

On stability of self-sustained volume discharge in working mixtures of non-chain electrochemical HF laser

A.A. Belevtsev, S.Yu. Kazantsev, I.G. Kononov, A.A. Lebedev, S.V. Podlesnykh, K.N. Firsov

Abstract. Burning voltage of a self-sustained volume discharge (SSVD) is studied as a function of the specific energy deposition in SF_5 with $C_2 - H_6$ and H_2 mixtures, which are working media of a non-chain electrochemical HF laser. It is established that the voltage rises linearly with increasing the specific energy deposition, the relative voltage rise in the $SF_6 - C_2H_6$ mixtures being noticeably higher than in pure SF_6 and $SF_6 - H_2$ mixtures. An assumption is suggested and substantiated on determining the role of molecule dissociation by the electron impact leading to the observed voltage rise. From experimental data we have found approximate energy expenditures of producing dissociation fragments including atomic fluorine in a discharge in pure SF_6 : $\bar{E}_d(F) = 5 \pm 1$ eV. The values of E_d well agree with literature data obtained by other experimental methods. A conclusion is drawn that the dissociation process is the main mechanism limiting the current density, which implies SSVD realisation without preliminary gas ionisation in working mixtures of a non-chain HF laser and determines a higher stability of the volume discharge in mixtures of SF_6 with hydrocarbons (deuterocarbons) as compared to mixtures with hydrogen (deuterium). A method is suggested and substantiated for numerical estimation of the limitation effect of the current density and its influence on the SSVD stability.

Keywords: non-chain electrochemical HF laser, SF_6 , self-sustained volume discharge, discharge instability, current density limitation, dissociation.

1. Introduction

A self-sustained volume discharge (SSVD) in dense gases is widely used for pumping gas lasers, which determines a long-standing interest on its studying [1–5]. One of most powerful electric discharge lasers operating in mid-IR wavelength range is a non-chain chemical HF(DF) laser.

A strongly electronegative gas SF_6 is often used as a donor of atomic fluorine in such lasers [4, 5]. The presence of this gas in the mixture limited for many years the possibility to increase the radiation energy of a non-chain HF(DF) laser due to the problems of realising the SSVD [4]. First, it was explained as follows. It is known that the key condition for initiating the SSVD is the presence of primary electrons in a discharge gap. The electron concentration should not be below some minimal value, which guarantees that developing heads of electron avalanches would overlap when a high voltage is applied across the gap [1, 3, 6]. To obtain primary electrons, gas preionisation by hard radiation is employed [1, 6, 7] or preliminary filling the gap by electrons due to their drift in an electric field [2, 8]. These methods are efficient for initiating the SSVD in weakly electronegative gases, for example, in working mixtures of electric discharge CO_2 lasers [2, 9], excimer lasers [10], etc. As far as strongly electronegative gases are concerned including those based on SF_6 , simple analysis shows that none of them is suitable because of high electron losses in attachment processes regardless of the method of preliminary gas ionisation [5, 11]. In other words, according to the established notion on volume discharge formation, obtaining the SSVD in strongly electronegative gases by standard methods is principally impossible. However, it was found [12] that at moderate pressures (10–200 Torr) a self-sustained discharge in SF_6 and gas mixtures comprising it develops in a volume form without preionisation in gaps with both a uniform electric field and strong edge rise of the field under the condition that the cathode surface has small-scale (~ 50 μm) inhomogeneities. Such unusual form of the SSVD was termed ‘self-initiated volume discharge’ in [5, 13].

It was established [14] that such a development of a self-sustained discharge is specific not only for SF_6 , but for a series of other strongly electronegative gases, namely, fluorides, iodides, and chlorides. The dynamics of a self-initiated volume discharge was thoroughly studied in [5, 15] by the example of SF_6 and SF_6 mixtures with H_2 and C_2H_6 , which are the working media for non-chain HF lasers. Usually, a discharge starts at an edge of a discharge gap in a domain of an enhanced local electric field in the form of single or several diffuse channels. As the electric energy deposited into plasma increases, the diffuse channels, which arise first, initiate new channels and the discharge ‘spreads’ spontaneously over the gap from the initial breakdown zone moving normally to the applied electric field at the voltage across the electrodes close to that of the static breakdown. A discovery of this form of breakdown has increased the

