Tetrafluoroethylene as a fuel component for an atomic fluorine generator of a supersonic continuous-wave chemical HF laser

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Abstract. **A cheap and widely accessible combination (one of non-saturated perfluorocarbons), i.e. C_2F_4 tetrafluoroethylene, is proposed as an alternative fuel component for an atomic fluorine generator of a supersonic cw chemical HF laser of self-contained type. Combustion products of the hypergolic $F_2-C_2F_4-He$ mixture do not contain active relaxants of vibrationally excited HF(v) molecules. According to estimates, the atomic fluorine yield under comparable conditions is about 70 % in relation to that of the atomic fluorine generator operating on the F_2-D_2-He mixture which is the 'basic' one for a HF laser. The specific power is reduced by 23 % with an improvement in a set of the fuel performance and a significant reduction in the cost of triggering the laser in the course of its bench tests.**

Keywords: supersonic cw chemical HF laser, atomic fluorine generator, tetrafluoroethylene.

1. Introduction

The unique advantages of HF/DF supersonic cw chemical lasers (HF/DF CWCLs) – high energy potential and the absence of need for an external energy source (the capability of operation in a self-contained regime) – contribute to the active search for their practical application in a number of new technologies. The practical implementation of an effective supersonic HF/DF CWCL of self-contained type requires optimisation within the framework of two problems related to the selection of fuel for an atomic fluorine generator. The first problem involves the search, evaluation and selection of fuel components based on the requirements imposed on them as sources of chemical energy, on the one hand, and as substances being in operation outside the laser, on the other. The second problem is to determine the optimal chemical composition of the fuel to ensure maximum energy performance of a laser.

Optimisation within the framework of these problems is complicated by a controversial situation, according to which the components that are technologically more suitable may be better than those that are less suitable but have higher energy parameters. Thus, based on the operating conditions, a fuel component may be banned. It is for this reason that the search for new components is aimed at identifying and studying

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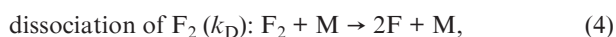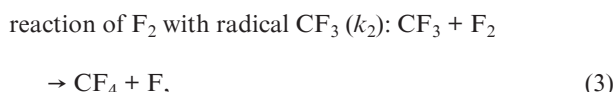compounds suitable for use as fluorine-containing oxidisers and primary fuels with the aim of replacing the 'basic' $F_2-D_2(H_2)-He$ fuel mixture, the characteristics of which are not quite satisfactory from the viewpoint of toxicity, fire and explosion hazard, storage capabilities and the cost of laser triggering.

In the field of HF/DF-CWCL development, research to improve the performance of fuels for the atomic fluorine generator are conducted in two directions: the selection of low-toxic fluorine-containing compounds to replace fluorine as an oxidiser, and the selection of relatively cheap high-energy hydrogen-free fuels to replace deuterium as a primary fuel.

As for the fluorine-containing oxidiser, as a result of many numerical [1] and experimental [2–4] studies of the HF/DF lasers, the NF_3 nitrogen trifluoride is recognised as the most effective compound in accordance with the 'energy performance–manufacturability' criterion.

The range of compounds suitable for use as primary fuels is quite wide and can be divided into two groups: hydrogen-containing compounds and their deuterium analogues, as well as compounds that do not contain hydrogen. In this case, while both groups of fuels are allowed in a DF laser, only hydrogen-free fuels can be used in a HF laser. The use of a much wider set of primary fuels in a DF laser due to the possibility of using hydrogen-containing compounds, for example, C_2H_4 ethylene, enables selection of a fuel that forms a minimum number of relaxants of vibrationally excited DF(v) molecules – 'cold' HF(0) molecules. It was established [5] that the transition to primary fuel (ethylene) instead of hydrogen provides an increase in specific energy yield by 30 %–40 %. Compensation for a reduction in the specific energy yield when replacing F_2 with NF_3 (also by 30 %–40 %) by its increase when replacing H_2 with C_2H_4 leads to the $NF_3-C_2H_4-He$ fuel composition, the energy characteristics of which are actually equivalent to the characteristics of the F_2-H_2-He fuel which is a 'basic' one for a DF laser. It is this circumstance that determines the use of the $NF_3-C_2H_4-He$ composition in all modern developments of high-power laser systems based on the DF CWCL [6], which gives reason to talk about the emergence of a new 'basic' fuel.

