Modeling HF Gain Generator F-Atom Flows

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This technical report has been reviewed and is approved for publication. Publication of this report does not constitute Air Force approval of the report's findings or conclusions. It is published only for the exchange and stimulation of ideas.

[Signature]

1/Lt William Welser IV
SMC/MTZ
Control volume analysis and one-dimensional reacting gasdynamics have been combined with a unique thermal-mechanical model of a combustor vessel to estimate production of fluorine atoms at the nozzle exit plane. Upon reaction with hydrogen, H₂, these F-atoms are a key species in the efficient production of internally energetic hydrogen fluoride (HF) molecules, which is the lasing material for an HF chemical laser. Thus, a quantitative estimate of F-Atom density and more specifically the degree of fluorine (F₂) dissociation, α, is an essential parameter for the accurate prediction of laser performance. This parameter, as a function of time, has been difficult to experimentally measure or to analyze in such lasers.

The method has been applied to the only cylindrical HF chemical laser in operation. Test conditions for the most recent tests of the Alpha Laser are analyzed. It is found for the baseline operating point that α reaches a plateau exceeding 0.9 after a sufficiently long operating time. Necessarily for the expediency of rapid calculations, such a model lacks the temporal and spatial fine details of the heat transfer processes, gasdynamics, and boundaries. Therefore, such a model is valuable for insight, but must be used with care for detailed design.
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1. Introduction

The cylindrical or annular HF chemical laser known as the Alpha Laser has been previously described. In this report, we will concentrate on the gain generator part of the system, which produces the F-atom radicals necessary for the production of the lasant, HF*, namely, HF with inverted vibrational and rotational state distributions. The HF* is produced by the chemical reaction

\[ F + H_2 \rightarrow HF + H, \quad \Delta H = 31.6 \text{ kcal/moles} \] (1)

The gain generator is the cylindrical vessel containing the combustor or plenum generating the F-atoms from a hot, chemically reacting mixture at a steady stagnation temperature approaching 2000K. The hot gases are exhausted from the plenum through an annular nozzle array serving as the cylindrical walls of the plenum, in an essentially azimuthally uniform radial flow. The nozzles in the array are a converging-diverging type to generate a supersonic jet exhaust into a good background vacuum. The trailing edges of aerodynamic blades at the nozzle exit planes inject cold hydrogen into the hot stream. The gain medium is formed by free jet mixing with the hydrogen. The annular gain medium is approximately 50 cm in radius with a width or “thickness” of 2.5–3.5 cm. An annular resonator structure, the HEXDARR (High EXtraction Efficiency Decentered Annular Ring Resonator), extracts laser power from this medium over a length of approximately 400 cm in a double pass. It is apparent from Eq. (1) that laser power performance will be strongly proportional to the F-atom flow out of the nozzle exit planes, but it will depend somewhat on the plenum stagnation temperature, reaction kinetics, and the degree of F₂ dissociation at that plane. The plenum temperature depends sensitively on the combustion process, as we will see, and it determines to a great extent the F-atom production and also the static temperature in the free jet. Both gas properties affect the small signal gain coefficients of each spectral line in the flow direction. Laser power performance will also be affected by the presence of small amounts of F₂ created by recombination in the plenum and in the nozzle arrays when the plenum temperature is not high enough. In this case, the free H-atom generated by Eq. (1) can react with F₂ in the so-called “hot reaction” of the chain to raise significantly the static temperature of the free jet, effectively lowering small signal gains and even shortening the annular gain width by enhancing quenching collision kinetics. The H + F₂ reaction has an exoergicity of 91 kcal/mole, almost 3 times that of Eq. (1). Unfortunately, in contrast to Eq. (1), little of this large heat release leads to productive HF* gain for lasing.

Thus, a major goal of gain generator design has been to minimize the presence of F₂ in the free jet. This challenge is difficult to meet because space laser requirements virtually dictate a regeneratively cooled plenum of low mass content. Yet, the complex structure is of sufficient size that practical laser operation occurs in a transient thermal and mechanical condition, which is difficult to model. The empirical Alpha Laser experience has been essential for understanding any future design approaches.
A simple model for predicting \( F_2 \) degree of dissociation is sought to guide conceptual design or test planning. In this work, we propose to develop a simple, spreadsheet combustor model that focuses on predicting F-atom production across the nozzle exit planes. Our objective is to provide sufficient insight to help understand observed laser performance data on a device such as the Alpha Laser and also to help in developing combustor operating points for future laser devices. Our approach is the classical application of Control Volume analysis to describe the combustor process.\(^2\) The advantage of this method is that the practitioner defines the boundary of study to eliminate much of the structural and reactive flow intricacies. Once the Control Volume is defined, analysis must be developed for transient nozzle throat changes due to thermal effects and their linkages to combustor plenum pressure changes. The quantitative development of this linkage is the one unique aspect of this report. In such an approach, one deliberately sacrifices the spatial and temporal details that might influence an actual differential fluid element, in quest of averaged quantities that will reflect the global behavior of the system with acceptable fidelity.

