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# Efficiency of using nitrogen trifluoride as an oxidiser in a supersonic continuous-wave chemical HF laser

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***Abstract.*** **We report the results of an experimental study of possible ways to increase the efficiency of using $NF_3$ nitrogen trifluoride as an oxidiser in an atomic fluorine generator of a supersonic continuous-wave chemical HF laser with a flat nozzle block corresponding to the nozzle–nozzle reagent mixing scheme. As such ways, we consider increasing the residence time of reagents in the combustion chamber and rising temperature in the chemical reaction zone. Comparison of specific energy characteristics of an HF laser operating with the use of the studied ($NF_3$) and 'basic' ($F_2$) oxidisers shows that the replacement of molecular fluorine with nitrogen trifluoride during the burning with deuterium in the combustion chamber of an atomic fluorine generator with allowance for the proposed measures leads to a reduction in the specific laser energy output by no more than 23 %–24 % at high burning completeness.**

***Keywords:*** *supersonic continuous-wave chemical HF laser, oxidiser, fluorine, nitrogen trifluoride, burning completeness, specific energy characteristics.*

## 1. Introduction

The use of $NF_3$ nitrogen trifluoride as a fluorine-containing oxidiser for an atomic fluorine generator in a supersonic HF/DF continuous-wave chemical laser (HF/DF CWCL) attracted the attention of researchers in the early 1970s as an alternative to molecular fluorine $F_2$. The latter is characterised by extremely low processability (not a completely satisfactory set of operational properties), very high corrosion activity and toxicity (an MPC of 0.03 mg $m^{-3}$), which makes it refer to the first (highest) hazard class substances [1]. In this regard, $NF_3$ has obvious advantages: it belongs to the fourth (lowest) hazard class (an MPC of 10 mg $m^{-3}$) [1], is significantly less corrosive, which ensures its compatibility with most construction materials, and also greatly simplifies storage and transportation. When working with fluorine-containing fuel components, the requirements for operating conditions increase significantly for high-power large-scale systems due to the significant mass consumption of these components and, consequently, the need to have a large stock of them in the storage system. For this reason, nitrogen trifluoride is used in all currently known and promising projects of high-power (100 kW and higher) systems based on HF/DF CWCLs (Table 1).

From the viewpoint of practical application, the most realistic, in our opinion, are the projects of using CWCLs on spaceborne platforms to protect the orbital station from micrometeorite and anthropogenic particles [2], as well as to clean up near-earth space from debris [3]. Experimental studies conducted under bench conditions have shown that CWCL models can operate in space vacuum for a long (about 100 s) time in a cw regime at a power level of several tens of kilowatts without compromising performance and any significant external energy consumption. For the above purposes, the use of HF CWCLs seems more appropriate, since, due to a higher gain, its energy potential exceeds the DF CWCL potential by approximately 25 %, and there are no restrictions on the radiation wavelength (the problem of passing laser radiation through the real atmosphere).

For spaceborne platforms, the energy efficiency of fuel – its specific energy output $N_\Sigma$ – is the most critical parameter, because it determines the mass and size characteristics of the laser system. The decrease in the chemical activity of nitrogen trifluoride could not but affect its energy, which raises the question: What is the true price for improving the processability? Information about the consequences of replacing $F_2$ with $NF_3$ for the HF/DF CWCL energy performance is highly controversial, and the vast majority of studies are related to DF CWCLs (see Table 1). Calculated estimates [4, 5] have shown that the replacement of molecular fluorine with nitrogen trifluoride in the same laser leads to a decrease in the HF CWCL specific energy output by 26 %. In the experiment, even with optimisation of the fuel chemical composition, the corresponding decrease reaches 40 % [6]. This fact testifies that the energy potential of nitrogen trifluoride is far from fully realised, and indicates the need to improve the design of the combustion chamber of the atomic fluorine generator in order to ensure a higher degree of realisation of the fluorine contained in nitrogen trifluoride.

There is no detailed information about the design of well-known foreign systems based on high-power HF CWCLs, and so it is not known what technical decisions were taken when designing an atomic fluorine generator operating with the use of $NF_3$. In this regard, when designing domestic promising systems, the issue of finding ways to increase the efficiency of using nitrogen trifluoride as an oxidiser in a HF CWCL atomic fluorine generator remains relevant. This paper is dedicated to the discussion of this problem.