A.A. Belevtsev Institute for High Energy Densities, Joint Institute for High Temperatures, Russian Academy of Sciences, ul. Izhorskaya 13/19, 125412 Moscow, Russia;

S.Yu. Kazantsev, I.G. Kononov, A.A. Lebedev, S.V. Podlesnykh, K.N. Firsov A.M. Prokhorov General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, 119991 Moscow, Russia; e-mail: k_firsov@rambler.ru

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output radiation energy of non-chain HF(DF) lasers by more than an order of magnitude, and a further possible increase was demonstrated [4–13].

It was shown [5–13] that the development of the SSVD in the form of a self-initiated volume discharge is explained by mechanisms which exist in strongly electronegative gases and limit the current density in a diffuse discharge channel, preventing all the energy stored in the energy source from flowing through the single channel. The result is that the discharge system tends to form new diffuse channels rather than transfer all the energy through a single channel in the course of increasing the energy deposited into plasma.

It was established [4, 5, 16, 17] that small additives (5%–20%) of some gases to SF₆, for example, hydrocarbons (or deuterocarbons) may noticeably increase the duration of stable burning and the SSVD homogeneity. It seems probable that such additives affect the SSVD stability by limiting the current density. However, no investigations have been performed yet on interrelation between the current limitation effect and SSVD stability in gas mixtures based on SF₆. Obviously, establishing such an interrelation requires understanding the mechanisms that limit the current density. Earlier, electron attachment to vibrationally excited SF₆ molecules [5], electron–ion recombination [15], and dissociation of SF₆ molecules by the electron impact [5] were considered as candidates for such mechanisms. It was shown [18] that electron attachment to vibrationally excited SF₆ molecules produced in SSVD plasma may be neglected. The rate constants for electron–ion recombination (β_{ei}) and for electron detachment from negative ions by the electron impact (k_d) under conditions of the SSVD in SF₆ were found [19, 20] to be close: $\beta_{ei} \approx k_d = (2-3) \times 10^{-7} \text{ cm}^3 \text{ s}^{-1}$. Hence, the influence of these processes on discharge characteristics is mainly compensated. A most probable candidate to the role of the main mechanism responsible for current density limitation in the gas mixtures with SF₆ is dissociation of SF₆ by electron impact, in which more than 80% of the energy deposited into SSVD plasma is consumed [21].

The present work is aimed at search for experimental evidences confirming the assumption that dissociation of SF₆ and other mixture components by electron impact plays the dominating role in the current limiting effect. It is also aimed at interpretation of different character of H₂ and C₂H₆ influence on the SSVD stability in working mixtures of the HF laser.

2. Experimental

The scheme of the experimental setup is shown in Fig. 1. A self-sustained volume discharge was initiated in the electrode geometry rod (cathode) – plane at the electrode separation $d = 48 \text{ mm}$ and gas mixture pressure $p = 9-30 \text{ Torr}$. The cathode was a copper wire 1 mm in diameter in polyethylene isolation. The SSVD with such a cathode develops across the discharge gap in the form of a diffuse channel closed to a single cathode spot [5]. The capacitor C with the capacitance of 0.8–3 nF was charged to a voltage of 13–40 kV, then it discharged through a varied inductivity L across the gap. The discharge was studied in SF₆ and in the mixtures of SF₆ with C₂H₂ and H₂. In order to fix the volume V_{dis} occupied by a SSVD plasma, the transverse dimension of the discharge was limited by a dielectric tube 8.5 mm in diameter (the limited discharge

method [5]). Without limitation, the volume V_{dis} increases as the electric energy deposited into SSVD plasma rises. The SSVD voltage and current were detected by a calibrated voltage attenuator and shunt and then recorded with a digital oscilloscope with the bandwidth of 100 MHz. In most experiments, the total duration of the discharge current (at the level of 0.1) was not longer than 100 ns in order to minimise the influence of reactions with chemically active fluorine atoms, which are produced in SF₆ dissociation by electron impact.

