

# Features of Gaseous Mixtures Combustion Initiated by High-Current Slipping Surface Discharge

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**Experimental investigation of decomposition of chlorofluorocarbons (CFCs) contained in  $H_2 + O_2$  or  $CH_4 + O_2$  gaseous mixtures, combustion of which is initiated by high-current pulse slipping surface discharge, has been carried out. It was found that a ternary gaseous mixture of  $H_2 + O_2 + CFC$  or  $CH_4 + O_2 + CFC$ , even with a large amount of the third component, maintains the explosion combustion property inherent in a binary  $H_2 + O_2$  or  $CH_4 + O_2$  mixture, based on the branched chain processes. High-efficient (near-complete) destruction of chlorine-fluorine containing substances has been demonstrated. Production rate of the investigated reactor with respect to the CFC decomposition is of the order of 200 kg/h. Combustion dynamics was investigated. It was shown that initiation by slipping surface discharge combustion propagation behavior has some peculiarities that are beyond the ordinary combustion wave or detonation wave mechanisms. Peculiarities of the observed phenomena may be attributed to the strong UV irradiated by a high-current slipping surface discharge.**

## Nomenclature

$L_c$	=	length of reactor chamber, cm
$M$	=	molecular weight
$p_p$	=	partial pressure, torr
$S$	=	reactor chamber cross section, $cm^2$
$v$	=	gas flow velocity, cm/s
$W$	=	energy cost of chlorofluorocarbons decomposition, kW · h/kg
$\eta$	=	productivity of reactor, kg/h
$\tau_c$	=	burnup time, s
$\Phi_c$	=	diameter of reactor chamber, cm

## Introduction

**I**N recent years the problem of decomposing chlorine- and/or fluorine-bearing substances has assumed special importance in connection with the role that these chemical compounds play in the progressively deteriorating ecological situation throughout the world. We mention freons destroying the ozone layer ( $CF_2Cl_2$ ,  $CFCl_3$ , etc.), compounds with a high greenhouse potential ( $CF_4$ , etc.), chemical warfare agents stockpiled in a number of countries, and so on.

The present work discusses the outcomes of the experimental studies on the decomposition of chlorine- and fluorine-bearing substances in a reactor, where the combustion in the  $H_2 + O_2 + CFC$  or  $CH_4 + O_2 + CFC$  is initiated by an electrical discharge. The purpose of such research is to search for ways to reduce essentially the energy cost of destruction as well as to increase the reactor capacity through involving a source of chemical power, that is, energy released in combustion of a mixture.

Combustion dynamics was investigated. It was shown that initiation by slipping surface discharge combustion propagation behavior

in  $H_2 + O_2$ ,  $H_2 + O_2 + CFC$ ,  $CH_4 + O_2$ , and  $CH_4 + O_2 + CFC$  gaseous mixtures has some peculiarities that are beyond the ordinary combustion wave or detonation wave mechanism.

## Scheme of the Experiment

The scheme of the reactor based on electrodischarge initiation of combustion is shown in Fig. 1.

The chamber of the reactor (1 in Fig. 1) represents a cylindrical quartz pipe of a diameter  $\Phi_c \approx 50$  mm and a length  $L_c \approx 100$ –200 mm. The chamber is evacuated up to a pressure of  $p_0 < 10^{-2}$  torr and filled with a working gas at pressures of  $100 \leq p \leq 500$  torr. It is in this gas mixture that combustion is initiated with the help of a discharger (2 in Fig. 1).

The discharger represents a multielectrode system fixed in a particular way<sup>1–4</sup> on a dielectric (crystal, Teflon®, ceramics) tube of a diameter  $\Phi_d \approx 6$  mm. The length of the metal-dielectric cylindrical discharge system is  $L_d \approx 40$  mm. The discharger is located close to one of the end flanges of the reactor chamber. When a high-voltage pulse is applied to the discharger, it gives rise to the formation of a high-current ( $I \leq 1$ –10 kA), low-threshold, slipping discharge along the discharger and an extended plasma layer at its surface. Such discharge systems have been widely applied at the General Physics Institute of the Russian Academy of Sciences for generating dense, hot collisionless plasma,<sup>1</sup> as sources of the metal plasma,<sup>2</sup> for excitation of converging toroidal shock waves in gas medium,<sup>3,4</sup> and so on.