Thus, whereas in the case of a DF laser the problem of finding an effective fuel composition for an atomic fluorine generator can be considered, in a first approximation, as already solved, for a HF laser it remains open, mainly due to the lack of a primary fuel which would to the fullest extent satisfy the 'energy performance–manufacturability' criterion. In this regard, unsaturated perfluorocarbons, particularly, C_2F_4 tetrafluoroethylene, are very promising.

The aim of this research work was a preliminary assessment of the prospects of using tetrafluoroethylene as a pri-

mary fuel for a HF CWCL in order to justify the feasibility of further detailed studies on the characteristics of laser radiation (energy, amplification, spectral) and active medium (spatial and gas-dynamic). There are two reasons that make this task relevant. The first reason is extremely high cost of deuterium which is used as the primary fuel for bench tests of HF lasers. In the world market, this cost is about $ 5000 per kilogram, while in the domestic market – about $ 2500. The second reason is the lack of detailed studies on perfluorocarbons as fuel components for this type of lasers.

2. Tetrafluoroethylene as a chemical component

The C_2F_4 tetrafluoroethylene is attractive due to its commercial availability (commercial designation is Khladon-114), low cost (tetrafluoroethylene produced by OJSC 'Kirovo-Chepetsk Chemical Plant' costs about $ 5 per kilogram), and extensive accumulated operating experience. The physicochemical properties of tetrafluoroethylene are quite fully described in reference book [7]. To evaluate the operational performance, it is essential that C_2F_4 under normal conditions is a colourless and odourless gas (critical temperature $T_{crit} = 306.45$ K, critical pressure $p_{crit} = 4$ MPa), which is flammable when mixed with air. It belongs to substances of the fourth (lowest) hazard class (its maximum permissible concentration in the operating zone air is 20 mg m^{-3}). It has no corrosion activity, so its operation can use a wide range of metallic and non-metallic materials, except for fluoroplast-4 which promotes polymerisation. The tendency to polymerisation (often explosive) can be eliminated by introducing a small admixture of stabilisers (tertiary amines), which virtually do not affect the physicochemical properties of the monomer.

The detailed mechanism of fluorocarbon-perfluorocarbon reaction is unknown. There are only scarce data on the rate constants k_i of the individual reaction stages. For simplicity of reasoning, we may assume that, under stationary conditions, the process of C_2F_4 combustion in fluorine consists of the following elementary stages:

dissociation of C_2F_4 (k_d): $C_2F_4 + M \rightarrow 2CF_2 + M$,   (1)

reaction of F_2 with radical CF_2 (k_1): $CF_2 + F_2$

   $\rightarrow CF_3 + F$,   (2)

reaction of F_2 with radical CF_3 (k_2): $CF_3 + F_2$

   $\rightarrow CF_4 + F$,   (3)

dissociation of F_2 (k_D): $F_2 + M \rightarrow 2F + M$,   (4)
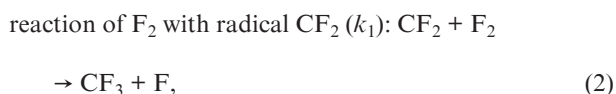
where M is an arbitrary particle of the fuel mixture in the combustion chamber of an atomic fluorine generator; and k_d, k_1, k_2, and k_D are reaction rate constants (1)–(4).

