Energy characteristics of a supersonic continuous-wave chemical HF laser using hydrazine decomposition products

I.A. Fedorov

Abstract. **We report the results of calculations and experimental studies of specific energy characteristics of a supersonic cw chemical HF laser with a flat nozzle block corresponding to the nozzle–nozzle reagent mixing scheme, in which catalytic decomposition products of hydrazine N_2H_4 ($H_2 + N_2 + NH_3$) are used as a secondary fuel. As a result of experiments with cold (300 K) model mixtures, the specific energy extraction of the laser is found to decrease in this case by 43 % compared with the laser utilising pure hydrogen. The calculations show that the main reason for a decrease in the specific energy of the laser is an increase in the temperature in the mixing zone of the oxidising gas and secondary fuel jets. A strong effect of ammonia is revealed. Taking into account the actual heating of the products (up to 1000 K) during the catalytic decomposition of N_2H_4 allows the level of specific energy extraction from the laser to approach that of energy extraction from the laser operated on pure hydrogen by 13 %, which will lead to a decrease in the specific energy extraction by only 30 % in the absence of NH_3 decomposition products.**

Keywords: supersonic continuous-wave chemical HF laser, secondary fuel, hydrazine decomposition products, ammonia, specific energy characteristics.

1. Introduction

A supersonic HF continuous-wave chemical laser (HF CWCL) is a four-component fuel system including a primary fuel (deuterium D_2), a fluorine-containing oxidising agent (usually fluorine F_2), an inert diluent (helium He) and a secondary fuel (hydrogen H_2). In such a system, an auxiliary chemical reaction of a fluorine-containing oxidising agent with primary fuel in the presence of an inert diluent is used to generate fluorine atoms, which play the role of active centres necessary to initiate a pump reaction: $\alpha F_2 + D_2 + \psi He \rightarrow 2DF + 2(\alpha - 1)F + \psi(\alpha - 1)He$ [α is the coefficient of excess oxidiser (fluorine) in the combustion chamber of the atomic fluorine generator, ψ is the degree of dilution of the fuel mixture with helium in the combustion chamber of the atomic fluorine generator); an exothermic reaction of fluorine atoms with secondary fuel is used for pumping: $F + H_2 \rightarrow HF(v) + H$. As a result, an active medium – vibrationally excited hydrogen fluoride molecules $HF(v)$ with an inverse distribution of populations over vibrational levels up to the fourth level ($v = 1 – 4$) – is formed in the laser chamber (resonator cavity).

The unique advantages of lasers of this type – their high energy potential, absence of the need for an external energy source and possibility of generating mid-IR radiation ($\lambda = 2.6 – 3.2$ μm) – stimulate an active search for the prospects of producing various versions of mobile laser systems on their basis. Thus, Karel'skii et al. [1] showed the possibility of using HF CWCLs for laser location and protection of spacecrafts from micrometeorite particles and space debris, and Bashkin et al. [2] considered the areas of the possible application of multi-frequency CWCLs for sensing the Earth's atmosphere for environmental control. In these cases, it is advisable to place the CWCLs on high-altitude carriers, i.e. spacecrafts or airplanes, as suggested, for example, in [3]. This raises the problem of finding the optimal technical implementation of the main subsystems of the laser system to ensure an acceptable balance between its energy and weight and size characteristics.

A significant part (50 %–70 %) of a mobile laser unit (by weight and size) is the fuel storage and supply subsystem, which includes compression devices, high-pressure cylinders and in some cases cryogenic equipment. The use of hydrogen as a secondary combustible in a gaseous or cryogenic state due to its low density in the gas phase and low temperature and density in the liquid phase creates problems associated with the weight and size characteristics of the storage subsystem and the storage resource of explosive and fire hazardous substances. To solve these problems in mobile laser systems, it is preferable to store hydrogen in the form of hydrogen-containing compounds, and to obtain it as necessary due to chemical reactions. From this came the idea of developing compact solid-fuel [4] and liquid [5] hydrogen gas generators that do not require the use of the above devices and can significantly improve the weight and size characteristics of the fuel component storage and supply subsystem for a mobile laser unit based on a HF CWCL.

The level of technical development of solid-fuel hydrogen generators is currently low and is at the stage of selecting generation methods, their analysis and assessment of the range of starting hydrogen-containing substances and formulations of mixtures on their basis. Direct laser experiments using a solid-fuel hydrogen generator were performed only on low-power (less than 10 W) laboratory facilities [6].

