COMPARATIVE CHARACTERISTICS OF EXCIMER XeCl LASER BASED ON He/Xe/HCl AND He/Xe/CF_2Cl_2 MIXTURES

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We carry out the comparative analysis of the discharge and emission dynamics in XeCl lasers on the basis of He:Xe:HCl and He:Xe:CF_2Cl_2 mixtures. According to the results of our experiments and a numerical simulation, the energies achieved in the investigated media are approximately equal, therefore, Freon-12 can be used for creating a competitive XeCl laser on the basis of a relatively harmless mixture.

1. Introduction

At the present time, the most prevalent halogen donors used in excimer lasers are molecular fluorine F_2 and hydrogen chloride HCl [1]. These substances are unsafe in operation because of their extreme toxicity. The maximum allowable concentration of hydrogen chloride in the working zone is equal to 5.0 mg/m^3, while that of fluorine amounts to 0.15 mg/m^3[2]. Both of them belong to danger class 2 [3]. Furthermore, F_2 and HCl are very chemically active, which puts strict demands to the material of a flash chamber.

In view of these reasons, the search for new mixtures based on comparatively harmless halogen-containing substances takes on a special significance. As a possible alternative, one can use different freons and, in particular, Freon-12 CF_2Cl_2. This compound is chemically inert and its maximum allowable concentration in the working zone equals 3000 mg/m^3, which corresponds to danger class 4 [4]. The advantage of the use of Freons as halogen-containing substances in excimer lasers is also proved by the optical diagnostics [5] of plasma in discharges in He and Xe with various halogen donors, which has testified to that the effectiveness of the formation of XeCl molecules in He:Xe:CCl_4 and He:Xe:CF_2Cl_2 mixtures is higher than that in He:Xe:HCl.

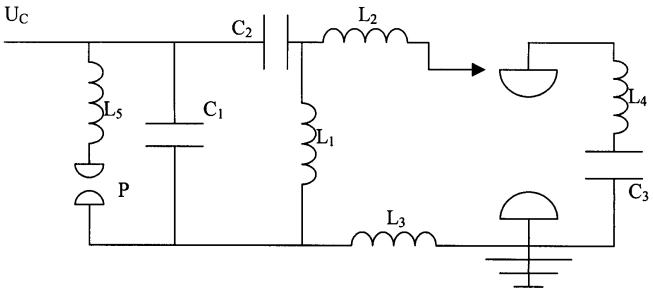

Fig. 1. Electric exciting circuit of the laser. $C_1 = C_2 = 25$ nF, $C_3 = 13$ nF, $L_1 = 2$ μH, $L_2 = 35$ nH

In the present work, we carry out the comparative analysis of the discharge and emission dynamics in XeCl lasers based on the He:Xe:HCl and He:Xe:CF_2Cl_2 mixtures. According to the results of our experiments and a numerical simulation, the energies achieved in the investigated media are approximately equal, therefore Freon-12 can be used for creating a competitive XeCl laser on the basis of a relatively harmless mixture.

2. Experimental Technique

Experimental investigations were carried out for an electric-discharge XeCl laser with automatic UV preionization based on a He:Xe:CF_2Cl_2 mixture. The chamber having 480 cm^3 in volume was produced from B caprolan while the electrodes were made of nickel. The volume of the active zone was equal to $33\times0.7\times1.3$ cm^3 (the latter value corresponds to the interelectrode distance). The chamber windows represented substrates made of KU-1 quartz. On the one side, the laser resonator was formed by an external dielectric mirror with a reflection factor of 99% at a wavelength of 308 nm. The other side represented the laser window.