The present work is the first attempt to use dischargers on a slipping surface discharge for initiating combustion in a gas mixture. In the work, two generators of high-voltage pulses were used as power supplies of dischargers (3 in Fig. 1), with the following parameters:

- 1) Generator  $G_1$  with low pulse energy has pulse amplitude  $U \approx 40$  kV, pulse duration  $\tau \approx 40$  ns, and energy in a pulse  $E \leq 0.1$  J.
- 2) Generator  $G_2$  with high pulse energy has  $U \approx 20$  kV,  $\tau \approx 20$   $\mu$ s, and  $E \leq 30$  J.

The concentration of freon and a number of products of its decomposition was determined by absorption spectroscopy using spectrophotometers IKS-29 and ZEISS SPECORD M80. The wave number spectrum of these devices is 200–4000  $cm^{-1}$  with the resolution  $\leq 0.5$   $cm^{-1}$  in the band 400–4000  $cm^{-1}$  and  $\leq 0.8$   $cm^{-1}$  in the range 200–400  $cm^{-1}$ . After the working mixtures were treated by ignited combustion, they were pumped over into a diagnostic dish, and the resulting products were examined by means of spectrophotometers from the absorption of the diagnostic radiation in the infrared spectral region corresponding to wave numbers from 400 to 1400  $cm^{-1}$ . The absorption cross sections for the corresponding spectral lines of  $CF_2Cl_2$ ,  $CF_4$ ,  $SiF_4$ , and  $SiCl_4$  were

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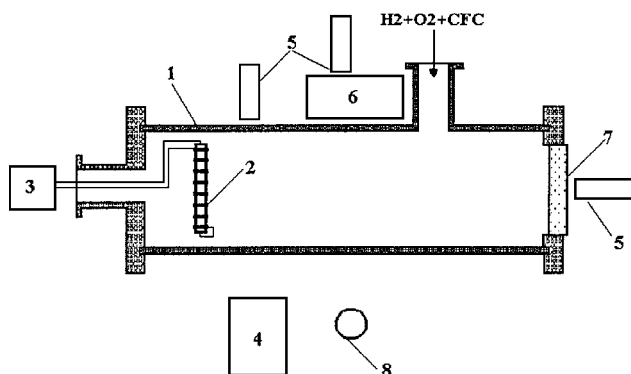
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**Table 1** Results of CFC decomposition measurements in  $H_2 + O_2 + CFC$  gaseous mixture

Mixture contents	Components ratio	Overall pressure, torr	Igniting pulse, energy, J	Degree of CFC decomposition, %
$H_2 + O_2 + CF_2Cl_2$	1:1:1	182	10	$98.7 \pm 0.4$
$H_2 + O_2 + CF_2Cl_2$	1:1:1	182	0.1	$98.5 \pm 0.5$
$H_2 + O_2 + CF_4$	1:1:0.5	170	10	89
$H_2 + O_2 + CF_4$	2:1:1	243	10	94
$H_2 + O_2 + CF_4$	1:1:0.5	152	0.1	40
$H_2 + O_2 + CF_4$	2:1:0.5	213	0.1	94
$H_2 + O_2 + CF_4$	2:1:1	243	0.1	91

**Table 2** Results of CFC decomposition measurements in  $CH_4 + O_2 + CFC$  gaseous mixture<sup>a</sup>

Mixture contents, torr	Degree of freon component decomposition, %
$CH_4:O_2:CF_2Cl_2 = 30:60:60$	83
$CH_4:O_2:CF_2Cl_2 = 30:60:15$	96
$CH_4:O_2:CF_2Cl_2 = 60:120:60$	99.4
$CH_4:O_2:CF_2Cl_2 = 60:120:90$	97.8
$CH_4:O_2:CF_2Cl_2 = 60:120:120$	90
$CH_4:O_2:CF_2Cl_2 = 15:30:9$	95.3
$CH_4:O_2:CF_2Cl_2 = 30:60:60$	90
$CH_4:O_2:CF_2Cl_2 = 15:30:15$	95.4