As for the liquid hydrogen generator, it can be designed on the basis of the catalytic decomposition of hydrazine N_2H_4 [5]. The choice of the latter is determined by a high (12.5 wt. %) content of hydrogen in its composition, high density (about 10 times higher than the H_2 density) and rich experience in use in spacecraft propulsion technology (N_2H_4 is

I.A. Fedorov Russian Scientific Center 'Applied Chemistry', ul. Krylenko 26A, 193232 St. Petersburg, Russia;
e-mail: appolo.47@mail.ru

widely used as a single-component rocket fuel) [7]. The latter circumstance is especially important when placing a HF CWCL on board a space carrier. Since N_2H_4 is a fuel for microrocket engines of the spacecraft orientation system, it can also be used to generate hydrogen when the laser is operating.

Hydrazine is decomposed on the catalyst according to the reaction [8]

$$N_2H_4 \rightarrow 2xH_2 + {}^4/_3(1-x)\,NH_3 + {}^1/_3(1+2x)\,N_2 + Q, \qquad (1)$$

where $x = 0-1$ is the degree of decomposition of ammonia; and $Q = (80 - 44x)$ kcal mol^{-1} is the amount of heat released during the hydrazine decomposition. The active substance of the catalyst is a metal from the group Ir, Ni, Nb, Mo and Ru. It is applied to the porous granules of the carrier substance, which is alumina Al_2O_3. Such a catalyst is capable of actively decomposing hydrazine at room temperature. The decomposition of hydrazine is an exothermic process, and the temperature of its decomposition products can reach 1000 K. It is clear that the by-products formed here – nitrogen and especially ammonia – can negatively affect the energy characteristics of the HF laser. The calculation and experimental assessment of the nature and extent of this influence is the goal of this work.

2. Conditions for performing experiments

A methodically experimental study was organised in such a way as to individually evaluate the effect of each of the hydrazine decomposition products – H_2, N_2 and NH_3. At the same time, first benchmark tests were carried out in which pure hydrogen was used as a secondary fuel, then tests with the addition of nitrogen to hydrogen and then with the addition of ammonia in a mixture of hydrogen and nitrogen. The molar composition of the mixtures was selected so that the nitrogen and ammonia content in them corresponded to the content of these substances in the decomposition products of hydrazine on real catalytic bags, namely: 61 %–65 % H_2, 31 %–32 % N_2 and 3 %–5 % NH_3. The braking temperature was maintained at 300 K.

The experiments were carried out on a bench setup using a model of an HF laser with an estimated power of 5 kW, equipped with a flat nozzle unit that corresponds to the nozzle–nozzle reagent mixing scheme [9]. In the benchmark tests, we used a fuel composition with the conditional formula

$$[D_2 + \alpha F_2 + \psi(\alpha - 1)\,He] + \alpha_2(\alpha - 1)H_2, \qquad (2)$$
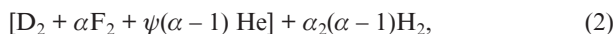
where $\alpha = n_{F_2}/n_{D_2}$; $\psi = n_{He}/(n_{F_2} - n_{D_2})$; $\alpha_2 = n_{H_2}/(n_{F_2} - n_{D_2})$ is the excess coefficient of secondary fuel (hydrogen) at the nozzle block section; and n_{H_2}, n_{F_2}, n_{D_2}, and n_{He} is the number of moles of hydrogen, fluorine, deuterium and helium.

In further tests, the hydrogen in the fuel composition (2) was successively replaced by the above mixtures. The ratio of the molar fractions of nitrogen and hydrogen in the model mixture was taken equal to their ratio in hydrazine: $n_{N_2}/n_{H_2} = 1:2$. The reproducibility of the experiments was ensured by three regimes, in which the two extreme regimes corresponded to the reference experiment, and the middle regime corresponded to the experiment with the addition of nitrogen or ammonia. In all tests, the composition of the working mixture in the atomic fluorine generator was kept constant and corresponded to the oxidiser excess coefficient $\alpha = 1.7$ and dilution degree $\psi = 10$, which were close to opti-

mal for the studied model of the HF laser, and the pressure $p_c = 0.09$ MPa corresponded to the maximum radiation generation power. The laser radiation power was measured by the closed resonator method using a mirror calorimeter (mirror diameter of 60 mm) with zero transparency according to the procedure [10] with an error of ±7 %. Part of the radiation emerging through a 2 mm diameter hole in the centre of one of the mirrors was directed to a Model 501 high-speed scanning spectrometer (OCLI Instruments). The spectrum was scanned in the range $\lambda = 2.6-3.2$ µm for a time of 1.25 s. An FSG-22-3A2 photoresistor based on germanium doped with gold, the signal of which was amplified and recorded by electronic (Tektronix-565) and light-beam (H-117) oscilloscopes, was used as a radiation receiver.