Evaluation of the characteristic times of elementary processes (1)–(4) for temperature $T = 1500$ K and pressure in the combustion chamber $p = 0.1$ MPa leads to the following results. The rate constant k_d in the temperature ranges $T = 1200–1800$ K and density $\rho = (0.9–1.7) \times 10^{-5}$ mol cm^{-3} is given by the relation [8]: $k_d = 10^{15.89}T^{0.5}\exp(-55.69/RT)$, where R is the gas constant. At the temperature $T = 1500$ K, $k_d = 2.6 \times 10^9$ cm^3 mol^{-1} s^{-1}, and the characteristic dissociation time of C_2F_4 is $t_d = 20$ μs.

The relations for the constants $k_1 = 10^{11.30}T^{0.5}\exp(-2.12/RT)$ and $k_2 = 10^{11.34}T^{0.5}\exp(-2.895/RT)$ are taken from reference book [9]. At $T = 1500$ K and F_2 volumetric content of 20 % in the fuel mixture, the characteristic reaction times (2) and (3) are $t_1 = 0.08$ μs and $t_2 = 1$ μs.

The dissociation of the F_2 excess by reaction (4) at the temperature $T = 1500$ K in accordance with the constant $k_D = 10^{12.66}T^{0.5}\exp(-2.849/RT)$ recommended for the temperature range $T = 1000–2000$ K in [10] occurs with the characteristic time $t_D = 170$ μs.

Therefore, as in the case of using the $F_2–D_2–He$ fuel which is primary for a HF laser, the slowest process under stationary conditions is the establishment of dissociation equilibrium in excess fluorine. The hypergolicity (self-ignitability) of the $F_2–C_2F_4$ mixture was experimentally tested. To this end, fluorine was fed into a quartz reactor in the form of a tube with an inner diameter of 8 mm along its axis, and tetrafluoroethylene was fed through the side wall. The pressure in the reactor was maintained at $p = 0.1$ MPa. The temperature in the reaction zone was measured by a thermocouple. When feeding the reagents into the reactor with a volume flow rate of 0.6–1 cm^3 s^{-1}, the mixture temperature increased to 523–573 K. An increase in the volume flow rate to 1.4–2 cm^3 s^{-1} led to the emergence of a yellow flame with a simultaneous temperature increase to 673–723 K. The temperature and concentration limits of self-ignition were not determined.

3. Thermodynamic calculation of combustion products

The calculation of equilibrium medium characteristics in an atomic fluorine generator operating on the $F_2–C_2F_4–He$ mixture was performed according to a simplified mathematical model similar to that proposed in [11] for a generator operating on the 'basic' $F_2–D_2–He$ mixture. Under typical operating conditions (pressure and temperature in the combustion chamber $p_c = 0.1–1.0$ MPa and $T_c = 1550–1880$ K), the combustion process is described by the equation:

$$\alpha F_2 + C_2F_4 + \gamma He = 2\alpha_{diss}(\alpha - 2)F$$

$$+ 2CF_4 + \gamma He + (1 - \alpha_{diss})(\alpha - 2)F_2, \quad (5)$$
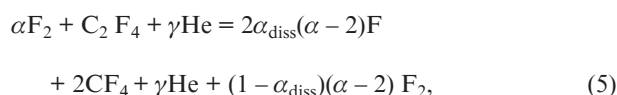
where α is the fluorine excess coefficient; $\alpha_{diss} = p_F/(p_F + 2p_{F_2})$ is the dissociation degree of molecular fluorine; $\gamma = 2(p_{CF_4} + p_{He})/(p_F + 2p_{F_2})$ is the helium excess coefficient with respect to the amount of C_2F_4; p_i is the partial pressure of the ith combustion product; and i are the reagents and combustion products of F, F_2, CF_4, and He.