The electric exciting circuit of a laser is depicted in Fig. 1. A two-stage high-voltage generator (HVG) was assembled according to the inverted lower stage circuit. The storage capacitors C_1 and C_2 as well as the peak one C_3 were composed from small-inductance pulse capacitors KVI-3. $C_1 = C_2 = 25$ nF, $C_3 = 13$ nF. The preionization was fulfilled during the charging of the peak capacitor by means of a discharge excited between the anode and a row of pins located at a distance of 2 mm astride the anode. The number of pins was

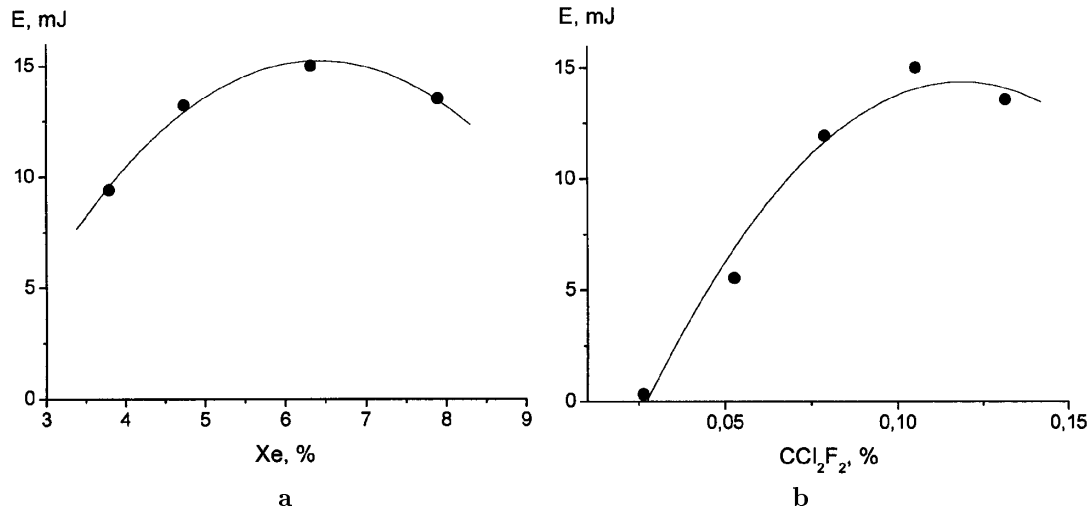

Fig. 2. Dependences of the emission energy on the concentration of Xe ($p(CF_2Cl_2) = 0.1\%$) (*a*) and CF_2Cl_2 (p(Xe)=6.3%) (*b*) measured at a total pressure of 2.5 atm.

made as large as possible and was equal to 54. The pins were connected to the storage capacitor through the decoupling inductances L_2 amounting to 1.9 μH. The total inductance L_2 was equal to 35 nH. The inductance L_1 used for the charging of the upper stage C_2 was equal to 2 μH. The storage capacitors P_1 and C_2 were charged from an external switch-mode power supply. The HVG commutation was fulfilled by means of a controlled discharger P.

The emission energy was measured with the help of an IMO-2N meter of the laser emission energy. The temporal characteristics of pulses of the emission and the discharge voltage and current were registered using a high-speed oscillograph S7-19. A light pulse was converted to an electric one by means of a coaxial FEK-22 SPU photocell. When measuring the voltage pulses, ohmic potential dividers were used including those belonging to the standard set of an S7-19 oscillograph. The current pulses were measured by means of the 0.01-Ohm shunt assembled from low-inductance resistances of the TVO type. By our estimates, the inaccuracy of measuring the energy and amplitude-temporal characteristics didn't exceed 20%.

In the present work, the emission energy of a laser was investigated depending on the pressure of Xe and CF_2Cl_2 at a constant total pressure of the mixture. With rising the total pressure, the emission energy increased, but a significant spread in the measured values of the emission energy was observed from one admission to another at pressures higher than 2.5 atm. It is probably associated with the necessity of a more exact admission of the halogen-containing substance. (By our estimates, the inaccuracy of the admission of a halogen-containing gas was equal to 8-10% while that of the Xe admission was much lower (<5%)). That's why, the dependence of the emission energy on the total pressure of the mixture was not investigated, and the total pressure was equal to 2.5 atm. In this case, a spread in values from one admission to another didn't exceed 5%. Fig. 2 shows the emission energy as a function of the Xe and CF_2Cl_2 concentrations. The maximal emission energy was obtained in the mixture He:Xe:CF_2Cl_2 = 93.6:6.3:0.1 and amounted to 15 mJ.

A XeCl laser on the basis of the He:Xe:HCl mixture was investigated in a number of publications. For the electric exciting circuit analogous to that used in the present paper, the emission energy reduced to the volume of our discharge chamber amounts to ~10-20 mJ [6].