As a demonstration of its application, the model will be used to compute the \( F_2 \) degree of dissociation, \( \alpha \), as a function of time for two tests with essentially the same flow rates in steady state, namely, HL902 and HL909B. A short model uncertainty discussion will follow.
2. Model Logic

The logic for building the thermal energy flux balance for the model is as follows:

1. Estimate thermal energy flowing in with reagents during interval $\Delta t$.

2. Estimate thermochemical energy flux released from reagents, assuming complete combustion.

3. Estimate net heat flux loss from the reagents or products during $\Delta t$ interval.

4. Estimate heat loss from combustor gases into the rings (and end domes) from temperature growth in the rings.
   a. Determine growth in combustor pressure $P_Q$ ($=P_C$) during the interval.
   b. Relate $P_Q$ growth to nozzle throat area $A^*$ closure. (1-D gasdynamics)
   c. Relate temperature growth in ring to $A^*$ decrease. (linear coefficient of thermal expansion)
   d. Compute net heat flux loss from gases to vessel, given vessel temperature growth.
   e. Estimate heat loss into combustor core (found negligible).

5. From NET heat flux from reagents and product gases (items 1–3), compute "stagnation" temperature $T_e$ of the combustor plenum. (i.e., no exhaust gasdynamics).

6. Compute F-atom density and F$_2$ degree of dissociation, $\alpha$, given $T_e$.

7. Slightly corrected for further recombination through the nozzle structure to exit plane (Appendix C)

The main innovation in this heat flux model is to use the mechanical changes of the combustor vessel as indicated by gasdynamics to deduce vessel temperature growth and, thereby, the net heat flux loss from the product gas, a quantity difficult to measure directly. This vessel temperature growth manifests the net heat flux to the vessel, the difference between heat loss from the product gases, and the regenerative cooling of the vessel by reagents. Since Items 1 and 2 are well known, we will concentrate on the development of Item 3 in the following sections.

It turns out that this model is applicable provided there are no mechanical changes in the vessel due solely to $P_c$ pressure changes. We will discover later that this assumption approaches validity beyond $t \geq 8$ s in the cases discussed here when the flows reach a true steady state and combustion is well established. (See Figure 3 and discussion.) An upgraded version of this model, which accommodates the detailed, fast transient behavior of $P_c$ and the role of the cold inner core of the vessel, is reported in References 9 and 10.
3. Control Volume

We assume in the time increment modeled (typically 1 s) that the flow is “steady.” The combustor gas properties such as molecular weight, \( W \), specific heat, \( c_p \), mole fractions of species, \( X_i \), and ratio of specific heats, \( \gamma \), are definable uniform quantities within a Control Volume space. These parameters for all species can be evaluated from the given reagent flowrates and gas properties and those same parameters deduced for the product species. Completed combustion and complete consumption of the oxidizer to products is assumed. There may be combustor gas property changes in succeeding time intervals due to slow changes in the flow rates of the reagents and the observed change in combustor pressure.

The choice of combustor Control Volume is illustrated in Figure 1. We attempt to choose the volume to avoid the need for specific three-dimensional details of the combustor vessel structure, including the heat flux into reagents during regenerative cooling of the vessel and the complex details of the actually mixing of reagents at an injector system before combustion.

The Control Volume suggests the balance equation for heat flux

\[
Q_{\text{dot} \text{out}} = Q_{\text{dot} \text{inflow}} + Q_{\text{dot} \text{thermochemical}} - Q_{\text{dot} \text{gas loss}}
\]  

(2)

![Diagram of Control Volume](image)

Figure 1. Illustration of the Control Volume chosen. The Control Volume is traced out by the dotted boundary, which intercepts the incoming pipes at the cryo-heat exchangers and the outflow to the nozzle throats.

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where

\[ Q_{\text{dot, inflow}} = \sum_i [ \text{mdot}_i c_p(T_{in,i}) ] \]

(3)

With mdot, the mass flow rate, \( c_p(T_{in}) \), the specific heat, and \( T_{in} \), the temperature at inflow of the ith reagent. We develop \( Q_{\text{dot, thermochemical}} \) and \( Q_{\text{dot, loss}} \) in successive sections below.
4. Thermochemistry

The combustion process begins with the early ignition by a burst of the hypergolic mixture of fluorine and deuterium. The bulk chemicals flowing into the plenum are nitrogen trifluoride, NF$_3$, and cryogenically cooled (to a nominal 80K) deuterium/helium mixture. In the combustion process, NF$_3$ readily dissociates at elevated temperature to produce F-atoms. A portion of these atoms reacts with D$_2$ to keep the combustor furnace going. At the plenum temperatures forecasted for the given Alpha Laser flowrates, the following chemical equation governs the process:

$$aD_2 + bNF_3 + cHe \rightarrow 2aDF + (1/2)bN_2 + \alpha (3b-2a)F + (1-\alpha)(3b-2a)F_2 + cHe,$$

where coefficients a, b, and c are mole fractions based on incoming flow rates, and $\alpha$ is the degree of dissociation of molecular fluorine.

The net heat of reaction, following Eq. (4), is

$$\Delta H_{net} = -a\Delta H_{D_2} - b\Delta H_{NF_3} - c\Delta H_{He} + 2a\Delta H_{DF} + 0.5b\Delta H_{N_2} + \alpha(3b-2a)\Delta H_{F}$$

$$+0.5(1-\alpha)(3b-2a)\Delta H_{F_2} + c\Delta H_{He}.$$

Of course, the heat of formation for He is moot, and those for D$_2$, F$_2$ and N$_2$ are zero. The heats of formation are taken at the condition of the gas crossing the Control Volume. $\Delta H$ for NF$_3$ is taken at 300K, and those for the combustion products, at a thermochemical temperature to be defined below. A consistent solution for the combustor will, therefore, require iteration, in which gas properties are adjusted as we home in to a temperature.