The content of other fluorocarbons in the equilibrium mixture under these conditions is negligible. Estimates of the chemical composition (partial pressures p_i) and the temperature T_c of the gaseous medium in the atomic fluorine generator in the equilibrium approximation were obtained by the standard thermodynamic calculation for a gas-dynamically locked nozzle with allowance for heat losses (by analogy with [11]) at a pressure $p_c = 0.1$ MPa. At the same time, other medium parameters were calculated (specific heat, adiabatic exponent, molecular weight, etc). For the degrees of molecular fluorine dissociation $\alpha_{diss} = 0.95$ and helium dilution $\gamma = 10$, the calculation data are summarised in Table 1. The calculation results made it possible to compare the parameters of the alternative $F_2–C_2F_4–He$ fuel and the 'basic' $F_2–D_2–He$ fuel.

Table 1. Results of thermodynamic calculation of the chemical composition and parameters of the combustion products of compared fuels in atomic fluorine generator.

Fuel (T_c = 1480 K)	$m/F_{crit}/$ g cm^{-2} s^{-1}	Reagent consumption/mol cm^{-2} s^{-1}							k	μ/g mol^{-1}
		F$_2$	C$_2$F$_4$	D$_2$	He	F	CF$_4$	DF		
F$_2$–C$_2$F$_4$–He	7.45	0.092	0.025	–	0.37	0.080	0.050	–	1.40	14.87
F$_2$–D$_2$–He	6.90	0.116	–	0.060	0.45	1.116	–	0.120	1.56	10.10

Note: mass flow rates m/F_{crit} are calculated per 1 cm^2 of the critical cross section area of the nozzle block; F_{crit} is the critical section area (cm^2); k is the adiabatic exponent; m is the mass flow rate of a reagent (g s^{-1}); and μ is the molecular weight of combustion products.

Thermodynamic calculation has shown that, under comparable conditions, the hypergolic F$_2$–C$_2$F$_4$–He fuel provides the atomic fluorine yield at the level of 70% of the value characteristic of the F$_2$–D$_2$–He fuel which is primary for the HF-laser, with a necessary degree of dilution of the fuel mixture with helium. In contrast to the combustion products of the 'basic' fuel containing 'cold' DF(0) molecules – active relaxants of vibrationally excited HF(v) molecules, only a minimal amount of low-active relaxants is present among the products of C$_2$F$_4$ combustion with F$_2$, namely the CF$_4$ molecules [the constant of relaxation of HF(v) molecules on the CF$_4$ molecules is two orders of magnitude smaller than that on the 'cold' DF(0) molecules].

4. Energy performance assessment

In order to make a well-argued decision on the replacement of fuel components, it is necessary to assess the impact of these components on the energy performance of the HF laser, i.e. to compare the alternative fuel with the 'basic' one. For an objective assessment of the comparative energy efficiency of various fuels for the atomic fluorine generator, this comparison should be carried out with optimal chemical compositions providing the maximum specific energy yield $N_\Sigma = N/m_\Sigma$ for each fuel (where N is the laser radiation power, m_Σ is the total mass flow rate of the fuel mixture). Determination of such compositions is associated with a significant amount of complex and time-consuming numerical calculations or expensive experiments. In this regard, it is advisable to use a different approach, namely, an express assessment using the technique [12] developed on the basis of generalisation of the results of numerical studies performed for a large group of fuel compositions in a wide range of changes in the relaxation characteristics of combustion products.

In accordance with this technique, the comparative energy efficiency ε of the two fuels being compared only accounts for a change in the molecular weight μ of the combustion products that affects the rate of mutual diffusion of reagents in the laser chamber, and a difference in their relaxation properties φ_{rel} with respect to vibrationally excited HF(v) molecules. This efficiency is determined by the formula [12]

$$\varepsilon = \frac{N_{\Sigma max1}}{N_{\Sigma max2}} = \frac{\mu_2}{\mu_1}\frac{\varphi_{rel1}}{\varphi_{rel2}}, \tag{6}$$

where $N_{\Sigma max1}$ and $N_{\Sigma max2}$ are the specific energy yields of a laser operating on the 1 (alternative) and 2 ('basic') fuels at optimal chemical compositions; μ_1 and μ_2 are the molecular weights of the combustion products of fuels 1 and 2; and φ_{rel1} and φ_{rel2} are the corrections accounting for the relaxation properties of the combustion products of fuels 1 and 2.