## 2. Structural features of the nitrogen trifluoride molecule and its effect on the HF laser energy performance

A significant reduction in the chemical aggressiveness and toxicity of nitrogen trifluoride compared to molecular fluorine, which allows it to be preferred in operational issues, is determined by the structural features of the $NF_3$ molecule.

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**Table 1.** Existing and promising system designs based on high-power supersonic cw chemical HF/DF lasers.

| System name | Working molecule accommodation | Fuel composition | Estimated power/MW | Firm (country) | References |
|---|---|---|---|---|---|
| BDL (Baseline Demonstration Laser) | HF/DF, ground-based | $NF_3-D_2(H_2)-He$ | 0.1 | TRW (USA) | [7] |
| NACL (Navy Advanced Chemical Laser) | HF/DF, ground-based | $NF_3-D_2(H_2)-He$ | 0.1 | TRW (USA) | [7] |
| MADS (Modular Army Demonstration System) | DF, ground-based | $NF_3-H_2-He$ | 0.1 | TRW (USA) | [7] |
| MIRACL (Mid-Infrared Advanced Chemical Laser) | DF, ground-based | $NF_3-H_2-He$ | 2.2 | TRW (USA) | [8] |
| Sigma | DF, ground-based | $NF_3-H_2-He$ | 2.2 | Rockwell International (USA) | [8] |
| Alpha | HF, spaceborne | $NF_3-D_2-He$ | 2.2 | TRW (USA) | [9] |
| THEL (Tactical High Energy Laser) | DF, ground-based | $NF_3-C_2H_4-He$ | 0.4 | TRW (USA), Taridan, IAI, Rafael (Israel) | [10] |
| MTHEL (Mobile Tactical High Energy Laser) | DF, ground-based | $NF_3-C_2H_4-He$ | 0.2 | TRW (USA), Taridan, IAI, Rafael (Israel) | [10] |
| Skyguard | DF, ground-based | $NF_3-C_2H_4-He$ | 0.3 | Northrop Grumman (USA) | [11] |
| Aviation system | HF/DF, air-based | $NF_3-(D_2)H_2-He$ | 0.1 | NPO Energomash (Russia) | [12] |
| Mobile system | DF, ground-based | $NF_3-C_2H_4-He$ | 0.3 | NPO Energomash (Russia) | [13] |
| Marine system | DF, above-water | $NF_3-C_2H_4-He$ | 0.3 | NPO Energomash (Russia) | [14] |

According to [15], the breaking energy of the first N–F bond is 230 kJ mol$^{-1}$, and that of the subsequent bonds is 335 and 230 kJ mol$^{-1}$, respectively. The dissociation energy of the $F_2$ molecule (breaking the F–F bond) is 159 kJ mol$^{-1}$.

A higher energy consumption for the dissociation of $NF_3$ molecule requires the use of larger amounts of oxidiser to obtain the same amounts of atomic fluorine F. For example, to obtain 1 g of fluorine and heat it to a temperature of 2000 K at a dilution degree of the mixture with helium $\psi$ = 10, it is necessary to burn 1.4 g of fluorine if the $F_2$ oxidiser is used, and 2 g of fluorine (contained in the $NF_3$ molecule) in the case of $NF_3$. This circumstance inevitably leads to a decrease in the specific energy output $N_\Sigma = N/m_\Sigma$ ($N$ is the absolute lasing power, and $m_\Sigma$ is the total mass fuel consumption), which characterises the efficiency of using fuel as a whole. The presence of nitrogen molecules in the oxidising gas composition (the F + DF + He + $F_2$ + $N_2$ mixture from an atomic fluorine generator) is accompanied by a decrease in the rate of its outflow from the nozzle block, a decrease in the lasing zone length and the appearance of additional relaxation of vibrationally excited HF($v$) molecules on $N_2$ molecules. This leads to a decrease in the specific power $N_F = N/m_F$ ($m_F$ is the mass consumption of atomic fluorine, and $v$ is the vibrational quantum number), which characterises the efficiency of using fluorine atoms.