<sup>a</sup> Igniting pulse energy 10 J.**Fig. 1** Scheme of experiment: 1, metallic or quartz chamber; 2, discharger initiating combustion; 3, high-voltage power supply; 4, streak camera PHER-7; 5, photomultipliers; 6, monochromator MDR-23; 7, quartz or KRS-6 window; and 8, UV lamp.

measured with an allowance for the gas pressure and the degree of absorption.

Dynamics of the reactor glow was investigated with the use of a photoelectric recorder (streak camera) PHER-7 (4 in Fig. 1). Time characteristics of the glow, integrated on the volume of the chamber and on the spectrum, were determined with the help of a photomultiplier FEU-106 (5 in Fig. 1). Viewing windows were made of quartz or of KRS-6 (TlBr + TlCl) (7 in Fig. 1).

## Results

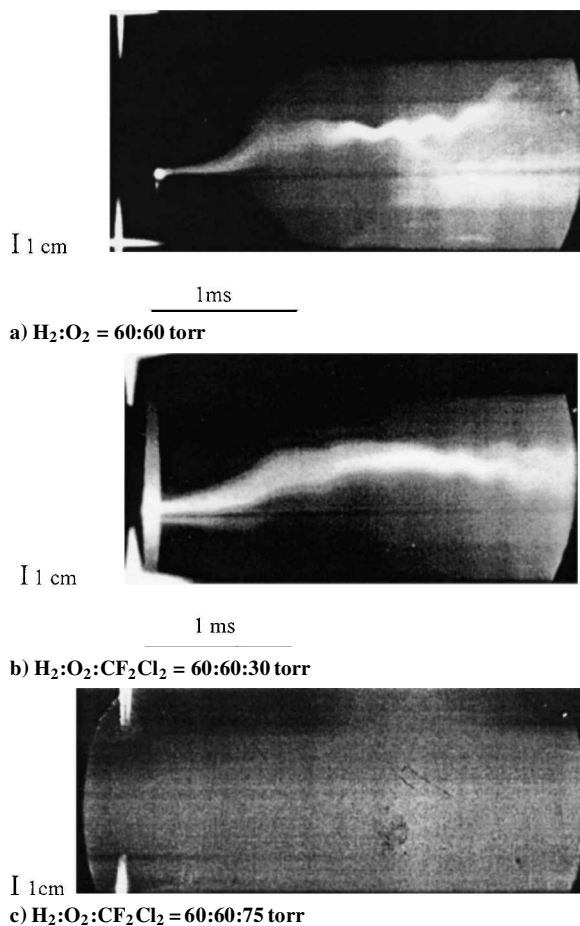
Excitation of a surface slipping discharge in the flammable gas mixture gives rise to the combustion of the gas mixture throughout the chamber volume and leads to changes in its chemical structure. The efficiency of the freon component plasmachemical decomposition is inferred by the characteristic results shown in Tables 1 and 2. Tables 1 and 2 show data on the degree of decomposition of two types of freon: freon-12 ( $CF_2Cl_2$ ) and freon-14 ( $CF_4$ ) for different mixture ratios, different initial pressures, and two different values of energy level in the pulse igniting the slipping surface discharge. In every case of exciting a surface discharge, the gas mixture combustion causes (in the majority of experiments, virtually complete) freon decomposition. Freon destruction was recorded over a broad range of pressures and hydrogen, oxygen, methane, and freon mixtures.