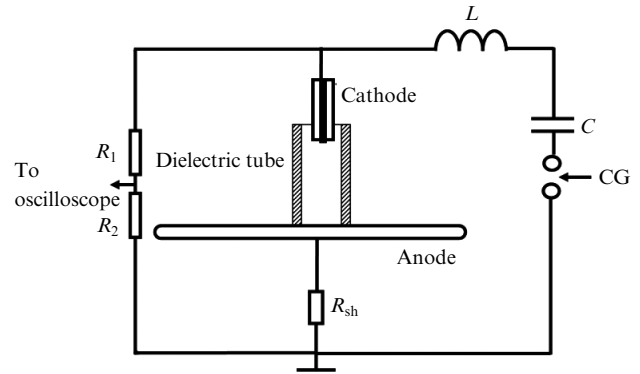


Figure 1. Experimental setup: (CG) controlled spark gap; (R_{sh}) current shunt; ($R_1 - R_2$) voltage divider; (C) capacitor; (L) inductance.

The specific energy deposition into SSVD plasma per molecule, w_{in} , is determined as

$$w_{\text{in}} = \frac{1}{V_{\text{dis}} N_0} \int_0^{t_{\text{max}}} U(t) I(t) dt, \quad (1)$$

where N_0 is the initial total concentration of molecules; $U(t)$ is the voltage across a discharge gap; $I(t)$ is the SSVD current; t_{max} is the time needed for the current to reach a maximum (the duration of the current pulse leading edge). Time is kept from the instant of the gap electrical breakdown.

In the experiments, the dependence $U_{\text{qs}}(w_{\text{in}})$ in a quasi-stationary stage of SSVD burning was taken. The magnitude of U_{qs} was measured at the instant corresponding to a maximum current, $U_{\text{qs}} = U(t_{\text{max}})$; hence, the integral in (1) is taken from zero to t_{max} . Variation in $U_{\text{qs}}(w_{\text{in}})$ with an increase in w_{in} was characterised by a parameter $\Delta = (U_{\text{qs}} - U_{\text{qs}}^0)/U_{\text{qs}}^0$, where U_{qs}^0 is the voltage corresponding to a minimal energy deposition into plasma, which is determined from the experimental dependence $U_{\text{qs}}(w_{\text{in}})$ at $w_{\text{in}} \rightarrow 0$.

The critical reduced electric field strength $(E/N)_{\text{cr}}$ (E is the electric field strength and N is the molecule concentration) responsible for U_{qs}^0 {in strongly electronegative gases the parameter $(E/N)_{\text{cr}}$ is close to E/N in a quasi-stationary discharge burning phase [15]}, was found from the relationship

$$U_{\text{qs}}^0 = (E/N)_{\text{cr}} d N_0 + U_{\text{k}}, \quad (2)$$

where U_{k} is the cathode drop (the thickness of the cathode layer in the considered pressure range may be neglected [22]). With this aim, in gases under study we detected the

dependence of U_{qs}^0 on the parameter pd at low specific energy depositions ($w_{in} < 0.05$ eV mol⁻¹). The pressure of the mixtures varied in the range 6–60 Torr. In these measurements, the SSVD domain was not limited by a dielectric tube. The dependences $U_{qs}^0(pd)$ for all gas mixtures are well approximated by linear functions.

3. Experimental results and discussion

Figure 2 presents the current and voltage oscillograms at two values of w_{in} for the SF₆:C₂H₆ = 5:1 mixture at a considerably long duration of the discharge current. One can see that at the higher energy deposition the voltage oscillogram distorts and U_{qs} increases, which indicates that the mechanisms limiting the SSVD current arise [5]. In Fig. 2b, the voltage oscillogram exhibits an abrupt fall after the current reaches a maximal value. This is related to a greater length of an incomplete spark channel advancing into the gap from the cathode with increasing w_{in} . With a further increase in w_{in} , the time lapse between the charge start and beginning of the voltage drop becomes shorter and finally the SSVD contracts. Hence, the energy deposition w_{in} was limited in the experiments so that voltage oscillograms would not distort. In addition, as was already mentioned the dependences $U_{qs}(w_{in})$ were recorded at a current duration no longer than 100 ns, which also favoured the SSVD at high w_{in} without producing a noticeable incomplete channel.