Fuel optimisation by chemical composition (coefficients $\alpha$ and $\psi$) indicates the possibility of implementing the $NF_3-D_2-He$ fuel composition within the existing small-scale HF CWCL models [16] with acceptable efficiency. However, neither the calculations nor the experiments took into consideration the burning incompleteness of fuel in the actual combustion chamber of an atomic fluorine generator. The fact is that for the correct calculation of heat and mass transfer in the combustion chamber, it is necessary to solve a three-dimensional system of Reynolds equations for the compressible gas, supplemented by diffusion equations for individual components of the mixture. This approach is quite complicated and requires very significant computing resources. For this reason, it is usually assumed that the reagents are mixed instantly, directly in the nozzle head plane. As for experiments, the study of the consequences of replacing reagents is usually conducted on a facility of the same design without allowance for their specifics. As a result, in both cases, the burning incompleteness of fuel turns out excluded from consideration.

It is known [15] that the high strength of the N–F bond makes it necessary to heat the reagents involved in reactions with $NF_3$ to a temperature of at least 600 K. As applied to HF CWCLs, this circumstance requires special ignition means to initiate the $NF_3$ burning reaction with $D_2$. Thermal decomposition of nitrogen trifluoride occurs at temperatures above 1200 K. In this case, the first bond, $NF_3 \rightarrow NF_2 + F +$ 230 kJ mol$^{-1}$, turns out first broken and an extremely stable $NF_2$ radical is formed, the stability of which is explained by the high (335 kJ mol$^{-1}$) dissociation energy of the second N–F bond of the $NF_3$ molecule. $NF_2$ fluoroazenes and $N_2F_2$ difluorodiazines formed during thermal dissociation of the $NF_2$ radical are unstable compounds that decompose into nitrogen and fluorine at temperatures above 400 K.

If the processes of ignition, mixing, heat and mass transfer, and also chemical kinetics are poorly arranged, the formation of an oxidising gas is possible in the combustion chamber of an atomic fluorine generator, which differs from that required both in temperature and chemical composition, and in which, along with DF, He, F and $N_2$ molecules, $F_2$, $NF_2$, $NF_3$ and $D_2$ molecules may appear. This leads to a substantial violation of the conditions for the pump reaction $F + H_2 \rightarrow HF(v) + H$ and, consequently, to a sharp drop in the power of the generated laser radiation.

## 3. Incompleteness of fuel burning in the combustion chamber of an atomic fluorine generator and methods for its reduction

The presence of incomplete fuel burning was revealed in the very first experiments aimed at using nitrogen trifluoride in HF CWCL models designed to operate on molecular fluorine. This is due to the different intensities of the processes of mixture formation and burning of different types of fuels in the combustion chamber of an atomic fluorine generator of the same design. In this situation, in order to obtain objective information about the effect of the chemical nature of the fuel on the laser efficiency, it is necessary to optimise the fuel burning process.

Due to the high rates of the chain reaction $F_2 + D_2$, which occurs already at the first stage with the release of a significant amount of heat, and to the moderate F–F binding energy (159 kJ $mol^{-1}$), it is obvious that the decisive factor to attain high completeness of fuel burning is the duration of the process of mixing the reagents (the time they are in the combustion chamber).

Fuel burning incompleteness during the HF CWCL operation using nitrogen trifluoride is associated not only with the features of chemical kinetics of the $D_2$ interaction with $NF_3$, but also with the dissociation features of the latter. Unfortunately, the absence of a kinetic model and kinetic constants precludes the possibility of a computational study of this issue. Given the above, for effective use of nitrogen trifluoride, special measures must be undertaken, aimed both at intensifying the process of mixing the fuel components and increasing the rates of chemical reactions.

Such special measures include the following: increasing the residence time of the reacting fuel components in the combustion chamber by changing its length and increasing the temperature in the chemical reaction zone of the $D_2 + NF_3$ component in order to accelerate the dissociation of the latter. Technically, this measure can be implemented in a dual-zone combustion chamber, in the first zone of which the increased temperature of the mixture is attained by reducing the amount of inert diluent (helium), while in the second zone the mixture is diluted to the required degree.

Note that dual-zone combustion chambers are widely used in rocket and space technology to produce a working fluid (generator gas) used in turbopump units of fuel supply systems for liquid rocket engines.

The purpose of this work is to experimentally study the proposed measures to improve the efficiency of using nitrogen trifluoride as an oxidiser for the HF CWCL atomic fluorine generator.