3. Numerical Simulation

When numerically simulating the discharge and emission dynamics, the kinetic equations for the mixture components were solved together with the equations describing the laser supply circuit and the Boltzmann equation for the electron energy distribution function in an electric field [7]. The power density of the laser emission was calculated by the following formula:

$$W = \frac{1}{2} c\hbar\omega N_{\rm ph} \frac{1}{\tilde{l}} \frac{(1-r)(1-d)}{1-r+rd}, \qquad (1)$$

where N_{ph} is the photon concentration, $\tilde{l} = l/\ln\left(\frac{1}{r(1-d)}\right)$, l — the length of the discharge chamber, $\hbar\omega$ — the energy of a radiation quantum, c — the velocity of light, $r = 0.08$ — the reflection factor of the output mirror, $d = 0.2$ — the diffraction losses in the resonator.

The scheme of the reactions used when simulating the plasma kinetics is represented in Table 1. The rates of processes 1—10 were calculated from the Boltzmann equation. In this case, the cross sections for the electron interactions with helium and xenon atoms were taken from [8—10] and [8, 11—13], respectively. The cross sections characterizing the electron interactions with Freon-12 molecules are given in [14]. The cross section for the electron attachment to vibrationally excited freon molecules was not found in the literature. The rate of this process was assumed to be equal to that of the electron attachment to unexcited CF_2Cl_2 molecules.

Instead of the formation of $XeCl^*$ molecules, the quenching of excited Xe^* atoms by CF_2Cl_2 molecules results in the dissociation of Freon-12 with possibility of 97% [19]. That's why, the ion-ion recombination remains the only channel resulting in the generation of radiative XeCl* molecules in the He:Xe:CF_2Cl_2 medium.

According to [17], the rate of thermodissociation of heteronuclear ions (reactions 29—34) is assumed to be equal to 5.0 cm^3/s. A variation of this value between wide limits doesn't noticeably influence the plasma kinetics. The rate of quenching by freon molecules (reaction 49) was not found in the literature. That's why, we use the typical value for the quenching rate of excimer molecules by halogens — 1.0×10^{-9} cm^3/s.

The optimal concentration of xenon in the mixture calculated at a charging voltage of 17 kV, a pressure of 2.5 atm, and the Freon-12 concentration of 0.1%

