

INVESTIGATION OF THE WORK CONDITIONS AND CHARACTERISTICS OF MULTIWAVE EMITTERS ON CHLORIDES AND FLUORIDES OF RARE GASES

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We present the results of a design and systematic investigation of spectral, temporary and resource characteristics of an electrodischarge excimer emitter with radiation on 175 nm ArCl (B-X) / 222 nm KrCl (B-X) / 236 nm XeCl (D-X) / 258 nm Cl₂ (D'-A') / 308 nm XeCl (B-X); 175 nm ArCl (B-X) / 193 nm ArF (B-X); 222 nm KrCl / 249 nm KrF / 258 nm Cl₂ / 308 nm XeCl / 353 nm XeF. Active media of lamps were formed by gas-mixtures: Ar/Kr/Xe/Cl₂, Ar/CF₂Cl₂, He/Kr/Xe/CF₂Cl₂, with the help of a powerful transverse discharge with a spark preionization. The optimum pressure of the Ar/Kr/Xe/Cl₂ mixture is 15-20 kPa, whereas the partial pressure of chlorine is 0.2–0.4 kPa. In order to obtain bands of comparable brightness, the partial pressure of Kr and Xe atoms should be in the range 0.2–0.5 kPa.

Using the CF₂Cl₂ molecules as unique carriers of Cl and F atoms, the multiwave operation mode of excimer emitters with brightness ration Kr and Xe chlorides to fluorides as 9/1 is also realized. The optimal content of CF₂Cl₂ molecules was 0.008–0.01 kPa (Ar/CF₂Cl₂ mixtures). Correlation of B-X-bands brightness of ArCl and ArF was 10. The radiation expectancy in the multiwave mode reached 5×10^3 – 1×10^5 pulses. Such emitters can be used in pulse photometry, high energy chemistry, biology and medicine.

1. Introduction

Excimer emitters (Eem) with transverse discharge (TVD) pumping [1, 2] are most powerful of the wide class of such electric-discharge radiators. Such radiators radiate as a rule, on a single wavelength. The investigation of a multiwave mode of operation of such excimer emitters is of interest for a number of applications in pulsed photometry, for acting simultaneously and selectively on individual high-energy bands of chemically or biologically active compounds.

This communication describes a study of the conditions for simultaneous formation of ArCl(B), KrCl(B), KrF(B), Cl₂(D'), XeCl(B) and XeF(B) molecules in a TVD-plasma based on Ar/Kr/Xe/Cl₂, Ar/CF₂Cl₂, He/Kr/Xe/CF₂Cl₂ and He/Kr/Xe/SF₆/HCl mixtures.

2. Experimental

TVD-pumped emitters had $18 \times 2,2 \times (0.5 - 1.0)$ cm³ active media with an interelectrode distance of 2.2 cm. A schematic diagram of the radiation source is presented in [3, 4]. Preionization of the interelectrode space is performed with the help of two rays of spark gaps with parallel triggering. A thyatron TGI 1000/25 is used as a switch for the pulsed power supply of the TVD. The discharge was struck by means of a double-loop LC-circuit in which the capacitance of the main storage capacitor was 30 nF and total capacitance of the peaking capacitors was 9,4 nF.

The radiation from the TVD plasma was recorded using a half-meter Seya-Namioka vacuum monochromator. The TVD chamber is tightly connected to the input slit of the vacuum monochromator through a CaF₂ window. A FEU-142 photomultiplier with a LiF window served as the photodetector. The

working wavelength range of the vacuum spectrometer was 130–350 nm. The relative calibration of the vacuum monochromator+FEU-142 system was carried out in the range $\Delta\lambda=165\text{--}350$ nm on the basis of the continuous radiation of hydrogen molecules. The emission in range $\Delta\lambda=200\text{--}600$ nm was studied by using an MDR-2 monochromator and photomultiplier “FOTON”. The power and temporal characteristics of emission in the UV spectrum range were controlled with the help of an IMO-2H device, a 14-FC electronic linear amplifier and 6LOR-04 high-speed oscilloscope.