Optimal chemical composition of each of the fuels, at which their energy characteristics should be compared, is determined on the basis of two conditions: optimal volume content x_F^{opt} = 0.118 of atomic fluorine in the combustion products, and optimal adiabatic temperature $T_c^{opt} \geqslant$ 1500 K of the combustion products (found previously for the 'basic' fuel, i.e., for the fuel with which the comparison is performed [in formula (6) it corresponds to the index 2]).

The conditional fuel formula can be written as

$$A + \alpha B + \psi C, \tag{7}$$

where A is the fuel; B is the oxidiser; C is the inert diluent; and α and ψ are the amounts of moles of oxidiser and inert diluent per mole of fuel.

For each of the fuels in question, at a selected pressure in the combustion chamber of the atomic fluorine generator, a series of thermodynamic calculations is performed, in which the coefficients α and ψ are varied. Thus obtained data on chemical composition and temperature of the combustion products are used to determine the α_{opt} and ψ_{opt} values that satisfy the two conditions noted above. The values of corrections ψ_{rel} accounting for the relaxation properties of combustion products are determined by the specific energy yield losses due to VT relaxation of vibrationally excited HF(v) molecules on the combustion by-products as functions of the relaxation factor k_{rel} [12]:

$$k_{rel} = \sum k_{VT}^i \psi_{F_2}^i, \tag{8}$$

where k_{VT}^i is the rate constant of VT relaxation of radiating molecules on the ith combustion product; $\psi_{F_2}^i = x_i/0.5x_F$ is the dilution degree of the ith combustion product per one mole of free, conditionally molecular fluorine; and x_i, x_F are the molar fractions of ith combustion product and free fluorine.

The operational estimate S, which reflects the complex of properties of both initial components and fuel as a whole, which are essential from the laser operation viewpoint, is determined by a wide range of indicators using data on the physical, chemical and toxicological properties of individual reagents and fuel as a whole with allowance for its availability (production scale and cost). A method for S determination is described in [12].

In the course of the express assessment, the F$_2$–D$_2$–He fuel was adopted as the 'basic' fuel. The conditional formulas for the fuels under comparison are as follows:

$$C_2F_4 + \alpha F_2 + \psi He, \tag{9}$$

$$D_2 + \alpha F_2 + \psi He. \tag{10}$$

The conditions accepted for conducting thermodynamic calculations are as follows: p_c = 0.1 MPa; T_c^{opt} = 1900 K (when using common-in-practice nozzle blocks with a geo-

Table 2. Results of thermodynamic calculation of combustion product parameters and relaxation corrections for compared fuels in atomic fluorine generator.

Fuel	x_F^{opt}	T_c^{opt}/K	α_{opt}	ψ_{opt}	μ/g mol^{-1}	ψ_{rel}
$F_2-C_2F_4-He$	0.118	1901	3.34	18.0	13.18	1.00
F_2-D_2-He	0.118	1900	2.50	11.9	9.00	0.89

metric degree $10-20$ of nozzle expansion, the value of T_c^{opt} without preliminary studies can be taken equal to $1800-2000$ K [11, 13]); $x_F^{opt} = 0.118$; $\alpha = 2.0-3.5$ and $\psi = 10-20$ [for fuel (9)]; and $\alpha = 1.5-2.5$ and $\psi = 3-15$ [for fuel (10)]. The calculation results are listed in Table 2.