Table 1

N	Reaction	Rate constant	Ref.
1	$e + He \rightarrow e + He^*$	Calculated	
2	$e + He \rightarrow e + e + He^+$	from the	
3	$e + Xe \rightarrow e + Xe^*$	Boltzmann	
4	$e + Xe \rightarrow e + e + Xe^+$	equation	
5	$e + Xe^* \rightarrow e + e + Xe^+$		
6	$e + C\ F_2Cl_2 \rightarrow e + CCl_2F_2^*$		
7	$e + C\ F_2Cl_2 \rightarrow e + CCl_2F_2(v)$		
8	$e + C\ F_2Cl_2\ (v) \rightarrow e + CCl_2F_2$		
9	$e + C\ F_2Cl_2 \rightarrow Cl^- + CClF_2$		
10	$e + C\ F_2Cl_2\ (v) \rightarrow Cl^- + CClF_2$		
11	$e + C\ F_2Cl_2 \rightarrow e + e + CCl_2F_2^+$		
12	$He^* + He^* \rightarrow He^+ + He + e$	2.0×10^{-10}	15
13	$Xe^* + Xe^* \rightarrow Xe + Xe^+ + e$	5.0×10^{-10}	15
14	$Xe^* + Xe^* \rightarrow Xe_2^+ + e$	1.0×10^{-9}	15
15	$Xe_2^* + Xe_2^* \rightarrow Xe_2^+ + 2Xe + e$	3.5×10^{-10}	15
16	$He^* + Xe \rightarrow Xe^+ + He + e$	7.5×10^{-11}	15
17	$He^+ + Xe \rightarrow Xe^+ + He$	1.0×10^{-11}	15
18	$He^* + 2He \rightarrow He_2^* + He$	0.43×10^{-33}	15
19	$Xe^* + 2Xe \rightarrow Xe_2^* + Xe$	8.0×10^{-32}	15
20	$Xe^* + Xe + He \rightarrow Xe_2^* + He$	1.4×10^{-32}	15
21	$Xe^+ + 2Xe \rightarrow Xe_2^+ + Xe$	3.6×10^{-31}	15
22	$Xe^+ + Xe + He \rightarrow Xe_2^+ + He$	1.3×10^{-31}	15
23	$Xe^+ + Xe + He \rightarrow HeXe^+ + Xe$	1.1×10^{-31}	16
24	$Xe^+ + He + He \rightarrow HeXe^+ + He$	1.1×10^{-31}	15
25	$He^+ + 2He \rightarrow He_2^+ + He$	8.0×10^{-32}	15
26	$He^+ + Xe + He \rightarrow He_2^+ + Xe$	1.0×10^{-31}	17
27	$He^+ + Xe + He \rightarrow HeXe^+ + He$	1.0×10^{-31}	17
28	$He^+ + 2Xe \rightarrow HeXe^+ + Xe$	1.0×10^{-31}	17
29	$HeXe^+ + He \rightarrow Xe^+ + 2He$	5.0×10^{-10}	17
30	$HeXe^+ + He \rightarrow He^+ + He + Xe$	5.0×10^{-10}	17
31	$HeXe^+ + He \rightarrow He_2^+ + Xe$	5.0×10^{-10}	17
32	$HeXe^+ + Xe \rightarrow Xe^+ + He + Xe$	5.0×10^{-10}	17
33	$HeXe^+ + Xe \rightarrow He^+ + 2Xe$	5.0×10^{-10}	17
34	$HeXe^+ + He \rightarrow Xe_2^+ + He$	5.0×10^{-10}	17
35	$Xe_2^+ + e \rightarrow Xe^* + Xe$	$2.3 \times 10^{-6} \times (0.026/T_e)^{0,5}$	17
36	$He_2^+ + e \rightarrow He^* + He$	$5.0 \times 10^{-10} \times (0.026/T_e)$	17
37	$Xe^+ + Cl^- \rightarrow XeCl^*$	Calculated	18
38	$Xe_2^+ + Cl^- \rightarrow XeCl^* + Xe$	by Flannery	
39	$He^+ + Cl^- \rightarrow He + Cl$	formulas	
40	$He_2^+ + Cl^- \rightarrow 2He + Cl$		
41	$Xe^* + C\ F_2Cl_2 \rightarrow XeCl^* + CClF_2$	0.5×10^{-11}	19
42	$Xe^* + C\ F_2Cl_2 \rightarrow$	1.6×10^{-10}	19
43	$XeCl^* + He \rightarrow Xe + He + Cl$	1.0×10^{-12}	15
44	$XeCl^* + Xe \rightarrow 2Xe + Cl$	2.0×10^{-12}	15
45	$XeCl^* + 2Xe \rightarrow Xe_2Cl^* + Xe$	4.0×10^{-31}	15
46	$XeCl^* + Xe + He \rightarrow Xe_2Cl* + He$	1.5×10^{-31}	15
47	$XeCl^* + 2He \rightarrow Xe + Cl + 2He$	1.0×10^{-33}	15
48	$XeCl^* + e \rightarrow Xe + Cl + e$	2.0×10^{-7}	15
49	$XeCl^* + CCl_2F_2 \rightarrow$	1.0×10^{-9}	20
50	$Xe_2Cl^* + He \rightarrow 2Xe + Cl + He$	5.0×10^{-13}	15
51	$Xe_2Cl^* + Xe \rightarrow 3Xe + Cl$	5.0×10^{-13}	15
52	$Xe_2Cl^* + e \rightarrow 2Xe + Cl + e$	2.0×10^{-7}	15
53	$Cl^- + h\nu \rightarrow Cl + e$	6.3×10^{-7}	15
54	$Xe^* + h\nu \rightarrow Xe^+ + e$	2.28×10^{-7}	15
55	$Xe_2^+ + h\nu \rightarrow Xe^* + Xe^+$	7.5×10^{-7}	15
56	$Xe_2^* \rightarrow 2Xe$	6.0×10^{7}	15
57	$Xe_2Cl^* + h\nu \rightarrow Xe + Cl + Xe^+ + e$	7.8×10^{-7}	15
58	$He^* + h\nu \rightarrow He^+ + e$	8.4×10^{-8}	15
59	$Xe_2^* + h\nu \rightarrow Xe^+ + Xe + e$	4.2×10^{-7}	15
60	$He_2^* \rightarrow 2He$	3.6×10^{8}	15
61	$HeXe^+ + hv \rightarrow Xe^+ + He$	4.5×10^{-7}	15
62	$Xe_2Cl^* \rightarrow 2Xe + Cl + h\nu_4$	7.4×10^{6}	15
63	$XeCl^* \rightarrow XeCl(X) + h\nu$	9.0×10^{7}	15
64	$XeCl^* + h\nu \rightarrow XeCl(X) + 2h\nu$	1.2×10^{-5}	15
65	$XeCl(X) \rightarrow Xe + Cl$	5.0×10^{8}	15
66	$XeCl(X) + hv \rightarrow XeCl^*$	1.2×10^{-5}	15