We determine

$$Q_{dot_{thermochemical}} = n_{dot_{in}} \cdot \Delta H_{net}.$$

Where $n_{dot_{in}}$ is the total molar flowrate of reagents. We also discovered in the course of building this model the high sensitivity of the computed results to very accurate thermodynamic data of the various species. We, therefore, resorted to look-up tables based on the established JANAF database$^{3,4,5}$ and to linear interpolation between the 100K temperature intervals of the database. (Ref. 5 is probably the most useful for providing digital data and functional extrapolations.)
5. Heat Flux Loss Term

The heat flux loss from the gases is given by

\[ Q_{\text{loss}} = Q_{\text{rings}} - Q_{\text{struts}} + Q_{\text{domes}} + Q_{\text{stagnant core}}, \tag{7} \]

with

\[ Q_{\text{rings}} = [Q_{\text{convective}} + Q_{\text{radiative}} - Q_{\text{regen}}]. \tag{7a} \]

\( Q_{\text{domes}} \) will become a small multiplicative correction factor on \( Q_{\text{rings}} \) in Eq. 16, while \( Q_{\text{struts}} \) and \( Q_{\text{stagnant core}} \) can be shown to be negligible heat conductors. Thus, evaluation of \( Q_{\text{loss}} \) centers on a good estimate of \( Q_{\text{rings}} \), the net heat flux gained by the rings.

In the time interval \( \Delta t \) of assumed "steady" flow, the total mass flow rates, individual mass flow rates, the specific heats, and the ratio of specific heats are all assumed constant. The construction of this model really assumes that input mass flow rates and, therefore, the output total mass flow rate remain constant in succeeding \( \Delta t \) intervals. Furthermore, the model assumes that in steady state, the vessel accrues no molecules; namely, the inflow of reagents equals the outflow of chemical products. This condition is only approached after 5 to 7 s following the ignition of the combustor. (Lasing begins about 5.5 s after ignition). The reason is that key reagent flows of NF\(_3\) and D\(_2\) must build up and are observed to asymptotically approach a true steady state in an exponential manner. Beyond this point in the actual laser, increases in pressure, \( P_o \), reflect mainly the result of thermal effects in the gas and the vessel. Then from the choked flow equation of one-dimensional gas dynamics the combustor pressure, \( P_o \), changes because of two effects: the square-root dependence on the stagnation temperature, \( T_o \), and more importantly, the inverse dependence on the throat area \( A^* \).

\[ P_o = C \sqrt{\frac{T_o}{A^*}}, \text{ where constant } C = \text{mdot} \sqrt{\frac{\gamma W}{R_o \left( \frac{2}{\gamma+1} \right)^{\gamma-1}}}, \tag{8} \]

where \( \text{mdot} \) is the total mass flow rate out of the vessel, \( \gamma \) is the ratio of specific heats, \( W \) is the molecular weight after combustion, and \( R_o \) is the universal gas constant. Properties \( W, \gamma, \) and \( c_p \) require calculations combining the individual properties taken at the gas stagnation temperature, \( T_e \), of the product species according to their mole fractions. For example:

\[ W = \left[ cW_{He} + 0.5bW_{N_2} + 2aW_{DF} + \alpha(3b - 2a)W_F + 0.5(1 - \alpha)(3b - 2a)W_{F_2} \right] \text{ndot}_{\text{in}}/\text{ndot}_{\text{out}}, \tag{9} \]
assuming full dissociation into F. Because the consistent combustor solution requires solving for \( T_o \), an iteration on properties must once again be performed to determine \( T_o \). In differential fractional terms for rate of change in the parameters, Eq. (8) becomes

\[
\frac{dP_o}{P_o} = -\frac{dA^*}{A^*} + 0.5 \frac{dT_o}{T_o}.
\]

From Eq. (10), one sees a direct relationship between a thermal gasdynamic parameter, \( P_o \), and a structural or mechanical parameter, \( A^* \), the throat area, which will be affected by thermal changes. We develop a second equation below to eliminate the fractional change in \( T_o \) (for the moment) such that a fractional change in \( P_o \) defines a fractional change in \( A^* \), and, therefore, the thermal condition of the rings in the vessel per unit time, from which \( Q_{\text{dot, rings}} \) can be derived.

In the Alpha Laser, \( A^* \) represents 25 primary nozzle throats plus two “half” nozzles on the ends of the ring stack. From Figure 2, we model one of the throats as \( \varepsilon \cdot c \), where \( c \) is the circumference of the ring, \( r \) is the radius, and \( \varepsilon \) is the slit width. Due to thermal effects, both may change dimensionally with time:

\[
A^* = n\varepsilon \cdot c = n\varepsilon \cdot 2\pi r, \ n = 26 \text{ throats.}
\]

This analysis is helped by the fact that there is absolutely no thermal expansion allowed in the direction of the gain generator cylinder’s axis, the z-direction, due to clamping referenced to massive invar rods of the Alpha Laser structure, as shown in Figure 2b.
Due to the clamping condition, the change in area of $A^*$ depends only on the thermal expansion of the half section of the ring cross section bordering a particular nozzle, and if one assumes all the expansions are identical for each of the 26 nozzles, $dA^* = n \cdot 2\pi r \cdot de + n \cdot e \cdot 2\pi dr$.