Given relation (6) and the data of Table 2, the assessment of the comparative energy efficiency of alternative $F_2-C_2F_4-$He fuel is determined as follows:

$$\varepsilon = \frac{N_\Sigma}{N_{\Sigma\,base}} = \frac{9.00/13.18}{1.00/0.89} = 0.77. \qquad (11)$$

This means that replacing deuterium with tetrafluoroethylene in the 'basic' F_2-D_2-He fuel composition reduces the specific energy yield N_Σ of the HF laser by 23%. At the same time, the operational properties of the alternative fuel in relation to that of the 'basic' one turn out somewhat improved: $S/S_{base} = 0.92$ (the ratio lesser than unity corresponds to a more suitable set of operational properties).

To confirm the reliability of the results obtained using the express assessment, one can refer to the experiments and numerical calculations performed during the HF/DF CWCL studies conducted by different authors. Comparative estimates of the energy efficiency of some fuel compositions obtained from the results of well-known studies on the self-contained CWCL with various types of nozzle blocks are presented in Table 3. The Table shows that the ratio ε of the specific laser energy yield obtained both by numerical simulation and experimentally is, in some cases, very close to the energy estimates for these fuels obtained by the express assessment technique.

The maximum difference of the energy estimates from the results of numerical calculations is 8%, while that from the experimental data is 17%, which should be considered very satisfactory.

5. Conclusions

To substantiate the feasibility of detailed studies on the characteristics of laser radiation and the HF CWCL active medium, a search was conducted on the preliminary assessment of the prospects for the use of tetrafluoroethylene as a primary fuel. The results of comparative evaluation of the energy efficiency of alternative ($F_2-C_2F_4-$He) and primary (F_2-D_2-He) fuels obtained by express assessment, the reliability of which is confirmed by comparison with experiments and numerical calculations performed by domestic and foreign authors, lead to the following conclusion.

Since the reduction in the specific energy performance of the HF laser when replacing deuterium with tetrafluoroethylene is not too large (23%), and the cost of C_2F_4 is almost 500 times lower than that of D_2, it should be recognised reasonable the use of cheap tetrafluoroethylene instead of expensive deuterium in situations when the aim of reaching the limiting specific laser energy performance is not pursued (for example, when conducting a large amount of bench tests to verify the laser design, and also in studies on the interaction of laser radiation with various objects). Proceeding from this, for a detailed comparative assessment of changes in the characteristics of laser radiation and active medium of an HF CWCL operating on the $F_2-C_2F_4-$He fuel, it is advisable to perform numerical calculations or experiments.

As for the practical issues of employing tetrafluoroethylene, its industrial production is being conducted in Russia at the OJSC 'Kirovo-Chepetsk Chemical Plant' (Kirov region) and the OJSC 'Galogen' (Perm), while the Russian Scientific

Table 3. Assessment results for comparative energy efficiency of various fuel compositions.

Fuel	Energy efficiency ε	Degree of difference from the express assessment (%)	Method of energy efficiency determination	Reference
NF_3-D_2-He	0.73	1	Numerical calculation	[14]
	0.71	4	Experiment	[2]
	0.74	–	Express assessment	[12]
$N_2F_4-D_2-He$	0.79	1	Numerical calculation	[2]
	0.94	17	Experiment	[2]
	0.80	–	Express assessment	[12]
$F_2-C_6F_6-He$	0.98	8	Numerical calculation	[15]
	0.79	13	Experiment	[15]
	0.91	–	Express assessment	[12]
$NF_3-C_6F_6-He$	0.59	2	Numerical calculation	[16]
	0.58	–	Express assessment	[12]
$F_2-C_2H_4-He$	1.06	7	Experiment	[3]
	0.99	–	Express assessment	[5]
NF_3-H_2-He	0.78	5	Numerical calculation	[14]
	0.80	8	Experiment	[17]
	0.74	–	Express assessment	[12]
$NF_3-C_2H_4-He$	0.75	13	Experiment	[4]
	0.66	–	Express assessment	[12]

Centre 'Applied Chemistry' has accumulated extensive expe-
rience in operating this product.

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