## 4. Experimental conditions

The experiments were performed on a bench using a HF CWCL model (a calculated power of 5 kW) equipped with a flat nozzle block corresponding to the nozzle–nozzle reagent mixing scheme [16]. The experiments were carried out in two series. In the first series of experiments, the effect of the combustion chamber length $L_c$ on the laser energy characteristics was investigated by means of changing the chamber length from 160 to 300 mm by sequentially installing one and two additional sections with a length of 70 mm each (the original length of the combustion chamber was 160 mm). Sections were cooled by water. For a fixed length $L_c$ and identical values of the operating parameters ($\alpha = n_{F_2}/n_{D_2}$, $\psi = n_{He}/(n_{F_2} - n_{D_2})$, $\alpha_2 = n_{H_2}/(n_{F_2} - n_{D_2})$ and $m_F$, where $n_{H_2}$, $n_{F_2}$, $n_{D_2}$, and $n_{He}$ are the amount of moles of hydrogen, fluorine, deuterium and helium, and $m_F$ is the mass flow rate of free molecular fluorine), we measured the laser energy characteristics using molecular fluorine and nitrogen trifluoride as an oxidiser. Each test included four regimes with a duration of 6 s. The first and third regimes were the control ones with $F_2$ supply, while the second and fourth regimes were the operating ones with $NF_3$ supply.

In the second series of experiments, we studied the effect of a temperature rise on the efficiency of using $NF_3$ in a dual-zone combustion chamber with $L_c$ = 230 mm, equipped with one additional section modified to supply the diluent to the second zone. In this case, the effect of the diluent supply method and the design features of the second zone mixer on the energy performance of the 'basic' fuel with the conditional formula $D_2 + \alpha F_2 + \psi(\alpha - 1)He + \alpha_2(\alpha - 1)H_2$ was initially determined. For this purpose, dual-regime operation of the combustion chamber was provided, with a duration of 6 s for each regime. In the first regime, all components were only fed through the nozzle head, while in the second regime part of the diluent (initially supplied through the nozzle head) was directed to the second zone mixer located in the immediate vicinity of the nozzle block inlet, with maintaining the total diluent (helium) consumption. According to estimates, this diluent supply scheme made it possible to raise the temperature of the fuel mixture in the first zone to 2900 K while maintaining the temperature at the nozzle block inlet at the level of 2000 K.

Then, the efficiency of using nitrogen trifluoride in a dual-zone diluent supply method was evaluated. To do this, in each single-regime experiment lasting 22 seconds, the HF CWCL energy characteristics were measured using two fuel compositions with different oxidisers ('basic' and alternative) with the same mixture parameters at the nozzle block inlet. Experiments were performed with the following chemical compositions of fuels: $\alpha = 1.5$, $\psi = 8.5$ (for the 'basic' $F_2-D_2-He$ fuel) and $\alpha = 1.5$, $\psi = 8.0$ (for the alternative $NF_3-D_2-He$ fuel). The selection of these parameters was guided by the following considerations. The oxidiser excess coefficient $\alpha = 1.5$ was taken from the condition of ensuring an acceptable ($T_c \sim$ 2000 K) temperature level in the atomic fluorine generator with allowance for the heat losses caused by cooling the combustion chamber at the typical degree of dilution of the fuel mixture with helium $\psi = 10$. It is known [17] that the effect of heat losses on the chemical composition of combustion products due to a decrease in the concentration of atomic fluorine stipulated by its recombination on the combustion chamber walls manifests itself at $T_c < 2000$ K. An increase in the dilution coefficient $\psi$ in the 'basic' composition compensates for the additional amount of diluent (nitrogen) released during the laser operation on nitrogen trifluoride by $\alpha/3$, in accordance with the conditional formula

$$D_2 + 2(\alpha/3)NF_3 + \psi(\alpha - 1)He \rightarrow 2DF + 2(\alpha - 1)F$$

$$+ \psi(\alpha - 1)He + (\alpha/3)N_2,$$

and allows experiments with the same dilution degree of both types of fuel to be performed. For the secondary fuel excess coefficient (hydrogen), a typical value of $\alpha_2 = 20$ was accepted. The lasing power was measured by the closed resonator method using a mirror calorimeter (mirror diameter 60 mm) with zero transparency according to the technique [17] with an error of ± 7%. To ignite $NF_3$, the simplest and most reliable chemical ignition method was used. It consisted in feeding molecular $F_2$ into the combustion chamber of an atomic fluorine generator for a start-up time (up to 0.5 s), followed by its gradual replacement with $NF_3$.

