Conclusion

The paper considers the problem of optimizing gas mixtures of multiband excilamps based on gas mixtures of one inert gas with several halogen carriers.

Simple expressions are presented that allow estimating the ratio of partial pressures of the excilamp gas mixture components, which makes it possible to accelerate the optimization of a multiband excilamp with the desired ratio of the radiation powers of the working molecules.

It is shown that the optimization of a multiband excilamp can be reduced to the problem of optimizing an excilamp on a two-component mixture, in which the percentage of inert gas and the total pressure of the working mixture are experimentally established.

For situations where it is necessary to use both exciplex and excimer radiation, an expression was obtained that regulates the relative contribution of powers depending on the inert gas concentration.

If the use of excimer radiation is not expected, then the maximum efficiency of the excilamp should be achieved while minimizing the escape of excited atoms of the inert gas to the formation of excimer molecules. Based on this, it is possible to estimate the ratio of partial pressures of an inert gas and a halogen carrier according to expression (19), setting the radiation power of excimer molecules lower than the power of exciplex ones. The degree of suppression of the radiation power of excimer molecules is selected based on the required power level of the excilamp.

The maximum efficiency of the excilamp should be observed when the intensity of the quenching of exciplex molecules is negligible. This circumstance is regulated by relation (22), which can be used as an additional condition for estimating the concentrations of the excilamp gas mixture components.

Conflict of interest

The authors declare that they have no conflict of interest.

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