Between the rigid centerlines in Figure 2a, any shrinkage in $e$ is due to thermal expansion of the two material half-lengths $L_i$. (Note $L_i$ is not the geometric half length): $de = -2dL_i$, and it follows that

$$dA^*/A^* = -2 \left[ (dL/L_i) \cdot (L/e) \right] + dr/r.$$  \hspace{1cm} (12)

It can be shown that $L_i/e$, taken constant in our calculations, is a very slowly varying function of time $t$, on the order of 2% increase during a typical test. (See Appendix B: Calculation of $L/e$.) The thermal linkage is through the coefficient of thermal expansion, CTE, to find an average temperature increase in combustor vessel rings:

$$dL/L_i = CTE \cdot \Delta T_{vessel}.$$  \hspace{1cm} (13)

Using the perfect gas law to describe the combustor gases yields another relationship for $dP_o/P_o$. With this additional relationship, $T_o$ and $r$ can be eliminated with Eqs. (10) and (12) to yield the sought relationship, deduction of the temperature change in the material based on merely the change in the combustor plenum pressure:

$$\Delta T_{GGA} = (\Delta P_o/P_o) + [4 \cdot CTE \cdot (L/e)]$$  \hspace{1cm} (14)

The estimation of the heat transfer out of the hot combustor gases to the cylindrical walls of the vessel depends on accurate knowledge of the mass of a gain generator ring. (See Appendix A: Mass Data.) Assuming the convective heat transfer process at the boundary of the end caps is the same, one also needs a knowledge of the mass of the end caps (i.e., the domes) of the vessel. The ring mass is self cooled or regeneratively cooled by the incoming cold reagents. In the Alpha Laser, the end caps are water cooled, providing a slight enhancement of heat transfer out the end domes.

There is a detail in handling the end domes heat transfer. As noted in Appendix A, the end-domes mass has been taken as 15% of the mass of the rings. The idea that the end domes bound both an active annular convective flow region and a stagnating core has been included. Because the domes are water cooled, an additional multiplicative correction, $f$, to Eq. 16 is

$$f = \left\{ 1 + 0.15 \cdot \left[ (T_{Al} - T_{water})/(T_e - T_{water}) \right] \right\}/1.15,$$  \hspace{1cm} (15)

with $T_{Al}$ as the ring temperature at the given time interval, $\Delta t$. Generally, $T_{water}$ equals 300K.

The heat transfer rate $Q_{dot}$ (watts) to all parts of the aluminum vessel then is given globally by
\[ Q_{\text{dot}, A} = M_{A} \cdot C_{p_{-A}} \cdot \Delta T_{A} \cdot f/\Delta t, \]  

(16)

with the vessel mass being given by \( M_{A} \), the specific heat of aluminum \( C_{p_{-A}} \), \( \Delta T_{A} \) by Eq. (13), \( f \) by Eq. (14), and the time interval, \( \Delta t \), of heat transfer for the \( \Delta P_{o} \) observed, namely, 1.0 s. The Control Volume approach combined with ideas relating to Eq. (7) link vessel material thermal growth to the combustor gas loss:

\[ Q_{\text{dot}, A} = Q_{\text{dot, gas loss}} \]  

(17)
6. Calculation of the Stagnation Temperature \( T_e \)

Finally, by the heat flux balance of Eq. (2), gas stagnation temperature, \( T_e \), in the combustor is

\[
T_e = \frac{(Q_{\text{inflow}} + Q_{\text{thermochemical}} - Q_{\text{gas loss}})}{(\text{mdot} \times c_{p-gas})}, \tag{18}
\]

where \( \text{mdot} \) is the measured mass flow rate of products from the combustor and \( c_{p-gas} \) is the specific heat at constant pressure of products computed according to accurate thermochemical data and mole fractions defined by products (e.g. Eq. 9). Gas properties are computed at \( T_e \). \( Q_{\text{inflow}} \) is the heat flux into the Control Volume due to heat capacity of reagents. \( Q_{\text{thermochemical}} \) is the heat flux added to the flow as products after complete combustion. Because the gas properties of products are computed at \( T_e \), iterations are also needed here to arrive at a consistent solution for \( T_e \), including combustor degree of dissociation effects.

In fact, it should be realized that this stagnation temperature is really a mythical "average" exactly defined by Eq. (18) within the context of one-dimensional gasdynamics analysis. The real gas condition is non-uniform. A differential fluid volume may experience a temperature up to a hypothetical "maximum" combustor temperature (sometimes called \( T_i \)) that could be computed without the "gas loss" heat flux term, but adding the heat flux of the reagents used in regenerative cooling. In such a simple model, we view \( T_e \) as an average "one-dimensional" stagnation temperature of the gas flowed through the nozzles into the laser cavity.
7. Estimation of F-Atom Production

For the given time interval, $\Delta t$, once $T_c$ is found, the estimation of F-atoms and molecular $F_2$ in the combustor follows conventional equilibrium chemistry calculations with $T_c$ as the defined flow “equilibrium” temperature. Inside the combustor, it is assumed that flow effects are negligible and there is sufficient dwell time for the mixture to equilibrate chemically. It has to be confirmed that there are no major bulk fluid dynamical recombination effects to generate $F_2$ through the nozzles. Indeed, there is a minor correction of flows from the combustor to the nozzle throat because fluid particle times are sufficiently long to allow some recombination. However, from the throat to the exit, the flow becomes on the average hypersonic, and any effects can be shown to be negligible (Appendix C). It can be shown that loss of F-atoms by wall and boundary layer recombination effects is negligible for this nozzle design, based on wall data discussed by N. Cohen et al.\textsuperscript{6} Thus, the principal loss of F-atoms is presumed to occur by three-body bulk recombination in the presence of diluents and other exhaust products.