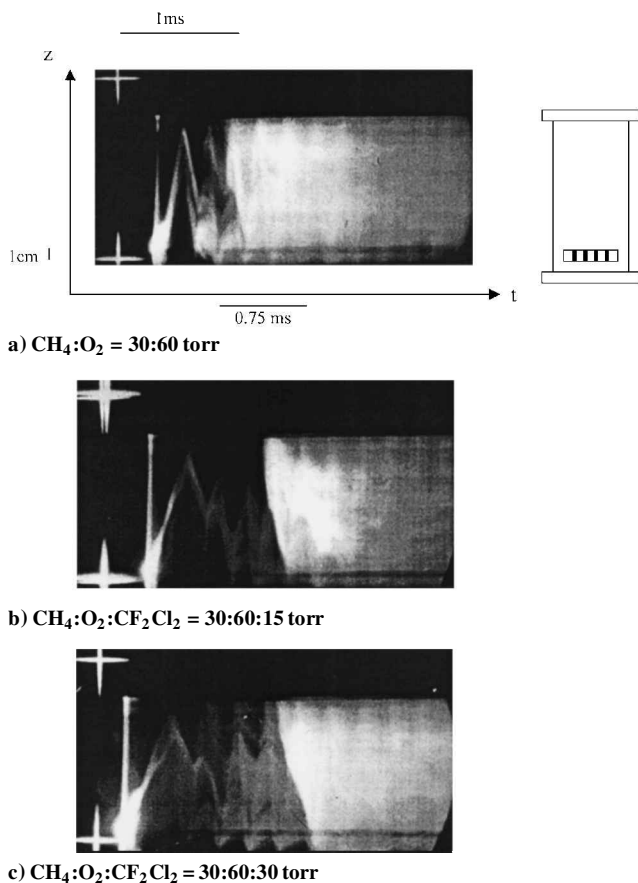
As the part of an exploration program,  $SiF_4$  and  $SiCl_4$  have been also used as a third component of the mixtures. It was shown that, as in the case of the mixtures containing freon-12 and freon-14, initiation of combustion by a discharge was accompanied by virtually complete destruction of the initial fluorine- and/or chlorine-bearing component. Experiments with  $SiF_4$  and  $SiCl_4$  have been performed only once. Therefore, data concerning these freons' decomposition are not included in Table 1, which contains results of repeated experiments.

The characteristic time relations of the mixture combustion are exhibited in a signal from the photomultiplier. It records the glow from the end face of the chamber integrated over the spectrum and the volume. At the origin of the beam sweep, a short splash of the glow is observed, the source of which is a breakdown of the metal-dielectric discharger. Then after some delay ( $\sim 2-3$  ms), which is much longer than the lifetime of a plasma formation ( $\sim 1-5$   $\mu$ s), a long-lived (10–100 ms) glow from the reacting volume of the gas mixture appears. This glow builds up rather slowly, reaches its peak, and then slowly decreases. Apparently, it is precisely this glow, which is characterized by the lifetime  $\Delta\tau$  determined by a half-height of a long-lived part of a pulse, that accounts for the process of the gas mixture combustion. The characteristic lifetime of the postdischarge glow  $\Delta\tau$  depends on the properties of the discharge initiating the combustion, decreasing with the increase of the energy in the pulse igniting the slipping surface discharge.

The dynamics of the glow from the working gas mixture exposed to an electrical discharge, which was localized at the end flange, was investigated with the help of the photoelectric recorder (streak camera) PHER-7. The slot of the recorder was placed along the axis of the chamber, creating a field of view area of length  $\Delta Z \approx 90$  mm and width  $\Delta Y \approx 5-6$  mm.

Characteristic perograms are shown in Figs. 2 and 3. The recorded light signal displacement along the  $X$  axis represents a

**Fig. 2** Characteristic photographs taken by streak camera under the combustion initiation in  $H_2 + O_2$  or  $H_2 + O_2 + CFC$  mixtures, generator  $G_1$ .