3. TVD in Ar/Cl₂, Ar/Kr(Xe)/Cl₂ and Ar/Kr/Xe/Cl₂ Mixtures

The main bands in TVD plasma on Ar/Cl₂ mixtures are 258 nm Cl₂ (D'-A') / 175 nm ArCl (B-X). Figure 1 shows the dependences of the emission intensities of the Cl₂* and ArCl* bands on the content of Cl₂ molecules and argon atoms in a discharge in the Ar/Cl₂ mixture. At $U_{ch} \leq 12,5$ kV, the optimal content of Cl₂ is 0,2–0,4 kPa.

For the Cl₂* band, a broad maximum of emission intensity is seen in the range of argon pressure in the TVD of 8–15 kPa, whereas for the B-X band of ArCl it is shifted to the region of 15–25 kPa. For TVD on the Ar/HCl and He/Ar/HCl mixtures, the optimal content of HCl molecules is 80 Pa and the maximal intensity of ArCl(B-X) emission decreases by 2–3 times. Such a behavior of the intensity of the ArCl(B-X) band can be due either to lower efficiency of ArCl(B) molecules generation as a result of the “harpoon” reaction $Ar^* + HCl(v) \rightarrow ArCl(B) + H$ as compared to the corresponding reaction with Cl₂ or to a considerable absorption of VUV radiation by hydrogen chloride. In the TVD based on Ar/Kr/Cl₂, Ar/Xe/Cl₂ and Ar/Kr/Xe/Cl₂ mixtures, the main radiation was concentrated at 175, 222, 258 nm; 175, 236, 258, 300 nm and 175 nm ArCl (B-X) / 222 nm KrCl (B-X) / 236 nm XeCl (D-X) / 258 nm Cl₂ (D'-A') / 308 nm XeCl (B-X) (Fig.2). A stable ignition of the TVD was achieved for the charging voltage $U_{ch} \geq 4.0$ kV.

An increase in the charging voltage from 4 to 12 kV led to the increase in the brightness of all of the observed bands by a factor of 2–5.

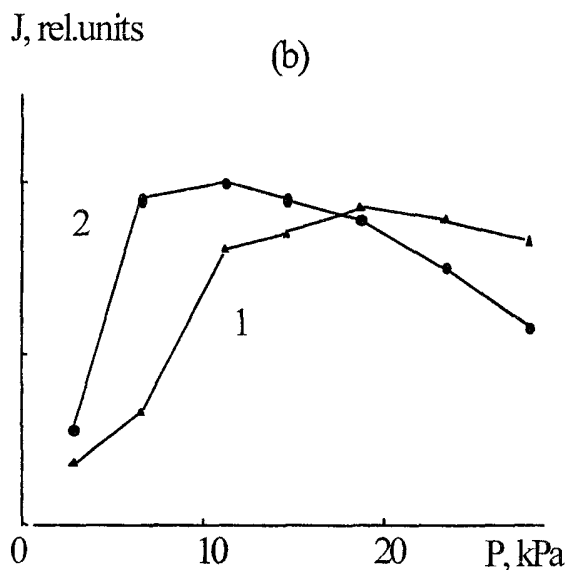
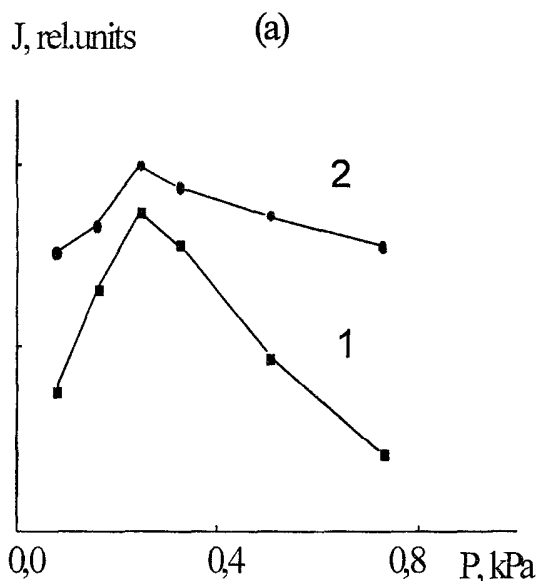