The dissociation of $F_2$ into F-atoms in this model is given by

$$k_f = 5 \times 10^{13} \sigma \exp\left(-35,000/R_0 T_0\right) \text{ (cm}^3\text{-mole}^{-1}\text{-s}^{-1}) \text{, (19)}$$

$\sigma = 1$ for all collision partners except He, $\sigma = 2$, $F_2$ itself, $\sigma = 2.7$, and F-atoms, $\sigma = 10$. The specific reference is N. Cohen and J. F. Bott.\textsuperscript{7} Instead of the more commonly JANAF-accepted bond strength of 38 kcal-mole$^{-1}$, we have used 35 kcal-mole$^{-1}$ as recommended by Cohen. The backward rate coefficient for this model is fitted to the early data summary\textsuperscript{6}

$$k_b = (2.6 \times 10^{16})/T_e^{1.05} \text{ (cm}^6\text{-mole}^{-2}\text{-s}^{-1}) \text{, (20)}$$

and the equilibrium constant $K$ is simply $k_f/k_b$. The maximum number of F-atoms (i.e., the F-atom density $[F_0]$) available in the combustor can be computed following Eq. (4) with unity degree of dissociation and for very high $T_e$, given the gas properties of the gas. For the 1-s interval of apparent steady state, this is equivalent to the flowrate of F atoms that will leave the plenum as well. However, the actual number of F-atoms in the combustor that makes up the desired F-atom flow rate depends on $T_e$. The solution for the actual F-atom density $[F]$ is quadratic, for the positive branch

$$[F] = 0.25 \times K \left\{1 + \sqrt{1 + 8F_0/K}\right\} \text{, (21)}$$

It follows that in the given local fluid particle, density $[F_2]$ and degree of dissociation, $\alpha$, are

$$[F_2] = 0.5 \left\{[[F_0] - [F]] \right\} \text{, (22)}$$
\[
\alpha = \left\{1 + 2 \cdot \left(\frac{[F_2]}{[F_1]}\right)\right\}^{-1}
\]  

(23)

The admittedly large uncertainty in the activation energy of Eq. (19) and the empirical curve fit in Eq. (20) mean the uncertainties in the estimations of F-atom and F\(_2\) densities will remain high except in the vicinity of unity \(\alpha\). Model uncertainties will be discussed further below. The efficient HF continuous-wave chemical laser usually requires the suppression of F\(_2\) species. This is achieved by ensuring sufficiently high \(T_e\), in the combustor. For a space system, a requirement for high \(T_e\) must be traded against the mass increase in reagents for producing the hotter gain generator.
The model is demonstrated using recent Alpha Laser input test data from Tests HL902 and HL909B. We caution at the onset that the model prediction of degree of dissociation $\alpha$ or of F-atom flow rate will remain unverified, for there are no independent direct experimental measurements. Nevertheless, certain observed behaviors of laser power as a function of time in HL902 and HL909B are consistent with model findings. Table 1 summarizes the flow condition for HL909B when the reagents have reached a near steady state at the onset of lasing ($t = 8$ s). In reality, both NF$_3$ and D$_2$ increase 1% or less during the time of lasing. Flow conditions are similar for HL902.

The key test observable is the measured combustor plenum pressure as a function of time from zero seconds to reagents shutoff. In the Alpha Laser test series, data are taken at 0.25-s intervals. This work will use data only at each 1.0-s interval for most of our cases because the rate of change of the pressure in this interval is sufficiently small, typically 5% or less. The estimated errors on this measurement are 3% or less.

From the HL909B combustor pressure $P_c$ as a function of time, one can deduce the apparent heat loss to the vessel for each 1-s block of the study. From the computed heat fluxes for a given $t$, $\Delta t$ block (Eqs. 3, 6, 16, 17), one can deduce the probable $T_e$ based on Eq. (18). This parameter is found to have a considerable range as a function of time, from 1423K to 1563K in this computation during lasing, from $t = 9$ s to $t = 15$ s. This spread is reflected in the computations for degree of dissociation $\alpha$, as exhibited in Figure 3. (diamonds)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>NF$_3$ Flowrate, mol/s</td>
<td>112.9 Entry at 300K</td>
</tr>
<tr>
<td>D$_2$ Flowrate, mol/s</td>
<td>111.5 Entry at 80K</td>
</tr>
<tr>
<td>He Flowrate, mol/s</td>
<td>573.5 Entry at 100K, taken as 80K</td>
</tr>
<tr>
<td>Reagents Molecular Weight, g/mol</td>
<td>13.50</td>
</tr>
<tr>
<td>Reagents Specific Heat $c_p$, J/g-K</td>
<td>1.928</td>
</tr>
<tr>
<td>Reagents Sp. Heat Ratio, $\gamma$</td>
<td>1.470</td>
</tr>
<tr>
<td><strong>Combustion Products</strong></td>
<td></td>
</tr>
<tr>
<td>Product: max-F (or F$_0$), mole/s</td>
<td>117.2 Complete reaction assumed</td>
</tr>
<tr>
<td>Product: N$_2$, mole/s</td>
<td>56.44 No recombination or loss</td>
</tr>
<tr>
<td>Product: DF, mole/s</td>
<td>221.4</td>
</tr>
<tr>
<td>Product Molecular Weight, g/mol</td>
<td>11.11</td>
</tr>
<tr>
<td>Product Specific Heat $c_p$, J/g-K</td>
<td>2.062</td>
</tr>
<tr>
<td>MdotF$_0$/A, kg/m$^2$</td>
<td>0.330</td>
</tr>
<tr>
<td>Product Sp. Heat Ratio, $\gamma$</td>
<td>1.561</td>
</tr>
</tbody>
</table>
The degree of dissociation or $\alpha$ curve labeled “Model (lasing),” from 9 to 15 s, exhibits an approach to “steady state” for times greater than 10 s, with $\alpha$ approaching 0.90. Here, the $P_c$ and reagent flow rate data are taken from test data 1, 8 from HL909B. Since data used are in 1-s intervals of time, high-frequency temporal details are disregarded in this model.