**Fig. 3** Characteristic photographs taken by streak camera under the combustion initiation in  $\text{CH}_4 + \text{O}_2$  and  $\text{CH}_4 + \text{O}_2 + \text{CFC}$  mixtures, generator  $G_2$ .

time displacement, whereas that along the  $Z$  axis represents an axial displacement. The full duration of the time sweep in a frame is  $\Delta\tau_p \cong 3$  ms.

The starting time of the streak camera is selected to catch the time of excitation and existence of the electrical discharge initiating the combustion in the reactor. A vertical glowing band in the left part of the pherograms corresponds to the lifetimes of a discharge on the initiating discharger.

The streak camera is adjusted to record the glow of one of the discharge gaps of the multielectrode device on the electrodischarge initiator. It is precisely the bright glow in the center of the band that represents the interelectrode space glow at the surface of the initiator.

In Fig. 2, characteristic photographs taken by the streak camera for combustion initiation with high-voltage pulse generator  $G_1$  are presented for the three cases:  $\text{H}_2:\text{O}_2 = 60:60$  torr (Fig. 2a),  $\text{H}_2:\text{O}_2:\text{CF}_2\text{Cl}_2 = 60:60:30$  torr (Fig. 2b), and  $\text{H}_2:\text{O}_2:\text{CF}_2\text{Cl}_2 = 60:60:75$  torr (Fig. 2c). In Figs. 2a and 2b the streak camera started recording at the same time as a high-voltage pulse was applied to the electrodischarge initiator. In Fig. 2c the PHER-7 was started 3 ms after starting the electrodischarge initiation, demonstrating the uniformity of luminosity well into the postdischarge time. In Fig. 3, characteristic photographs for the  $G_2$  generator supplying the discharger are shown; the compositions are  $\text{CH}_4:\text{O}_2 = 36:60$  torr (Fig. 3a),  $\text{CH}_4:\text{O}_2:\text{CF}_2\text{Cl}_2 = 30:60:15$  torr (Fig. 3b), and  $\text{CH}_4:\text{O}_2:\text{CF}_2\text{Cl}_2 = 30:60:30$  torr (Fig. 3c).

## Discussion

Electrical discharge is one of the most widely used techniques for initiating combustion in gaseous combustible mixtures. It has been comprehensively discussed in a series of monographs.<sup>5,6</sup> As a rule, electric discharges represent a relatively low-power spark excited between two electrodes immersed in combustible gas medium

(analogous to a spark in internal combustion engines). In the present work, a high-current pulse slipping surface discharge was used instead of a spark.

A comparison with already published data points out, in the performed research based on using a new high-power electrodischarge device, results that are of obvious interest for physics (physicochemistry), as well as for actual applications.

The mere fact of the fast combustion of a three-component gas mixture is very interesting per se. A two-component mixture (oxygen-hydrogen or oxygen-methane), such as the combustible gas, has been studied quite extensively (for example, Refs. 6–9). The ignition (with the fast, explosive, energy release) of an oxygen-hydrogen or oxygen-methane mixture occurs by chain branching processes described by well known and thoroughly developed reaction schemes.<sup>6–8</sup>

Addition of a third component (freon,  $\text{SiF}_4$ ,  $\text{SiCl}_4$ ) with a partial pressure comparable to that of oxygen and hydrogen, renders the chain process questionable because the availability of a significant number of chlorine- or fluorine-bearing molecules may interrupt the chain of interactions between the elements of the two-component mixture. However, experiments have shown that addition of freon (chlorine or fluorine bearing) maintains the process of chain ignition.

Thus, the mere existence of fast combustion in a three-component mixture may be regarded as novelty.

The dynamics of combustion initiated by a pulse surface slipping discharge is of interest in the study of the physics of the observable phenomena. From the high-speed photography, one observes that discharge on the initiator gives rise to a glow wave lifting off the discharger and going into the surrounding gas medium. At the beginning of the process, the wave is propagated in a direction orthogonal to the initiator, with a virtually constant velocity. However, at some distance  $\Delta Z_1$  from the discharger, the wave slows down. Then, after a small time interval, a glow flash along the whole length of the reactor chamber is observed.