*Note. Rate constants of n-particle reactions are given in units of $cm^{3(n-1)}/s$.

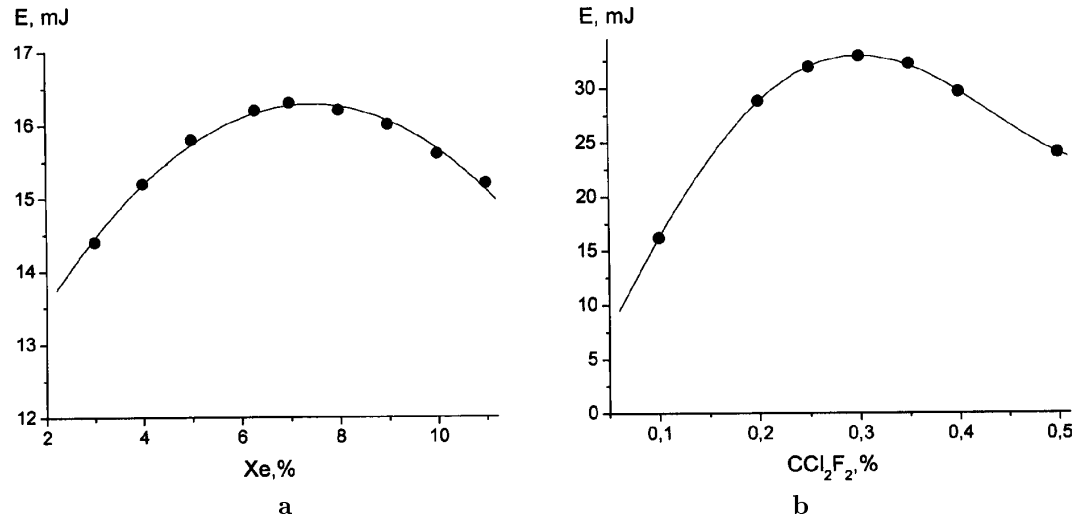

Fig. 3. Calculated dependences of the emission energy on the concentration of Xe ($p(CF_2Cl_2) = 0.1\%$) (*a*) and CF_2Cl_2 (p(Xe)=6.3%) (*b*). $U_c = 17$kV, $p = 2.5$ atm.

amounts to 7% (Fig. 3,*a*). This result agrees well with the experimental value 6.3%. The increase of the xenon concentration in the range from 3 to 10% results in a more efficient generation of excited $XeCl^*$ molecules due to the increasing number of positive Xe ions participating in the process of ion-ion recombination, which represents the dominant channel of the formation of $XeCl^*$ molecules. In this case, the density of chlorine ions remains practically invariable. Still, the dependence of the emission energy on the inert gas concentration reaches a maximum at 7%, which is stipulated by an increase of the total rates of photoionization $k_{54}Xe^*h\nu$ (54) and $k_{59}Xe_2^*h\nu$ (59) due to the rising densities of Xe^* and Xe_2^*.