Comparison of HL902 degree of dissociation results can be made with those of HL909B. The model can be run with HL902 pressure and flow conditions, which are very similar. HL902 $\alpha$’s (grey triangles, labeled “HL902 Model”) are slightly lower than those of HL909B, but within 5%. The good agreement is confirmation of the contention that test reagent flow rates have been the same for at least 10 tests in the series, spanning HL902 to HL909B. Comparison can also be made with a contractor model (squares, labeled “TRW HL909B”) based on a similar heat flux balance approach, which uses data from embedded thermocouples in the rings. The TRW model seems to predict higher $\alpha$ at earlier times averaged over a much more steady state, possibly an indication of insensitivity of the thermocouples to more rapid changes in gas flow conditions. The thermocouples are highly sensitive to vessel thermal non-uniformities and the quality of their placements. The trend toward agreement in degree of dissociation (within 10%) of two model approaches at late times past 10 s. suggests progress toward prediction of this elusive key parameter.

We have included a fourth computation (Model P-3)9, 10, which attempts to synthesize the transient degree of dissociation in HL909B toward steady state in the case of both pressure and thermal transient changes. As we suggested earlier, both the current and the advanced models converge to the same dissociation results as flows reach a steady state, at $t \geq 8$–10 s.
The absence of direct measurements for F-atoms or $\alpha$ at the nozzle array exit plane makes model validation difficult. We must resort to plausibility discussions that demonstrate the model's consistency with other observed data. We examined three types of data:

1. Typical thermocouple data for the rings
2. Spectra behavior as a function of time beyond 11 s
3. $P(t)$, outcoupled power as a function of time, seen to be slowly increasing

In all cases, the model seems to provide insight into data variations or demonstrate consistency.

In the Alpha Laser, thermocouples were mounted manually in various gain generator ring positions. Although the absolute values had considerable scatter, they seemed to exhibit time constants longer than a fraction of a second, and a common trend appeared to be an average temperature increase of 15K/s over most of the running time. The model prediction from Eq. (14) is shown in Figure 4, together with a comparison to a 15K/s slope line. It can be seen that there is qualitative, even quantitative agreement over a wide span of time.

The observation of spectra at times greater than approximately 11 s in a lasing test can be seen from the data of relative laser power as a function of time, as in Figure 5. The framed region corresponds to a lasing time region when $\alpha$ has not reached a steady state. For times greater than around 10.5 s, it can be seen that most spectral lines are steadier as a function of time.

The best demonstration of an approach to steadiness is $P_2(6)$ and $P_2(7)$ while $P_1(7)$ declines slightly. Steady spectral signals are an indication that laser power has attained a steady condition. In turn, this implies proportionally that the F-atom flowrate and therefore $\alpha$ has become constant for longer times.

![Estimated Average Vessel Temperature](image)

Figure 4. Growth in ring temperature computed by the model. The line delineates 15K/s.
as forecast by our model. The increases as a function of time in $P_1(5)$ and $P_2(5)$ have also been correctly attributed to gain generator thermal ring growth. Ring growth pushes the gain width outward to better couple with the mode width of the resonator and away from the obstruction caused by protective clippers at the nozzle exit plane. Lower $J$ lasing lines such as $J = 5$ are more sensitive to this thermal growth because of the sharper peaked spatial distributions of the low $J$ small-signal gains upstream in the medium. However, for HL909B, $P_1(6)$ experiences an anomalous "dip" at 13.5 s before recovering. Thus, in fact, HL909B data are not as convincing a validation as some earlier spectral data, such as HL909A, in showing steadier power as $\alpha$ approaches a steady value at long operating times.

The third check is on observed laser power as a function of time, $P(t)$, as shown in Table 2. In this table, the reported outcoupled laser power from test HL909B is normalized to its maximum observed value, between 12 and 14 s. To a zeroth-order-level approximation, we assume that the laser power extracted must be proportional to the F-atom flow into the resonator, and therefore degree of dissociation, normalized to our computed quantity, at 14 s.

This normalization can be compared with the actual similar trend in normalized data to suggest that the slight empirical increase in laser power as function of time, $P(t)$, might be due partly to the slow increase to steady state of parameter $\alpha$. As noted earlier, ring growth also contributes to $P(t)$ increase, and may be the reason for the larger part of the $P(t)$ increase after 10 s. Nevertheless the time-dependent pressure curve, $P_c$, plus slight temporal increase in reagent flows generate a performance prediction from our model consistent with an observed power trend.
Table 2. Comparison of Adjusted Model to Normalized HL909B Outcoupled Power

<table>
<thead>
<tr>
<th>Time (s)</th>
<th>Normalized</th>
<th>HL909B</th>
</tr>
</thead>
<tbody>
<tr>
<td>9</td>
<td>0.86</td>
<td>0.71</td>
</tr>
<tr>
<td>10</td>
<td>0.96</td>
<td>0.87</td>
</tr>
<tr>
<td>11</td>
<td>0.92</td>
<td>0.96</td>
</tr>
<tr>
<td>12</td>
<td>0.96</td>
<td>1.00</td>
</tr>
<tr>
<td>13</td>
<td>0.98</td>
<td>1.02</td>
</tr>
<tr>
<td>14</td>
<td>1.00</td>
<td>1.00</td>
</tr>
</tbody>
</table>

Outcoupled powers are normalized to values at t = 14 s.