Such is the scenario of the gas medium response to a pulse discharge at the initial stage preceding the basic process of the gas mixture combustion in the volume of the reactor. The time intervals spanned by the initial stage do not exceed 3 ms and are comparable to the time delay of the initiation of the glow recorded by the photomultiplier and attributed to the volume combustion of a mixture. The time of the combustion itself significantly exceeds the lifetime of the initial glow wave, and depending on the energy contribution to a discharge initiating the combustion, is up to 10–90 ms.

The observable glow wave is not a detonation wave because the measured propagation velocity is much less than the detonation velocity in a binary mixtures (which, according to Refs. 10 and 11, amounts to some  $10^5$  cm/s) and, in the case of the low-energy initiation, is even less than the velocity of sound in a gas medium.

For the case of high energy, the wave may hardly be attributed to the combustion waves described in the literature because velocities of those waves at standard ignition are lower than those recorded in the present experiment and strongly depend on even a small percentage of a third gas admixture to hydrogen-oxygen or methane-oxygen mixtures (for example, see Ref. 7).

The other part of this scenario is even more unusual because, once the initial glow has halted, a flash along the whole length of the chamber is observed, followed by the more or less homogeneous combustion of the mixture as a whole.

Application of the high-current initiator on a slipping surface discharge significantly changes the process of ignition of a gaseous volume remote from the initiator (as compared with standard low-power, two-electrode spark ignition or ignition with a heated metal filament).<sup>5,10</sup> The standard dynamics of combustion described by thermal or detonation waves propagating from a point of local heating (or a breakdown) of a gas medium is replaced (in initiating a high-current slipping discharge) by a complicated sequence of phenomena. The sequence consists in exciting a wave that lifts off the initiator and halts in the sufficient vicinity of the initiator, with the subsequent homogeneous combustion in the whole volume of the reactor. In this case, the glow wave cannot be identified either with a combustion wave or with a detonation wave.

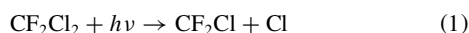
The combustion time for a gas mixture depends mainly on the energy released at initiation, decreasing with the increase of it (40–90 ms for a low-energy generator and 5–10 ms for a high-energy generator). This property of combustion initiated by a high-current surface slipping discharge is also of interest from the standpoint of fundamental physics. At the same time, the detected phenomenon opens up fresh opportunities for applications, permitting the influence of the combustion rate of an oxygen–hydrogen mixture through the parameters of an initiator.

Analyzing the results implies that the radiation of the high-current surface discharge plays a decisive role in the observable pattern. The intensity of this radiation and its spectrum should differ from the intensity and spectrum of a spark commonly used as an initiator of combustion in gas mixtures.

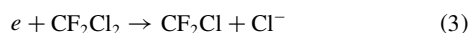
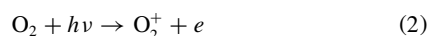
The specificity of a high-current ( $I \cong 100\text{--}5000\text{ A}$ ) slipping discharge as an emanating object has been outlined earlier in a series of publications. In particular, in Ref. 12, it is observed that there is a high fraction of a far UV component in the slipping discharge spectrum, which leads to the formation of an anomalously long-lived photoionized plasma in the gas medium surrounding the discharge. In Ref. 13, highly effective decomposition of freon in the volume of a gas mixture filling the reactor (argon plus freon and air plus freon) was also explained by the singularities of the slipping discharge radiation leading to photoionization, photoexcitation, and photodissociation of the gas remote from the energy release area.

The ability of a high-current surface discharge to affect significantly the properties of gas medium in a volume that considerably exceeds the volume occupied by a plasma formation may be regarded as one of the peculiarities of this type of gas discharge.