In addition to the kinetic processes, the optimal concentration of a halogen-containing gas in the active medium of the laser is greatly influenced by the voltage reached between the electrodes of the discharge gap, i.e. by the parameters of the supply circuit. With the rising content of Freon-12 in the mixture, the breakdown voltage abruptly increases, so the discharge is hardly initiated at CCl_2F_2 concentrations higher than 0.3—0.4%. As a result, the electron density rapidly decreases, and consequently so does the concentration of Xe^+ ions. On the other hand, an increase of the Freon-12 concentration is accompanied with a more effective generation of negative chlorine ions. That's why, the total rate of ion-ion recombination $k_{37}N_{Xe^+}N_{Cl^-}$, which determines the efficiency of generating the excimer molecules $XeCl^*$, reaches a maximum. According to the results of numerical simulations, the optimal conditions for forming the excited $XeCl^*$ molecules are realized in the case where the content of Freon-12 in the medium amounts to 0.3% (Fig. 3,*b*).

In order to draw a conclusion about the effectiveness of the laser under investigation, its output characteristics should be compared to those of an analogous XeCl laser based on the standard mixture with HCl serving as a halogen-containing gas. For this purpose, we calculated the discharge and emission dynamics of a XeCl laser based on the He:Xe:HCl mixture for the same exciting circuit, charging voltage, pressure, and mixture ratio.

A HCl molecule is characterized by a number of peculiarities that separate it among other halogen donors used in excimer inert-gas halide lasers. First, the cross section for the electron attachment to an HCl molecule in the ground state is rather low and cannot provide the generation of Cl^- ions at a rate sufficient for the $XeCl^*$ formation in the process of ion-ion recombination. However, the vibrational excitation of HCl molecules is accompanied with an increase of the attachment cross section by orders of magnitude. Second, the quenching of excited Xe^* atoms by hydrogen chloride results in the dissociation of HCl with probability of 98% [19]. That's why, the ion-ion recombination represents the dominant channel of the generation of excimer $XeCl^*$ molecules.

The reactions with the participation of HCl molecules considered in the kinetic scheme are given in Table 2. The cross sections for processes 1—5 are taken from [11], while those for reactions 6,7 — from [21].

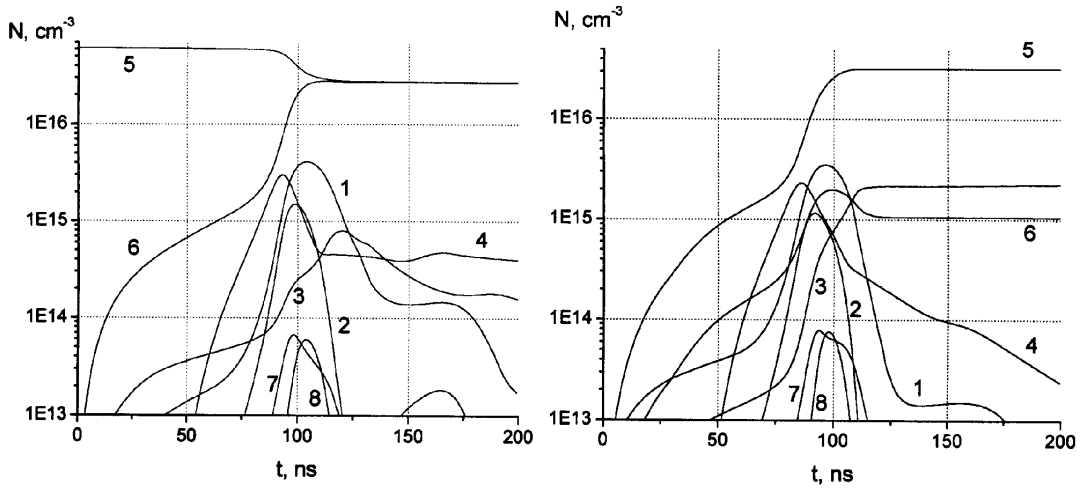

Fig. 4. Temporal evolutions of the basic plasma components in the He:Xe:CCl_2F_2mixture (*a*): *1* — N_e, *2* — Xe^+, *3* — Cl^- , *4* — Xe^*, *5* — CCl_2F_2, *6* — $CCl_2F_2(v)$, *7* — $XeCl^*$, *8* — $h\nu$ and in the He:Xe:HCl mixture (*b*): *1* — N_e, *2* — Xe^+, *3* — Cl^- , *4* — Xe^*, *5* — $HCl(v=1)$, *6* — $HCl(v=2)$, *7* — $XeCl^*$, *8* — $h\nu$