The general trends exhibited by the three empirical checks suggest that the model is consistent with observations. However, verification remains inconclusive without more data and experiments.
9. Model Uncertainty

The exponential structure of the equations for F-atom density, Eqs. 19–20 combined, suggest that any calculation of that parameter using empirical data will be subjected to accumulating errors in the F$_2$ to F density ratio in Eq. (23). If one supposes that the ratio of densities of F$_2$ to F-atoms can be established to the reasonable uncertainty of ±20%, then the fractional uncertainty, $\Delta\alpha/\alpha$, can be shown to depend on the value of $\alpha$ as in Figure 6. The uncertainty is computed assuming a root-sum-square fractional error dependence for independent error sources in experiments that are not repeated.

For example, the uncertainty in estimating $\alpha = 0.9$ is ±2.5%. On the other hand, if $\alpha$ is computed to be 0.4, the uncertainty is ±12%. The uncertainty in knowing $\alpha$ approaches the uncertainty in establishing the F$_2$/F density ratio for very small values of $\alpha$. For any value of $\alpha$, the density of F$_2$ is difficult to establish even though the total number of F-atoms can be easily computed from thermochemical calculations. There is obviously less uncertainty in this density as $\alpha$ approaches unity; thus, it is clear that the model is most useful for degrees of dissociation approaching unity.

![Error in Degree of Dissociation Computation assuming 20% error in density ratio [F$_2$/F]](image)

Figure 6. The uncertainty in computing degree of dissociation, $\alpha$. 

**Error in Degree of Dissociation Computation assuming 20% error in density ratio [F$_2$/F]**

- $\delta\alpha/\alpha$ vs. $\alpha$ for $\pm 20\%$ uncertainty.

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10. Closure

In reality, accuracy limitations of experimental laser data and the chemical kinetic database, especially the high sensitivity of equilibrium constant K to small uncertainties, may make it futile to strive for high accuracy in the density ratio. Then the utility of such models for detailed design is necessarily limited. We contend that such models are important, perhaps even invaluable, tools for guidance and insight, but may not be very useful for detailed design or planning except when combustor temperatures are set sufficiently high, with the degree of fluorine dissociation nearing unity.

The reader is cautioned that although this model is approximately correct as the combustor approaches "steady state," subsequent work generated a better formulation to handle the large unsteadiness in flow parameters at the onset of ignition and the approach to this quasi-steady-state condition. It was also later found that the combustor stagnation pressure, $P_c$, could not be successfully modeled as a function of time without developing the concept of a colder inner cylindrical core in this model. These adjustments are contained in the latest and final versions of the model.
Appendix A—Mass Data for the Alpha Gain Generator

The following mass data was acquired from the Alpha Laser Optimization program.

<table>
<thead>
<tr>
<th>Component</th>
<th>Mass (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Normal Ring</td>
<td>11.6 kg</td>
</tr>
<tr>
<td>End Ring</td>
<td>11.0 kg</td>
</tr>
<tr>
<td>Sub assemblies per ring</td>
<td>0.85 kg</td>
</tr>
<tr>
<td>Domes</td>
<td>1.15 kg</td>
</tr>
</tbody>
</table>
Appendix B—Calculation of $L/\varepsilon$ as a Function of Run Time

In a somewhat circular argument, we assume $L/\varepsilon$ is nearly constant, with $L = L_o = 18.2 \text{ mm}$ and $\varepsilon = \varepsilon_o = 0.6634 \text{ mm}$ defined by the system geometry. By Eq. (14), we can estimate $\Delta T$-GGA. Using this temperature, we can estimate the increase in $L$ (Eq. 13) and the decrease in $\varepsilon$ (Eq.12) as a function of time.