3. Results of experiments

In the first series of tests, we determined the dependences of the specific power $N_F = N/m_F$ and the specific energy extraction $N_\Sigma = N/m_\Sigma$ on the excess fuel ratio (pure hydrogen) α_2, where N is the absolute laser radiation power; m_F is the mass flow rate of free atomic fluorine; and m_Σ is the total mass flow rate of the fuel mixture.

The obtained dependences (Figs 1 and 2) indicate that the addition of nitrogen to the secondary fuel jet (pure hydrogen) leads to a significant decrease in the specific energy characteristics of the HF laser. Thus, the maximum specific power N_F, which characterises the efficiency of converting chemical energy into coherent radiation energy, decreases by 33 % (Fig. 1), and the maximum specific power extraction N_Σ decreases by 43 % (Fig. 2). In using an $H_2 + N_2$ mixture, the α_2
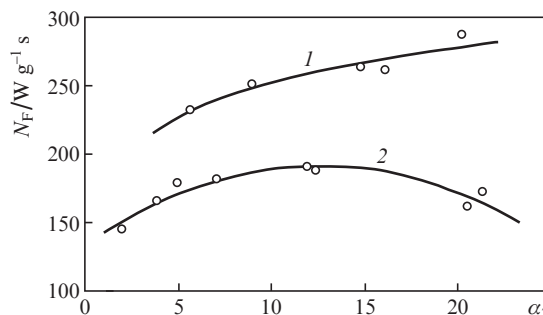

Figure 1. Dependences of the specific power N_F of the HF laser on the coefficient of the excess secondary fuel (pure H_2) when using (*1*) H_2 and (*2*) $H_2 + N_2$ mixture as a secondary fuel.
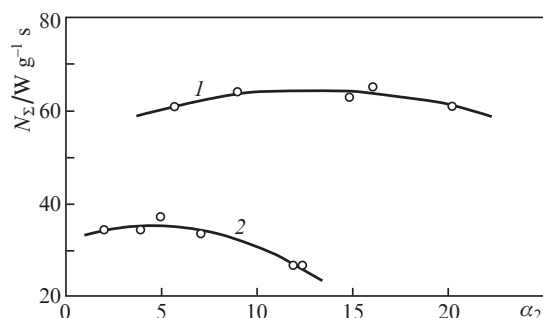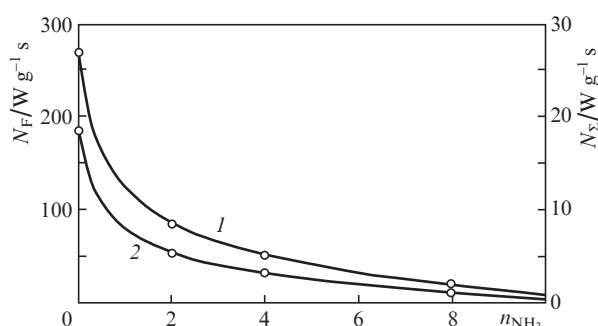

Figure 2. Dependences of the specific energy extraction N_Σ of the HF laser on the coefficient of the excess secondary fuel (pure H_2) when using (*1*) H_2 and (*2*) $H_2 + N_2$ mixture as a secondary fuel.

dependences of specific energy characteristics become more pronounced, and their maximum levels are achieved at significantly lower values of the coefficient α_2 than when using pure hydrogen as a secondary fuel.

The experiments to evaluate the effect of the additive in the secondary fuel of ammonia in the second series of tests were carried out at a fixed mass flow rate of the $H_2 + N_2$ mixture corresponding to the maximum specific power N_F determined in the previous series of tests. It turned out that the presence of ammonia in the secondary fuel jet greatly affects the specific energy characteristics of the laser: with an increase in the ammonia content in the mixture, the values of N_F and N_Σ decrease sharply, and with a volumetric composition of the mixture $60\%\,H_2 + 30\%\,N_2 + 10\%\,NH_3$ radiation generation is virtually absent (Fig. 3).

Figure 3. Dependences of the specific energy characteristics (*1*) N_F and (*2*) N_Σ of the HF laser on the volume ammonia content n_{NH_3} in the secondary fuel ($H_2 + N_2 + NH_3$ mixture).