Fig. 1. Dependences of emission intensity of the bands $\lambda=175$ nm ArCl (1) and 258 nm Cl₂* (2) on the content of chlorine molecules ($P_{Ar} = 13,3$ kPa, $U_{ch} = 10$ kV) (a) and argon atoms ($P_{Cl_2} = 0,24$ kPa, $U_{ch} = 10$ kV) (b) in the discharge when operating on the Ar/Cl₂ mixture.

The optimum argon pressure is in the range 15–20 kPa. The greatest brightness of the XeCl (D-X) radiation was observed at reduced pressures of the mixture ($P \leq 3$ kPa). At moderate charging voltage ($U_{ch} \leq 12$ kV)

and $P \geq 30$ kPa, contraction of the TVD was observed. XeCl(D,B-X) , KrCl(B-X) , ArCl(B-X) and $\text{Cl}_2(\text{D}'\text{-A}')$ radiation bands of comparable brightness could be obtained in the regions of low ($P=5\text{--}10$ kPa) and high ($P=20\text{--}30$ kPa) pressures of the Ar/Kr/Xe/Cl_2 mixture.

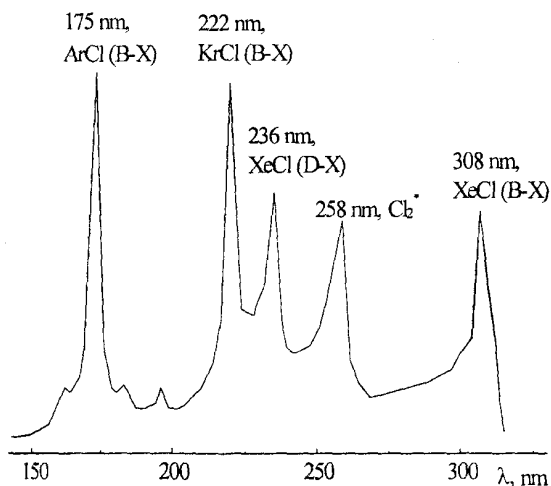


Fig. 2. Emission spectrum of an electric-discharge plasma formed from Ar/Kr/Xe/Cl_2 mixture ($P=15$ kPa, $U_{ch}=12$ kV).

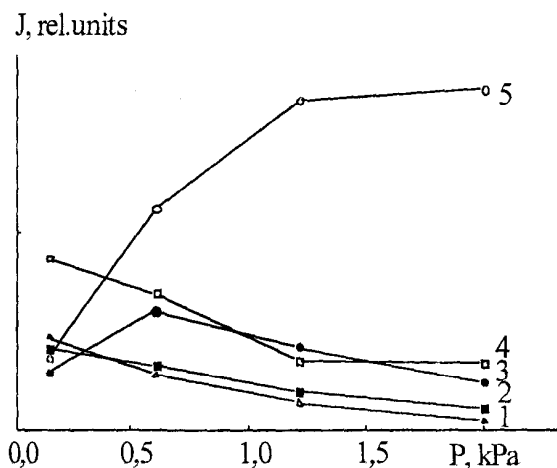


Fig. 3. Dependences of the brightness J of bands with $\lambda=175$ nm (1), 222 nm (2), 236 nm (3), 258 nm (4) and 308 nm (5) in a TVD formed the $\text{Ar/Kr/Xe/Cl}_2=13.3/0.4/P_{\text{Xe}}/0.24$ kPa mixture on the xenon pressure ($U_{ch}=12$ kV).