Figure 3 presents the dependences of U_{qs} versus w_{in} for SF₆ and mixtures of SF₆ with C₂H₆ or H₂. In the range $w_{in} = 0 - 0.7$ eV mol⁻¹ the dependences can be sufficiently well approximated by a linear function

$$U_{qs} = U_{qs}^0 + Kw_{in}, \quad (3)$$

where K is the constant dependent on the gas mixture composition. A noticeable growth of U_{qs} at a higher energy deposition into plasma proves existence of the mechanisms limiting the current density. In the experiments, a single diffuse channel is modelled, whose possibility to expand is limited by the walls of a dielectric tube. In the absence of the tube, the volume occupied by the discharge would grow with the energy deposition due to mechanisms limiting the current density. In the case of a plane electrode geometry, these mechanisms would prevent all the energy stored in a capacitor from flowing through a single channel and would stimulate the discharge system to transfer to a new state with an origin of additional diffuse channels [5, 15].

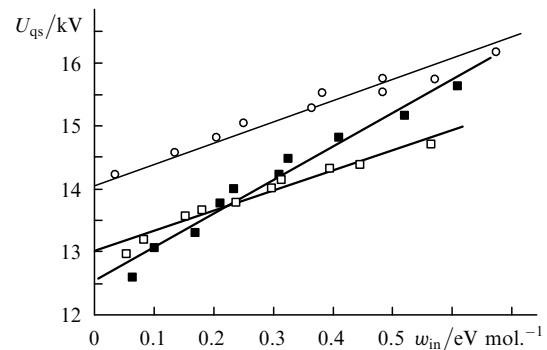


Figure 3. Dependences of U_{qs} on w_{in} at $t_{max} = 30$ ns and $p = 24$ Torr for the SSVD in SF₆ (○), and in the mixtures SF₆:C₂H₆ = 5:1 (■) and SF₆:H₂ = 5:1 (□).

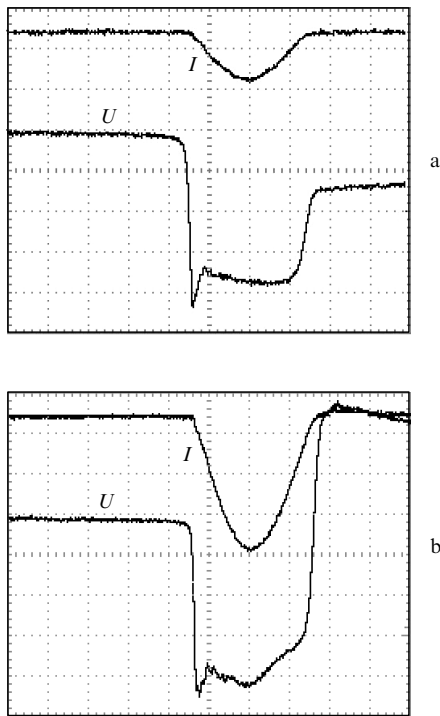


Figure 2. Oscillograms of voltage U and current I for a limited SSVD in the SF₆:C₂H₆ = 1:5 mixture at the pressure $p = 15$ Torr and specific energy deposition $w_{in} = 0.7$ (a) and 2 eV mol⁻¹ (b). Time scale is 50 ns div⁻¹.

As was mentioned, the dissociation of neutral molecules through an electron impact is the most probable mechanism of current density limitation in a diffuse channel. A qualitative estimate of the efficiency of this process in the considered conditions becomes significant.

Assume the mixture comprises S components, each with n_l different dissociation channels. Let each dissociation act of l th component in the i th channel produce m_{li} fragments. Then the rate of an increase in the net concentration $N_d^{(il)}$ for dissociation products in this channel is given by the expression

$$\frac{dN_d^{(il)}}{dt} = \frac{1}{e} m_{li} \eta_d^{(il)}(E/N) J e, \quad 1 \leq i \leq n_l, \quad 1 \leq l \leq S, \quad (4)$$

where

$$\eta_d^{(il)}(E/N) = \frac{k_d^{(il)}(E/N)}{u_e(E/N)} \frac{\delta_l}{E/N}; \quad \delta_l = \frac{N_l}{N}; \quad (5)$$