The experimental data allow us to formulate preliminary requirements to the catalysts for the decomposition of N_2H_4 as applied to an HF laser.

4. Conditions for performing calculations and their results

To explain the obtained experimental results, we performed a series of calculations. The calculation analysis was based on a complex of mathematical models of processes occurring in the elements of the gas-dynamic path of the HF CWCL. The parameters of the combustion products at the output of the atomic fluorine generator were determined in the course of thermodynamic calculation with the experimentally known values of pressure p_c in the combustion chamber, the coefficient of excess oxidiser α and the degree of dilution of the mixture with helium ψ. The flow parameters at the nozzle

block section were determined from the calculation of the inviscid one-dimensional frozen flow in the nozzles of oxidising gas and secondary fuel. The parameters of the active medium in the laser chamber and the energy characteristics of the HF laser were calculated based on the approximation of the boundary layer according to the procedure [11] using kinetic data [12]. The use of such models related to lower dimensional models leads to an overestimation of the energy characteristics of the HF laser. However, given the purpose of this study, which is to assess the effect of by-products of hydrazine decomposition, and not to accurately determine energy characteristics, this circumstance should be considered insignificant. It is important that the computational resources required for the calculations are significantly reduced.

The calculation results are presented in Table 1. Calculation Nos 1 and 2 correspond to the conditions for carrying out benchmark tests and tests in which a mixture of hydrogen with nitrogen was used as secondary fuel (see Figs 1 and 2). Their results are in qualitative agreement with experimental data. Thus, the decrease in the specific energy characteristics of N_F and N_Σ with the addition of nitrogen to hydrogen in the experiment is 33% and 43%, respectively, and their calculated values decrease by 54% and 60%. The most probable reasons for this decrease are the following three factors: (i) vibrational–translational (VT) relaxation of vibrationally excited HF(v) molecules on nitrogen molecules; (ii) deterioration of the gas-dynamic conditions of mixing of the jets of oxidising gas (a mixture of $F + F_2 + DF + He$ from an atomic fluorine generator) and secondary fuel caused by a decrease in the rate of flow of the secondary fuel jet from the nozzles due to an increase in its molecular weight at a fixed nozzle geometry; and (iii) deterioration of the diffusion properties of the secondary fuel jet due to an increase in its molecular weight.

To assess the relative role of each of the listed factors, we performed calculation Nos 3 and 4. Calculation No. 3 was carried out without taking into account VT relaxation of vibrationally excited HF(v) molecules on N_2 molecules. It can be seen that the results of calculation Nos 2 and 3 virtually coincide, which indicates an insignificant role of this factor.

To assess the role of the gas-dynamic conditions for mixing reagents (differences in the rates of outflow from nozzles of jets of pure hydrogen and hydrogen diluted with nitrogen), we performed calculation No. 4. The rate of outflow from a nozzle of a mixture of hydrogen and nitrogen was assumed to be equal to the rate of outflow from a nozzle of pure hydrogen. One can see from Table 1 that the values of the specific energy characteristics of the HF CWCL significantly increased compared with their values according to the results of calculation No. 2.
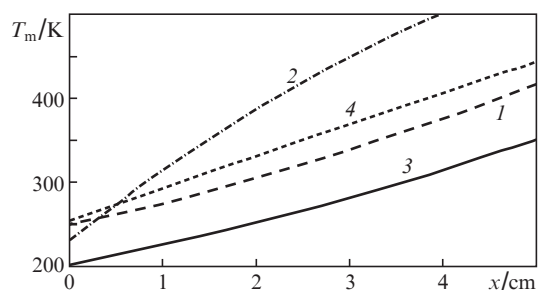
In order to understand the reasons for such a strong influence of the rate of the outflow of secondary fuel jets on the

Table 1. Results of calculations.

Calculation No.	Secondary fuel composition (% vol.)	α_2	u_{fuel}/m s^{-1}	T_{fuel}/K	u_{oxi}/m s^{-1}	N_F/W g^{-1} s	N_Σ/W g^{-1} s
1	$100\%\,H_2$	5.7	2370	300	2966	910	218
2	$67\%\,H_2 + 33\%\,N_2$	1.9	1027	300	2966	419	85
3	$67\%\,H_2 + 33\%\,N_2$	1.9	1027	300	2966	420	85
4	$67\%\,H_2 + 33\%\,N_2$	4.3	2370	300	2966	660	104
5	$67\%\,H_2 + 33\%\,N_2$	1.0	1846	1000	2966	543	111