For the experiments with the three-component mixtures, it seems probable as well that during its short lifetime (from a few tens of nanoseconds up to several microseconds) a high-current discharge pressed to the surface of the discharger leads not only to the initiation of plasmachemical phenomena at its immediate vicinity, but also, through a UV radiation component, to the modification of the gas medium characteristics virtually in the whole volume of the reactor. For example, the flash of UV radiation transforms the initial mixture of molecules  $\text{H}_2$ ,  $\text{O}_2$ , and  $\text{CF}_2\text{Cl}_2$  (or any other freon) into a new mixture containing, among the initial components, new ones that are extremely active chemically. Thus, for example, a series of processes may give rise to atomic chlorine (or negative ions of chlorine). Among these processes, the reaction of direct photodissociation must be mentioned:

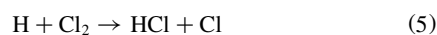


as well as a series of indirect processes related to photoionization, such as



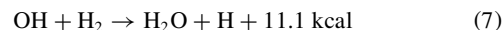
UV radiation may also lead to the photodissociation of  $\text{O}_2$  and production of electron-excited metastable atoms of oxygen.<sup>14</sup> There is also a possibility of direct or indirect dissociation of hydrogen molecules.

All of the preceding processes [Eqs. (1–3)] may increase the probability of ignition, up to the point of spontaneous ignition, of the three-component mixture. Thus, the production of atomic chlorine may give rise to a new chain cycle, which is complementary to the oxygen–hydrogen one, namely,<sup>6</sup>



Production of atomic hydrogen and atomic oxygen may initiate a thermal explosion in the hydrogen–oxygen mixture.<sup>6,9</sup> The role of primary centers of ignition may involve hydrogen atoms emerging

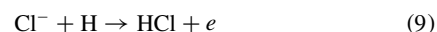
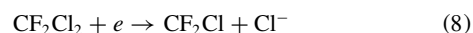
due to photochemical processes and participating in a catalytic cycle characterized by a significant energy release:



Thus, in the volume of the reactor, available to UV radiation generated by a high-current slipping surface discharge, combustibility of the mixture is increased up to the possibility of spontaneous ignition.

Photoexcitation of molecular oxygen and its deexcitation during the propagation of hydrodynamic perturbation from the discharger surface may also be related to a glow wave observed in the initial phase of the process preceding the combustion.

When analyzing chemical and discharge kinetics of the reactor, one has to bear in mind the possibility of a catalytic process involving an electronic component, the role of which is fast and deep decomposition of freons.<sup>15</sup> We mean, for example, the following cycle of reactions:



The already described experiments are independent of value, pointing toward the possibility of creating a high-performance reactor for studying both ozone-depleting freons and freons with a high greenhouse potential.

Basing on the results of the present experiment, it is easy to evaluate efficiency of a reactor representing a discharger of a described type placed in a cylindrical chamber of a cross section  $S$  and length  $L_c$ . For this purpose we may use a simple relationship,

$$\eta \approx 2 \cdot 10^{-7} p_p \cdot M \cdot S \cdot L_c / \tau_c \text{ kg/h} \quad (10)$$

For  $p_p = 100 \text{ torr}$ ,  $S = 20 \text{ cm}^2$ ,  $L_c = 50 \text{ cm}$ , and  $\tau_c = 10^{-2} \text{ s}$ , we obtain

$$\eta \approx 200 \text{ kg/h}$$

In this case, the consumed electrical power will not exceed 50 W. That means that the energy cost of decomposition, that is, expenditures of electrical energy, will be approximately

$$W \approx 2 \cdot 10^{-4} \text{ kW} \cdot \text{h/kg}$$

To ensure such a high efficiency of the reactor, the gas flow velocity through its chamber should be equal to

$$v \approx L_c / \tau_c \approx 5 \times 10^3 \text{ cm/s} \quad (11)$$

which is quite simple to accomplish.

## Conclusions

Performance studies similar to those described in this paper in various gaseous-phase combustible mixtures, including a methane–air mixture, are of current interest. The possibility of explosive utilization of chlorofluorocarbons in  $\text{H}_2\text{--O}_2$  or  $\text{CH}_4\text{--O}_2$  mixtures will make the plasmachemical reactor cheaper.

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