e is the electron charge; J is the current density; N_l is the concentration of the l th component; $k_d^{(il)}(E/N)$ and $u_e(E/N)$ are the rate of dissociation by the electron impact for the l th component in the i th channel and the electron drift velocity, respectively, which depend on the reduced electric field strength E/N . The introduced parameter $\eta_d^{(il)}(E/N)$ has a simple physical meaning. Indeed, it follows from (4) that $e/\eta_d^{(il)}(E/N)$ is the energy expenditure for a single dissociation act of the l th component in the i th channel. The net concentration over all dissociation fragments produced by the instant of maximal current is

$$\Delta N_d = \frac{1}{e} \sum_l \sum_i m_{il} \int_0^{t_{\max}} \eta_d^{(il)}(E/N) J E dt. \quad (6)$$

At comparatively small specific energy depositions, the reduced electric field strength E/N slightly varies with an increase in the discharge current. In addition, the numerator and denominator in (5) are monotonically growing functions of the parameter E/N so that the parameter $\eta_d^{(il)}(E/N)$ in the particular conditions of our experiments (the energy is deposited into plasma in a quasi-stationary discharge phase at the value of E/N close to constant) weakly depends on E/N . In particular, using the calculation data on kinetic coefficients for SF₆ [21] one can easily see that in pure SF₆ $\eta_d^{(il)}(E/N)$ increases by less than 10% even under the five-fold rise of the reduced electric field strength from 100 to 500 Td. Thus, we may take the term $\eta_d^{(il)}(E/N)$ in (6) outside the integration sign. Finally, in view of (1) we obtain

$$\Delta N_d = \frac{1}{e} w_{\text{in}} N_0 \sum_l \sum_i m_{il} \eta_d^{(il)}(E/N). \quad (7)$$

Let us determine the degree of dissociation α as the excess (as compared to the initial concentration N_0) concentration of particles $\Delta \tilde{N}_d$ in the dissociated gas relative to N_0 : $\alpha = \Delta \tilde{N}_d / N_0$. Since one molecule of the initial component disappears in each dissociation act, we have

$$\Delta \tilde{N}_d = \frac{1}{e} w_{\text{in}} N_0 \sum_l \sum_i (m_{il} - 1) \eta_d^{(il)}(E/N). \quad (8)$$

The energy spent on producing one of the fragments of the initial mixture regardless of their nature is

$$E_d = \frac{w_{\text{in}} N_0}{\Delta N_d} = \frac{e}{\sum_l \sum_i m_{il} \eta_d^{(il)}(E/N)}. \quad (9)$$

The parameters E_d and α are related by the obvious expression

$$E_d = \frac{w_{\text{in}}}{\alpha} \xi, \quad \xi = 1 - \frac{\sum_l \sum_i \eta_d^{(il)}}{\sum_l \sum_i m_{il} \eta_d^{(il)}}. \quad (10)$$

To determine the parameter ξ we need sufficient information about all dissociation channels, which can hardly be found presently. However, one may expect that in such mixtures, as it follows from (10), ξ would slightly differ from unity.

We may express E_d in terms of the parameters measured in the experiments assuming that the SSVD burning voltage rise at higher energy depositions is only caused by dissociation. According to (2) and (3) we have

$$(E/N) dN_0 (1 + \alpha) + U_k = U_{\text{qs}}^0 + K w_{\text{in}}. \quad (11)$$

In a dissociating gas, the reduced electric field strength in a discharge may, generally speaking, differ from the critical value $(E/N)_{\text{cr}}$ in the absence of dissociation: $E/N = (E/N)_{\text{cr}} (1 + \varepsilon)$. At small degrees of dissociation ($\alpha \ll 1$) this distinction, however, is small and to a sufficient accuracy we may assume $|\varepsilon| \ll 1$. In view of this fact from (2) and (11) we may write the expression

$$(E/N)_{\text{cr}} (\varepsilon + \alpha) dN_0 = K w_{\text{in}}. \quad (12)$$

The experimental data obtained give information about neither the sign nor absolute values of ε . We can only speak about an approximate estimation of α or the ratio E_d/ξ . In particular, neglecting ε as compared to α we arrive at the expression

$$\frac{w_{\text{in}}}{\alpha} = \frac{(E/N)_{\text{cr}} dN_0}{K}. \quad (13)$$

The values of w_{in}/α found under this assumption from the experimental data are presented in Table 1 and allow one to make some principal conclusions.