Note: α_2 is the coefficient of the excess secondary fuel (hydrogen) at the nozzle block section; u_{fuel} and u_{oxi} are the rates of outflow from secondary fuel and oxidizing gas nozzles, respectively; T_{fuel} is the braking temperature of the secondary fuel jet.

laser energy, we should turn to the results of calculating the longitudinal distribution of the average consumption temperature T_m of the active medium in the laser chamber (Fig. 4). It can be seen that when using $H_2 + N_2$ as a secondary fuel mixture at $T = 300$ K [curve (2)], the temperature T_m significantly exceeds the temperature in the case of the same mixture, but with identical gas-dynamic parameters of the secondary fuel and oxidising gas [curve (3)]. It is well known [12] that a rise of the active medium temperature leads to deterioration in the energy characteristics of a CWCL. Consequently, a decrease in the specific energy characteristics of the HF laser after the addition of nitrogen to hydrogen is associated, first of all, with a sharp temperature rise in the mixing zone due to the increased role of viscous dissipation of the mechanical energy of the gas jet with an increase in the velocity difference between the mixed supersonic jets of oxidising gas and secondary fuel.

Figure 5. Dependence of the fluorine burnout in the laser chamber on the distance x when using (1) H_2 as a secondary fuel, (2) $H_2 + N_2$ mixture at a temperature of 300 K, (3) $H_2 + N_2$ mixture at equal flow rates of secondary fuel and oxidising gas jets from the nozzle and (4) $H_2 + N_2$ mixture at a temperature of 1000 K; c_{F_i} is the mass fraction of atomic fluorine in the ith section of the laser chamber; c_{F_0} is the mass fraction of atomic fluorine in the output section of the nozzle block.
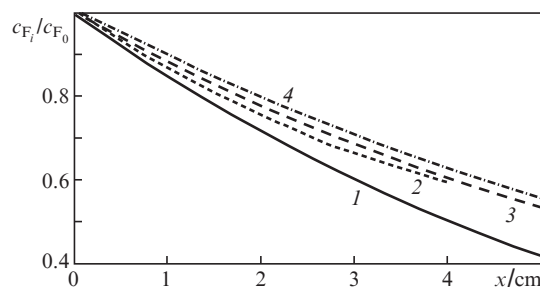

Figure 4. Longitudinal distributions of the average consumption temperature of the active medium in the laser chamber when using (1) H_2 as a secondary fuel, (2) $H_2 + N_2$ mixture at a temperature of 300 K, (3) $H_2 + N_2$ mixture at equal flow rates of secondary fuel and oxidising gas jets from the nozzle and (4) $H_2 + N_2$ mixture at a temperature of 1000 K; x is the distance from the output section of the nozzle block.

The third of the above factors also plays an important role in reducing the specific energy of the HF laser. This follows from the analysis of the burnup curves of atomic fluorine in the laser chamber (Fig. 5). The fluorine burnout rate for calculation No. 1 using pure hydrogen [curve (1)] turned out to be the highest. With the addition of nitrogen, the rate of burnout of fluorine atoms decreases [curve (2)], despite an increase in the average consumption temperature T_m of the active medium [see Fig. 4, curve (2)]. This circumstance unequivocally indicates that an increase in the molecular mass of the secondary fuel when nitrogen is added to the hydrogen jets leads to a decrease in its mixing rate with the jet of oxidising gas, to a decrease in the generation rate of vibrationally excited HF(v) molecules and to a decrease in the specific energy characteristics of the laser.

The most probable reasons for the sharp dependence of the specific energy characteristics of the HF laser on the ammonia content in the secondary fuel jet (see Fig. 3) may be the VT relaxation of vibrationally excited HF(v) molecules on NH_3 molecules or products of its interaction with fluorine, which, are possibly formed by mixing jets of secondary fuel and oxidising gas in the laser chamber, as well as their volume absorption of HF laser radiation.