As the harpoon reaction is practically absent in both media, the efficiency of the formation of excited $XeCl^*$ molecules is determined by the total rate of ion-ion recombination $k_{37}Xe^+Cl^-$. Therefore, the advantage of using one of the analyzed halogen-containing gases depends on the efficiency of the generation of chlorine negative ions.

The temporal evolutions of the basic plasma components in the He:Xe:CCl_2F_2 and He:Xe:HCl media calculated at a pressure of 2.5 atm., a charging voltage of 17 kV, and a mixture ratio of 93.6:6.3:0.1 are depicted in Fig. 4. According to the results of numerical simulations, CF_2Cl_2 and HCl molecules provide the approximately equal concentrations of negative chlorine ions in the discharge. That's why the densities of excited $XeCl^*$ molecules and photons also have comparable magnitudes. Finally, the emission energy reached in the He:Xe:CF_2Cl_2 mixture amounts to 16.2 mJ, while that reached in the He:Xe:HCl mixture is equal to 15.7 mJ.

Table 2

N	Reaction	Rate constant, cm^3/c	Ref.
1	$e + HCl \to e + HCl^*$	Calculated	
2	$e + HCl \to e + HCl\ (v{=}1)$	from the	
3	$e + HCl \to e + HCl\ (v{=}2)$	Boltzmann	
4	$e + HCl \to H + Cl^-$	equation	
5	$e + HCl\ (v{=}1) \to H + Cl^-$		
6	$e + HCl\ (v{=}2) \to H + Cl^-$		
7	$e + HCl \to e + e + HCl^+$		
8	$Xe^* + HCl \to XeCl^* + H$	$< 1.0 \times 10^{-11}$	19
9	$Xe^* + HCl \to Xe + H + Cl$	5.6×10^{-10}	19
10	$Xe^* + HCl\ (v{=}1) \to XeCl^* + H$	$< 1.0 \times 10^{-11}$	19
11	$Xe^* + HCl\ (v{=}1) \to Xe + H + Cl$	5.6×10^{-10}	15
12	$Xe^* + HCl\ (v{=}2) \to XeCl^* + H$	$< 1.0 \times 10^{-11}$	19
13	$Xe^* + HCl\ (v{=}2) \to Xe + H + Cl$	2.0×10^{-10}	15
14	$XeCl^* + HCl \to Xe + Cl + HCl$	8.0×10^{-10}	15
15	$XeCl^* + HCl\ (v{=}1) \to Xe + Cl + HCl$	7.7×10^{-10}	15
16	$XeCl^* + HCl\ (v{=}2) \to Xe + Cl + HCl$	7.7×10^{-10}	15
17	$Xe_2Cl^* + HCl \to 2Xe + Cl + HCl$	2.6×10^{-10}	15

4. Conclusion

Our theoretical and experimental investigations have demonstrated that the active media He:Xe:CF_2Cl_2 and He:Xe:HCl provide approximately equal emission energies. Therefore, making use of Freon-12 as a halogen-containing gas allows one to create a competitive XeCl laser based on a low-toxic chemically inert mixture.

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ПОРІВНЯЛЬНІ ХАРАКТЕРИСТИКИ ЕКСИМЕРНОГО XeCl-ЛАЗЕРА НА СУМІШАХ He/Xe/HCl ТА He/Xe/CF_2Cl_2

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Р е з ю м е

Проведено порівняльний аналіз динаміки розряду і випромінювання XeCl-лазера на сумішах He:Xe:HCl та He:Xe:CF_2Cl_2. Згідно з результатами експерименту і чисельного моделювання в цих сумішах досягається приблизно однакова енергія випромінювання, так що фреон-12 може використовуватись для створення конкурентоздатного XeCl-лазера на порівняно безпечній суміші.