An increase in Xe pressure (in Ar/Xe/Cl_2 and Ar/Kr/Xe/Cl_2 mixtures) leads to the decrease in the brightness of bands at $\lambda=236$, 222 and 175 nm and to the increase in the

brightness of the band at $\lambda=308$ nm. When the Xe pressure was increased above 0.1 kPa, the saturation of the growth (decrease) of the brightness of all the bands is observed at Xe pressure of 0.5 kPa (Fig. 3). The bands of comparable brightness can be obtained in the investigated mixture at the xenon pressure of 0.2-0.5 kPa.

For the TVD plasma with $p < 10\text{--}20$ kPa the role of the ion-ion recombination ($\text{Ar}^+ + \text{Cl}^- + [\text{Ar}] \rightarrow \text{ArCl(B,C,D)} + [\text{Ar}]$) diminishes, as the pressure decreases from 100 to 10 kPa, since the rate constant for the given reaction decreases by an order of magnitude [5]. In the initial stage of the discharge, the harpoon reactions: $\text{Ar}(^3\text{P}_2) + \text{Cl}_2 \rightarrow \text{ArCl}^* + \text{Cl}$ ($k = 7 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ [6]) predominates. The excitation of the rare-gas atoms in the TVD plasma is important for this reaction.

The radiation-intensity distribution in the TVD plasma is therefore determined to a large extent by the transfer of energy via the Ar-Kr, Xe channel [6, 7], which can also be interpreted as the formation of excimer molecules in a multicomponent active medium.

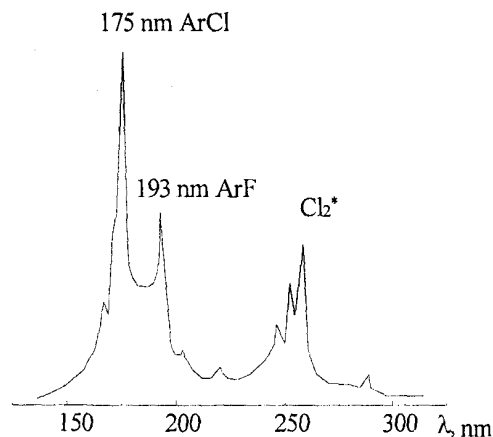


Fig. 4. Spectra emission of plasma TVD in mixtures $\text{Ar/CF}_2\text{Cl}_2=12/0.04$ kPa.

4. TVD in $\text{Ar/CF}_2\text{Cl}_2$ and $\text{He/Kr/Xe/CF}_2\text{Cl}_2$ Mixtures

Plasma radiation spectra in the range 130–150 nm and brightness of the 175-nm

ArCl, 193-nm ArF and 258-nm Cl₂* bands, obtained in the discharge in the mixture of Ar/CF₂Cl₂ = (1-15)/0,008-0,15) kPa were investigated. It was shown that the given TVD was a multiwave source on the ArCl, ArF and Cl₂ (D'-A') molecular transitions (Fig. 4). The optimal content of CF₂Cl₂ molecules was 0.008-0.01 kPa, Ar atoms – 10–15 kPa (Fig. 5). The ratio of the B-X band brightnesses in ArCl and ArF was 10 [8]. This value is approximately equal to the ratio of [Cl] and [F] ion densities, generated in the reaction of dissociative attachment of electrons to CF₂Cl₂ molecules [9]. Radiation of excimer molecules was observed in the immediate afterglow of the transverse space discharge. The radiation intensity in the visible spectral region was lower by approximately two orders of magnitude, compared to the UV region. The basic characteristics of the multiwave excimer emitter (in He/Kr/Xe/CF₂Cl₂ and He/Kr/CF₂Cl₂, He/Xe/CF₂Cl₂ mixtures) are as follows: 222/249/258/308/353 nm; total power of UV radiation ≤5 kW; pulse duration 200-400 ns; pulse repetition rate 1–10 Hz; service life of the gas mixture ≥(2–3)×10⁴ pulses; the working media are He/Kr(Xe)/CF₂Cl₂ and He/Kr/CF₂Cl₂, at a pressure of 10-40 kPa; and the optimum ratio between the concentrations of Kr and Xe atoms in the four-component mixture is (3–8) / (0.2–0.4) kPa.