## 5. Experimental results

Obviously, with a fixed design of the nozzle head of the atomic fluorine generator, there is an optimal length of its combustion chamber for each fuel composition. If the combustion chamber length is less than the optimal one, the fuel capabilities are not fully utilised due to incomplete burning. If the combustion chamber is too long, the laser efficiency is reduced due to unreasonably large heat losses and losses of fluorine atoms due to their recombination on the cooled walls. Therefore, determining the optimal length of the combustion chamber for each of the studied fuels is a necessary step in providing objective conditions for comparing their energy performance.

Experimental data illustrating the effect of the combustion chamber length on the HF CWCL energy characteristics are shown in Figs 1 and 2. One can see that they are distinguished by a smooth character and the presence of maxima. The increase in the specific laser energy performance with increasing the combustion chamber length is explained by an increase in the fuel burning completeness. A further increase in the combustion chamber length leads, due to an increase in the cooled surface, to an increase in heat losses and losses of atomic fluorine and, thus, to a decrease in both the specific power $N_F$ (Fig. 1) and the specific energy output $N_\Sigma$ (Fig. 2).

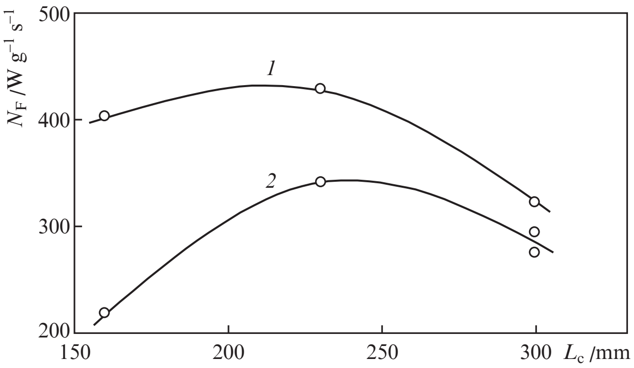


**Figure 1.** Dependence of the HF CWCL specific power on the combustion chamber length in an atomic fluorine generator: (*1*) $F_2$–$D_2$–He fuel, and (*2*) $NF_3$–$D_2$–He fuel.

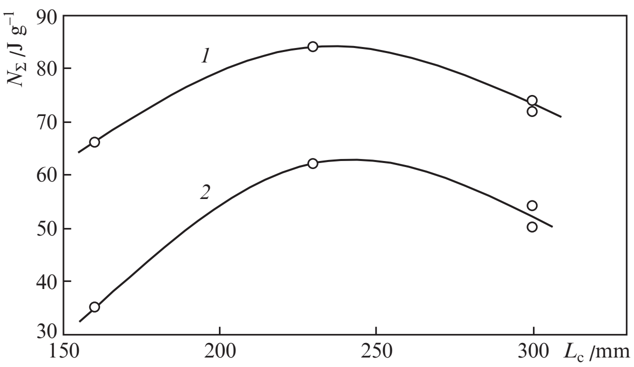


**Figure 2.** Dependences of the HF CWCL specific energy output on the combustion chamber length in an atomic fluorine generator: (*1*) $F_2$–$D_2$–He fuel, and (*2*) $NF_3$–$D_2$–He fuel.

The obtained results indicate the presence of the optimal length $L_c^{opt}$ of the combustion chamber for each of the fuel compositions in question. In particular, when using fluorine, the value of $L_c^{opt}$ turned out equal to 210–230 mm, while using nitrogen trifluoride, to 250 mm. Maximum values of the specific energy characteristics of the HF CWCL model under study, corresponding to the optimal lengths of the combustion chamber, are given in Table 2. They can be used to determine the coefficients that characterise the efficiency of using fluorine in nitrogen trifluoride,

$$k_F = (N_F^{max})_{NF_3}/(N_F^{max})_{F_2},$$

and overall fuel utilisation efficiency:

$$k_\Sigma = (N_\Sigma^{max})_{NF_3}/(N_\Sigma^{max})_{F_2}.$$

The coefficient $k_F^{exp}$ obtained in the experiment turned out to be 0.80, while its calculated value $k_F^{theor}$ determined using the calculation model [18] without allowance for burning incompleteness was 0.79. A fairly good agreement between the experimental and calculated values of the $k_F$ coefficient indicates that the high burning completeness of $NF_3$ with $D_2$ in the HF CWCL model under study was attained with the correct choice of the combustion chamber length. The experimental coefficient of fuel utilisation efficiency $k_\Sigma^{exp}$ was 0.76, i.e. when replacing fluorine with nitrogen trifluoride as an oxidiser, the specific energy output decreased by 24%. It should be noted that the values found for the combustion chamber length are only optimal for the atomic fluorine generator of this design with this particular reagent injection system.