First, the degree of dissociation α at noticeable energy depositions (see Fig. 3) may exceed 10%. In other words, indeed, in the considered conditions the dissociation of mixtures under study by electron impact occurs quite efficiently.

Second, one can see that w_{in}/α strongly depends on the gas composition. For the mixtures of SF₆ with ethane, it is sufficiently lower than for pure SF₆ or for a mixture of SF₆ with hydrogen. This, seemingly, is related to the fact that in dissociation process, C₂H₆ molecules may decay to more than two fragments (see relationship (8)). In addition, the dissociation constant for H₂ molecule by electron impact is considerably small since the total dissociation cross-section reaches a maximum of $\sim 8 \times 10^{-17}$ cm² only at the electron energy of ~ 15 eV, and the efficient dissociation mechanism through excitation of vibration levels has no time to 'switch on' during the discharge process [23]. At higher relative contents of ethane (greater δ_l), the parameter w_{in}/α in accordance with (8), should fall, because the dissociation rate constant of $k_d^{(il)}(E/N)$ for ethane in the considered conditions is surely greater than for SF₆. This completely agrees with the data from Table 1.

Table 1. Values of the parameter w_{in} for SF₆ and SF₆-H₂ and SF₆-C₂H₆ mixtures.

Mixture	$\frac{w_{\text{in}}}{\alpha}$ / eV mol. ⁻¹
Pure SF ₆	5 ± 1
SF ₆ :H ₂ = 5:1	5.2 ± 1.1
SF ₆ :C ₂ H ₆ = 5:1	3.1 ± 0.9

The following circumstance substantially justifies the assumptions made in estimating α . According to (24) in the case of the SSVD in pure SF₆, the dominating dissociation channel is



In (24) for these conditions we experimentally determined the energy expenditure $\tilde{E}_d(\text{F})$ of producing a fluorine atom, which is equal to the ratio of the specific energy deposition $w_{\text{in}} N_0$ to the total concentration of fluorine atoms produced in the dissociation process $\Delta \tilde{N}_d(\text{F})$. Since according to reaction (14), we have $\Delta \tilde{N}_d(\text{F}) = \Delta \tilde{N}_d$ then in view of relationship (8) we obtain

$$\tilde{E}_d(\text{F}) = \frac{w_{\text{in}} N_0}{\Delta \tilde{N}_d(\text{F})} = \frac{w_{\text{in}} N_0}{\Delta \tilde{N}_d} = \frac{w_{\text{in}}}{\alpha}. \quad (15)$$

In [24] and [25] $\tilde{E}_d(\text{F}) = 4_{-0.8}^{+1.2}$ and 4.5 eV were obtained, respectively, which is in good agreement with our result: $w_{\text{in}}/\alpha = 5 \pm 1$ eV mol.⁻¹ (see Table 1). This implicitly

confirms the dominating role of dissociation reaction (14) in pure SF₆.

Note that the energy expenditure E_d of producing a single fragment in reaction (14) is somewhat less. Indeed, in this case $S = 1$, $m_{il} = 3$, $\xi = 2/3$. By using the data from Table 1 we obtain from (10) $E_d \approx 3.3$ eV.

For mixtures of SF₆ with C₂H₆, the values of w_{in}/α presented in Table 1 cannot be used for determining the energy expenditure of producing fluorine atoms because of the lack of detailed information about dissociation channels. However, these give a reasonable view of some average energy expenditure on producing a dissociation fragment regardless of its nature.

In Fig. 4, the parameter $\Delta = (U_{qs} - U_{qs}^0)/U_{qs}^0$ calculated from the data of Fig. 3 is shown versus w_{in} . One can see that Δ for the mixture SF₆-C₂H₆ is, generally, larger and increases with w_{in} more rapidly than for SF₆ and for the mixture SF₆-H₂. Within the accuracy of linear approximation (3) we have

$$\Delta = \frac{K}{U_{qs}^0} w_{in}, \quad (16)$$

i.e., Δ increases proportionally to w_{in} .