5. Conclusions

Thus, the study of the conditions for the simultaneous formation of rare-gas chlorides, fluorides and Cl₂* TVD-plasma formed from an Ar/Cl₂, Ar/Kr/Xe/Cl₂, Ar/CF₂Cl₂ and He/Kr/Xe/CF₂Cl₂ mixtures showed that it is a multiwave source of radiation based on the band systems with λ=175/258 nm; 175/222/236/258/308 nm; 175/193 nm and 222/249/258/308/353 nm.

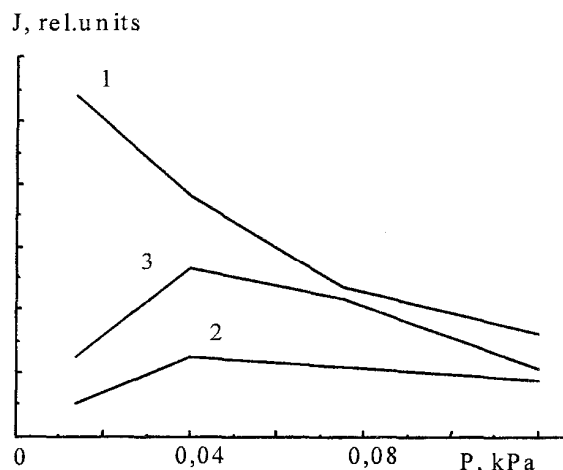


Fig. 5. Dependences of emission brightness of the bands with λ=175 nm ArCl (1), 193 nm ArF (2) and 258nm Cl₂* (3) upon the composition of freon-12 molecule in TVD on mixture Ar/CF₂Cl₂ by P_{Ar}=12 kPa and U_{ch}=10 kV.

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ДОСЛІДЖЕННЯ УМОВ РОБОТИ ТА ХАРАКТЕРИСТИК БАГАТОХВИЛЬОВОГО ВИПРОМІНЮВАЧА НА ХЛОРИДАХ ТА ФТОРИДАХ ІНЕРТНИХ ГАЗІВ

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Подано результати розробки та систематичних досліджень спектральних, часових та ресурсних характеристик електророзрядних ексімерних ламп, що випромінюють на 175 нм ArCl(B-X) / 222 нм KrCl(B-X) / 236 нм XeCl(D-X) / 258 нм Cl₂ (D'-A') / 308 нм XeCl (B-X); 175 нм ArCl(B-X) / 193 нм ArF(B-X); 222 нм KrCl / 249 нм KrF / 258 нм Cl₂ * / 308 XeCl / 353 нм XeF. Активне середовище лампи було сформовано на основі газових сумішей : Ag/Kr/Xe/Cl₂ , Ag/CF₂Cl₂ , He/Kr/Xe/CF₂Cl₂ , які збуджувались за допомогою поперечного розряду з іскровою перейонізацією. Оптимальний тиск суміші Ag/Kr/Xe/Cl₂ складав 15-20 кПа, парціальний тиск хлору був рівним 0.2-0.4 кПа. Для одержання приблизно рівних за величиною яскравостей смуг випромінювання парціальний тиск Kr та Xe повинен бути в межах 0.2–0.5 кПа.

При використанні молекул CF₂Cl₂ як єдиного носія атомів Cl та F було реалізовано багатохвильовий ексімерний випромінювач зі співвідношенням яскравостей хлоридів Kr та Xe до відповідних фторидних молекул як 9/1. Оптимальна концентрація молекул CF₂Cl₂ була рівною 0.008-0.01 кПа (суміш Ag/CF₂Cl₂). Співвідношення між яскравостями В-Х смуг молекул ArCl та ArF дорівнювало 10. Ресурс роботи багатохвильового випромінювача становив 5×10³–1×10⁵ імпульсів. Даний випромінювач може використовуватися в імпульсній фотометрії, хімії високих енергій, біології та медицині.