Experiments were performed to determine the combustion chamber efficiency using a dual-zone scheme, with parameters $\alpha_{opt}$ and $\psi_{opt}$ optimal for each type of fuel [19]: $\alpha_{opt}$ = 1.68, $\psi_{opt}$ = 12.6 (for 'basic' $F_2$–$D_2$–He fuel) and $\alpha_{opt}$ = 1.48, $\psi_{opt}$ = 12 (for alternative $NF_3$–$D_2$–He fuel). The following values of the coefficients were obtained: $k_F^{exp}$ = 0.98 and $k_\Sigma^{exp}$ = 0.77 (Table 2). The value of the $k_F^{theor}$ coefficient calculated with the optimised operating parameters for both types of fuel was 0.95. The large value of the $k_F^{exp}$ coefficient and its proximity to $k_F^{theor}$ makes it possible to speak of high completeness of fuel burning in a dual-zone atomic fluorine generator; moreover, it exceeds the burning completeness in a length-optimised combustion chamber, with the specific energy output decreased by 23%.

Thus, the measures taken to increase the completeness of the burning of nitrogen trifluoride with deuterium in the com-

**Table 2.** Experimental maximum values of the specific energy characteristics of the HF CWCL model under study and fluorine utilisation factors in nitrogen trifluoride and fuel.

| Combustion chamber features | Fuel | $L_c^{opt}$/mm | $N_F^{max}$/W g$^{-1}$ s$^{-1}$ | $N_\Sigma^{max}$/J g$^{-1}$ | $k_F^{exp}$ | $k_F^{theor}$ | $k_\Sigma^{exp}$ |
|---|---|---|---|---|---|---|---|
| Length-optimised chamber | $F_2$–$D_2$–He | 210–230 | 435 | 83 | – | – | – |
| | $NF_3$–$D_2$–He | 250 | 346 | 63 | 0.80 | 0.79 | 0.76 |
| Dual-zone chamber ($L_c$ = 230 mm) | $F_2$–$D_2$–He | – | 228 | 57 | – | – | – |
| | $NF_3$–$D_2$–He | – | 232 | 44 | 0.98 | 0.95 | 0.77 |

bustion chamber of an atomic fluorine generator within the framework of the HF CWCL model under study proved to be quite effective.

## 6. Conclusions

The results of this study, in the framework of which special measures were undertaken to increase the burning completeness of nitrogen trifluoride with deuterium, have shown that increasing the residence time of components in the combustion chamber by increasing its length, and raising the temperature in the zone of chemical reaction of $NF_3$ with $D_2$ in order to accelerate the nitrogen trifluoride dissociation in the dual-zone combustion chamber made it possible, with replacing $F_2$ by $NF_3$, to limit the reduction in the specific energy output of the studied HF-CWCL model to 23%–24%. Since such a decrease in the specific energy performance virtually (considering measurement errors) coincides with the calculated one (26%), this fact indicates the attainment of high completeness of the $NF_3-D_2-He$ fuel burning in the combustion chamber of the atomic fluorine generator of a small-scale laser installation under study and, accordingly, high efficiency of nitrogen trifluoride consumption. The difference in the energy characteristics of the NF CWCL operating with $F_2$ and $NF_3$ can be reduced by increasing the laser installation scale (increasing the active medium length in the direction of the resonator's optical axis and, accordingly, reducing the threshold gain).

Nonetheless, when replacing fuel components, it is still necessary to optimise both the laser operating parameters and the gas-dynamic path design (nozzle head, combustion chamber of atomic fluorine generator, and nozzle block). This is also indicated by the comparison of the fields of the active medium flow in the cavity of the HF CWCL resonator operating with the use of the $F_2-D_2-He$ and $NF_3-D_2-He$ fuel compositions. The results of this comparison demonstrate differences both in the flow gas-dynamics (in the geometry of characteristic flow regions and the profiles of total and static pressure) and in the processes occurring in the resonator cavity (in the intensity of active medium glow in the regions of visible and lasing spectra). They may comprise the subject of a separate study.

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