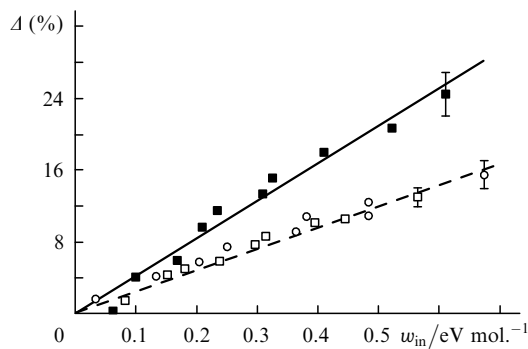


Figure 4. Dependence of parameter $\Delta = (U_{qs} - U_{qs}^0)/U_{qs}^0$ on w_{in} at $p = 24$ Torr and $t_{max} = 30$ ns for the SSVD in SF₆ (○) and in the mixtures SF₆:C₂H₆ = 5:1 (■) and SF₆:H₂ = 5:1 (□).

In our experiments we also established that in the mixtures of SF₆ with hydrocarbons at constant w_{in} the value of Δ increases with a percentage of hydrocarbon in the mixture (however, not above 50%), and the increase (reduction) in Δ correlates with an increase (reduction) in the SSVD stability. Note that Δ and w_{in}/α explicitly correlate: at the same w_{in} , the smaller w_{in}/α the greater the parameter Δ . To this end, the easier the mixture dissociates in SSVD plasma, the greater is the relative increase in the SSVD burning voltage related to the current density limitation. Actually, the parameter Δ determines the possibility of initiating the SSVD in the form of a self-sustained volume discharge and, in the same other conditions, its stability factor.

Note that the linear dependence of the experimental parameter U_{qs} and Δ on w_{in} , the theoretical consideration given above, and the good agreement of the energy expenditure of producing a fluorine atom in the present work with the corresponding data obtained by other experimental methods [24, 25] confirm the great role of the dissociation of SF₆ and other components in working

media of HF lasers by an electron impact in the effect of current density limitation.

Thus, from the experimental material obtained we may conclude that in the mixtures of SF₆ with C₂H₆, the effect of current density limitation, related to molecule dissociation in SSVD plasma, is more pronounced than in pure SF₆ or in its mixtures with hydrogen. Hence, new diffuse channels in SSVD developing in SF₆ mixtures with hydrocarbons in a plane-electrode system should arise at a lower electric power deposited into the plasma of a previously created channel than in the mixtures with a higher molecule dissociation energy. The resulting number of the channels should increase, i.e., the SSVD homogeneity should become better and its stability (the duration of stable burning) should rise due to a reduction in the current flowing through a separate cathode spot. It seems that these factors are responsible for higher stability and homogeneity of the SSVD in the mixtures of SF₆ with hydrocarbons (deuterocarbons) than in the mixtures of SF₆ with hydrogen (deuterium). These facts were mentioned earlier [4, 5, 16, 17] and are also confirmed by the experiments of the present work.

4. Conclusions

Thus, we have established that in the gas mixtures comprising SF₆ including working mixtures of a non-chain HF laser the SSVD burning voltage actually linearly rises with the energy deposited into plasma.

An approximate energy expenditure of producing dissociation fragments in working mixtures of HF lasers was found from experimental data under the assumption that the molecule dissociation by the electron impact plays a main role in the increase in the SSVD burning voltage. The energy expenditure of producing a fluorine atom in an electric discharge in pure SF₆ was determined, which well agrees with the results obtained by other methods [24, 25].

It is found that ethane added to SF₆, in contrast to addition of hydrogen, enhances the effect of current density limitation. It considerably rises the rate of the voltage increase across SSVD plasma at a higher energy deposited into plasma due to the dissociation of hydrocarbon molecules by the electron impact at large values of E/N . The latter are determined by the characteristics of SF₆ as the main mixture component. We may assume that the enhanced effect of current density limitation is the main reason for the increased SSVD stability and homogeneity in the cases where hydrocarbons are added to SF₆.

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