The role of these factors is confirmed by an analysis of the emission spectra measured during the experiments. Eight transitions were identified from the spectrograms (indices indicate the lower transition level): P_0 (9–12) bands $v = 1 \rightarrow 0$

and P_1 (8–11) bands $v = 2 \rightarrow 1$ in the range $\lambda = 2.82–3.04$ μm. As the ammonia content in the active medium increases, two trends are clearly observed, i.e. intensive 'pumping' of power from the vibrational band $v = 2 \rightarrow 1$ to the band $v = 1 \rightarrow 0$ and a sharp decrease in the laser radiation power with virtually no change in the overall spectrum boundaries. It can be assumed that these processes are caused, on the one hand, by VT relaxation of vibrationally excited HF(v) molecules on NH_3 molecules:

$$HF(v = 2) + NH_3 \rightarrow HF(v = 1) + NH_3, \qquad (3)$$

$$HF(v = 1) + NH_3 \rightarrow HF(v = 0) + NH_3. \qquad (4)$$
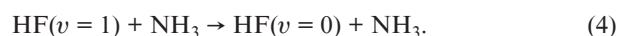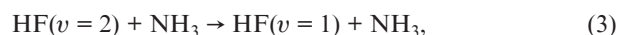
With increasing ammonia concentration, VT relaxation intensifies, and process (3) seems to be predominant, which ensures the redistribution of power between the bands due to increased inversion at the transitions of the $v = 1 \rightarrow 0$ band.

On the other hand, the volume absorption of laser radiation by ammonia molecules can significantly affect on the amount of the output power. Verification of this assumption was carried out as follows. We established in our experiments that the presence of NH_3 in an amount of more than 8% vol. in the active medium leads to a complete breakdown of lasing (see Fig. 3). If this effect is caused by volume absorption of radiation, then it will take place in the presence of ammonia not only in the active medium, but also outside it, for example, along the path of a laser beam to a radiation power meter. The latter scenario was implemented in special experiments when the HF laser was operated in the absence of impurities (pure hydrogen), but with the resonator mirror tubes purged with a mixture of $N_2 + 8\%$ vol. NH_3. The results of these experiments confirmed the presence of absorption of HF laser radiation by ammonia, especially by the P_1(8–10) lines, which is not surprising since these lines are included in the absorption spectrum of the NH_3 molecule [13]. As for the volume absorption of the HF laser radiation by nitrogen molecules, it is unlikely, because the generated lines do not fall into the absorption spectrum of the N_2 molecule [13].

The mixture of nitrogen and hydrogen obtained as a result of the catalytic decomposition of hydrazine is characterised by a rather high (1000 K and higher) temperature. In the preparation of model mixtures, this circumstance, due to technical difficulties associated with the preliminary heating of hydrogen, was not taken into account. Therefore, of inter-

est is an assessment of the influence of the braking temperature of the secondary fuel jet (a mixture of nitrogen and hydrogen) on the specific energy characteristics of the HF laser. To obtain such an assessment, calculation No. 5 was performed. In it, unlike calculation No. 2, the temperature of the secondary fuel was assumed to be 1000 K, close to the actual temperature of the hydrazine decomposition products. It turned out that the flow rate of the secondary fuel jet increased by 1.8 times. This led to a decrease in the difference in the velocities of the flow rates of the secondary fuel and oxidising gas jets, and, consequently, to a decrease in the average consumption temperature T_m of the active medium [see Fig. 4, curve (*4*)], which occurs despite an increase in the temperature of the secondary fuel at the nozzle block exit. This circumstance very favourably affects the specific energy characteristics of the HF CWCL and draws them 13% closer to the corresponding characteristics of a laser operating using pure hydrogen, but in the absence of ammonia.

5. Conclusions

The results of the study showed the fundamental possibility of using hydrazine decomposition products as a secondary fuel in a supersonic HF laser. When operating on cold (T = 300 K) model mixtures, it was found that the addition of 30% vol.−35% vol. nitrogen to the secondary fuel composition corresponding to its content in hydrazine decomposition products reduces the specific power N_F by 33%, and specific energy extraction N_Σ by 43% of the level obtained by using pure hydrogen in the optimal operation mode of the investigated HF laser model. Taking into account the actual heating of the products to 1000 K during the catalytic decomposition of hydrazine allows the specific energy level of the laser to approach the level achieved during its operation with pure hydrogen by 13%, which will lead to a decrease in specific energy extraction by only 30% in the absence of NH_3 decomposition products. If the nozzle unit of the HF laser is specially adapted for operation using the catalytic decomposition of N_2H_4 as a secondary fuel by equalising the rates of the secondary fuel and oxidising gas jets, this figure can be further reduced.

For the practical implementation of a liquid hydrogen generator based on the catalytic decomposition of hydrazine, it is necessary to exclude ammonia from the composition of its decomposition products. The solution to this problem involves the development of special reactors, catalysts and catalytic packages, a thorough design study and organisation of the thermal regime of the decomposition process, as well as the use of chemical ammonia absorbers. All these measures require experimental testing [14].

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