<table>
<thead>
<tr>
<th>$t$ (s)</th>
<th>$p_o$ (psia)</th>
<th>$\Delta t$ (1 s)</th>
<th>$\Delta p_o/p_o$</th>
<th>$\Delta T$-GGA</th>
<th>$\varepsilon$ (mm)</th>
<th>$L$ (mm)</th>
<th>$L/\varepsilon$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>30</td>
<td>4 to 5</td>
<td>0.0933</td>
<td>37.0</td>
<td>0.6638</td>
<td>18.20</td>
<td>27.42</td>
</tr>
<tr>
<td>5</td>
<td>32.8</td>
<td>5 to 6</td>
<td>0.1006</td>
<td>39.0</td>
<td>0.6621</td>
<td>18.22</td>
<td>27.51</td>
</tr>
<tr>
<td>6</td>
<td>36.1</td>
<td>6 to 7</td>
<td>0.0443</td>
<td>22.1</td>
<td>0.6595</td>
<td>18.24</td>
<td>27.66</td>
</tr>
<tr>
<td>7</td>
<td>37.7</td>
<td>7 to 8</td>
<td>0.0557</td>
<td>17.6</td>
<td>0.6584</td>
<td>18.25</td>
<td>27.72</td>
</tr>
<tr>
<td>8</td>
<td>39.8</td>
<td>8 to 9</td>
<td>0.0477</td>
<td>22.1</td>
<td>0.6576</td>
<td>18.26</td>
<td>27.76</td>
</tr>
<tr>
<td>9</td>
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<td>9 to 10</td>
<td>0.0432</td>
<td>17.1</td>
<td>0.6568</td>
<td>18.26</td>
<td>27.81</td>
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<tr>
<td>10</td>
<td>43.5</td>
<td>10 to 11</td>
<td>0.0460</td>
<td>18.2</td>
<td>0.6559</td>
<td>18.27</td>
<td>27.86</td>
</tr>
<tr>
<td>11</td>
<td>45.5</td>
<td>11 to 12</td>
<td>0.0352</td>
<td>13.9</td>
<td>0.6553</td>
<td>18.28</td>
<td>27.89</td>
</tr>
<tr>
<td>12</td>
<td>47.1</td>
<td>12 to 13</td>
<td>0.0382</td>
<td>15.2</td>
<td>0.6546</td>
<td>18.28</td>
<td>27.93</td>
</tr>
<tr>
<td>13</td>
<td>48.9</td>
<td>13 to 14</td>
<td>0.0225</td>
<td>8.9</td>
<td>0.6542</td>
<td>18.29</td>
<td>27.95</td>
</tr>
<tr>
<td>14</td>
<td>50</td>
<td>14 to 15</td>
<td>0.0200</td>
<td>7.9</td>
<td>0.6538</td>
<td>18.29</td>
<td>27.98</td>
</tr>
</tbody>
</table>

From $t = 4$ s to $t = 15$ s, $L/\varepsilon$ increases only around 2%, thus validating our assumption that $L/\varepsilon$ is nearly constant.

We have noted through References 9 and 10 that estimating $\Delta T$-GGA by this method for early times may not be exactly correct because time-dependent changes in $p_o$ may depend on flow variations as well as the thermal effects of this model.
Appendix C—Estimate of Volumetric F-Atom Loss from Plenum to Exit

We contended that the volumetric loss in a one-dimensional nozzle flow from plenum to exit of F-Atoms is around 1%. The contour pattern, A/A* for the HYWN nozzle was made available by the contractor. This does not include boundary layer effects. The table below summarizes our result for a high degree of fluorine dissociation in the plenum. Parameter “x” is flow in the radial direction, “A/A*” is the nozzle area expansion, “M” is the Mach number, “T/T_o” is the relative static temperature at station x, “[F]” is the molar concentration of F-atoms (Eqs. 19–21), and “alpha” (Eq. 23) is the degree of dissociation. The ratio of alpha at x = 27.3 mm and x = 0 mm (plenum) shows less than a 1% loss.

<table>
<thead>
<tr>
<th>x(mm)</th>
<th>A/A*</th>
<th>M</th>
<th>T/T_o</th>
<th>[F]</th>
<th>alpha</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>7.41</td>
<td>0.08</td>
<td>0.9983</td>
<td>1.9371E-06</td>
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<tr>
<td>1.1</td>
<td>6.48</td>
<td>0.09</td>
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<td>2.2</td>
<td>4.63</td>
<td>0.12</td>
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<td>1.9008E-06</td>
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<tr>
<td>3.3</td>
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<td>0.21</td>
<td>0.9879</td>
<td>1.8652E-06</td>
<td>0.959</td>
</tr>
<tr>
<td>4.4</td>
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<td>0.40</td>
<td>0.9571</td>
<td>1.7583E-06</td>
<td>0.958</td>
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<tr>
<td>5.7</td>
<td>1.00</td>
<td>1.00</td>
<td>0.7810</td>
<td>1.2212E-06</td>
<td>0.958</td>
</tr>
<tr>
<td>0.0</td>
<td>1.00</td>
<td>1.00</td>
<td>0.7810</td>
<td>1.2217E-06</td>
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</tr>
<tr>
<td>1.1</td>
<td>3.45</td>
<td>3.04</td>
<td>0.2790</td>
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<tr>
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<td>3.23</td>
<td>0.2553</td>
<td>1.6839E-07</td>
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<tr>
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<td>3.94</td>
<td>0.1871</td>
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<tr>
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<td>4.48</td>
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<tr>
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<td>4.69</td>
<td>0.1396</td>
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</tr>
<tr>
<td>6.8</td>
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<tr>
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<tr>
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<tr>
<td>20.4</td>
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<td>6.46</td>
<td>0.0787</td>
<td>2.0433E-08</td>
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</tr>
<tr>
<td>22.7</td>
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<td>6.55</td>
<td>0.0767</td>
<td>1.9528E-08</td>
<td>0.958</td>
</tr>
<tr>
<td>25</td>
<td>32.61</td>
<td>6.89</td>
<td>0.0738</td>
<td>1.8222E-08</td>
<td>0.958</td>
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<tr>
<td>27.3</td>
<td>34.99</td>
<td>6.83</td>
<td>0.0709</td>
<td>1.6966E-08</td>
<td>0.958</td>
</tr>
</tbody>
</table>

In this calculation, we have taken plenum conditions to be T_o = 1858K, p_o = 39.8 psia, \( \gamma = 1.56 \), and \( W = 11.1 \text{ g-mol}^{-1} \) as determined by mole fractions of products (See, for example, Table 1). Initial alpha= 0.964 in the plenum.
References


5. Ibid. NIST Standard Database 13 or NIST13.


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