NONEQUILIBRIUM COMBUSTION EFFECTS IN SUPersonic STREAMS

by

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# TABLE OF CONTENTS

SUMMARY .................................................................................. 1

INTRODUCTION ........................................................................... 2
  Scope of Present Investigation ................................................ 3

ANALYTICAL PROGRAM ................................................................. 4

EXPERIMENTAL PROGRAM .......................................................... 19
  Gas Generator ........................................................................... 19
  Hydrogen Heater ..................................................................... 21
  Supersonic Test Section .......................................................... 21
  Experimental Results ............................................................. 21
  Gas Generator Experiments .................................................... 24
  Supersonic Combustion Experiments ...................................... 24

DISCUSSION OF ANALYTICAL AND EXPERIMENTAL RESULTS .... 35

CONCLUSIONS ............................................................................ 43

BIBLIOGRAPHY ............................................................................. 44
# LIST OF FIGURES AND TABLES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Schematic of the Ignition Model</td>
<td>5</td>
</tr>
<tr>
<td>2</td>
<td>Equilibrium and Finite-Rate Compositions for G.G. Products</td>
<td>6</td>
</tr>
<tr>
<td>3</td>
<td>Analytical Mechanism for Investigation</td>
<td>8</td>
</tr>
<tr>
<td>4</td>
<td>Ignition Delay for Base Case Species</td>
<td>9</td>
</tr>
<tr>
<td>5</td>
<td>Effect of Temperature on Induction Times</td>
<td>10</td>
</tr>
<tr>
<td>6</td>
<td>Effect of Equivalence Ratio on Delay Times</td>
<td>11</td>
</tr>
<tr>
<td>7</td>
<td>Effect of Water Vapor on Induction Times</td>
<td>13</td>
</tr>
<tr>
<td>8</td>
<td>Ignition Delay Times for Different Correlations</td>
<td>14</td>
</tr>
<tr>
<td>9</td>
<td>Comparison of Analytical and Experimental Results</td>
<td>15</td>
</tr>
<tr>
<td>10</td>
<td>Ignition Delay for Vitiated Air/Hydrogen</td>
<td>17</td>
</tr>
<tr>
<td>11</td>
<td>Ignition Delay for Heated Air/Hydrogen</td>
<td>18</td>
</tr>
<tr>
<td>12</td>
<td>Schematic of Experimental Set-Up</td>
<td>20</td>
</tr>
<tr>
<td>13</td>
<td>Gas Generator Assembly</td>
<td>22</td>
</tr>
<tr>
<td>14</td>
<td>History of Ignition Phenomenon of Vitiated Air and Hydrogen</td>
<td>26</td>
</tr>
<tr>
<td>15</td>
<td>Ignition Delay Measuring Apparatus</td>
<td>29</td>
</tr>
<tr>
<td>16</td>
<td>Ignition Delay - Run 41</td>
<td>30</td>
</tr>
<tr>
<td>17</td>
<td>Ignition Delay - Run 43</td>
<td>31</td>
</tr>
<tr>
<td>18</td>
<td>Graph Ignition Delay Versus H₂ Temperature</td>
<td>33</td>
</tr>
<tr>
<td>19</td>
<td>Graph Ignition Delay Versus Equivalence Ratio</td>
<td>34</td>
</tr>
<tr>
<td>20</td>
<td>Ignition Delay 100% Mixing - Run 41</td>
<td>37</td>
</tr>
<tr>
<td>21</td>
<td>Ignition Delay 40% Mixing - Run 41</td>
<td>39</td>
</tr>
<tr>
<td>22</td>
<td>Graphical Comparison of Analytical and Experimental Ignition Delay Results as Function of H₂ Temperature</td>
<td>41</td>
</tr>
<tr>
<td>23</td>
<td>Graphical Comparison of Analytical and Experimental Ignition Delay Results as Function of Equivalence Ratio</td>
<td>42</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Table</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Ignition Delay Experiments</td>
<td>23</td>
</tr>
</tbody>
</table>
I. SUMMARY

Theoretical and experimental studies have demonstrated the potential for aircraft capable of operating at hypersonic speeds within the atmosphere with an air breathing engine employing supersonic combustion. Duplication of true flight conditions in ground testing these propulsive systems requires a significant extension of existing facility capabilities and operating ranges. However, the nature of the flow at these extreme conditions does not completely simulate the actual flight conditions experienced by the Supersonic Combustion Ramjet (SCRAMJET) engine because the flowing gas stream is not in complete thermal and chemical equilibrium.

This research program is a theoretical and experimental investigation of the effect of nonequilibrium conditions upon the performance of combustors employing supersonic flows. Calculations and experiments are made regarding the effects on the ignition of hydrogen of the nonequilibrium species (free radicals, atoms, water vapor, etc.) obtained using vitiated air.

Results of this investigation show that the nonequilibrium free-radical content from a supersonic vitiated air source will cause early ignition of the hydrogen. An analysis of heated air expended from a high temperature source to test section conditions also indicates that there is sufficient free radical content in the incoming flow to cause early ignition. Water vapor, an inherent contaminant in the generation of vitiated air, was found to reduce the ignition delay period under the experimental conditions considered.
II. INTRODUCTION

Propulsion systems utilizing supersonic combustion have recently been recognized as having strong potential for vehicles operating in the hypersonic flight regime. As flight velocities are increased above Mach Number 6, the stagnation temperatures and pressures required for meaningful ground test duplication becomes increasingly difficult to achieve, particularly where large test sections and long experimental run times are required. A typical supersonic combustion ramjet engine (SCRAMJET) cruising at a Mach number of 6 at an altitude of 80,000 feet would encounter stagnation temperatures and pressures in the realm of 2500°R and 150 psia with air mass flows of 150 pounds per second. Dr. Antonio Ferri (1)* pointed out that the most urgent problem to be solved before a supersonic combustion engine could be perfected was the development of ground facilities for testing these engines. Within the present state-of-the-art of ground test facilities, it is just about impossible to test a complete engine under conditions duplicating all the parameters corresponding to high Mach number flight, particularly for a time duration sufficient to investigate the time-dependent facets of the combustion process. In the supersonic combustion problem as applied to the SCRAMJET, the chemical composition of the flowing medium, the mixing of the streams of fuel and air, the chemical reactions taking place, and the reaction rates involved are the important factors to be considered (2). The primary parameters requiring actual duplication in the test cell are the static temperature, static pressure, composition of the gases, residence time in the combustor, and the velocity relationship between the mixing streams. Duplication of the severe environments associated with low altitude high Mach number flight regimes using state-of-the-art ground test facilities, is possible at Mach numbers up to approximately 7, but it is difficult due to the size and operational cost of a clean air heater capable of delivering the necessary high temperature, pressure and quantity air mass flows. An alternate approach to this problem is to generate a fluid medium labelled "vitiated air", a synthetic mixture of gases having the same percentage oxygen content and molecular weight as true air. The most significant difference between vitiated air and true air is the contaminating constituents such as water vapor and carbon dioxide emanating from the chemical reaction of generation. These contaminating constituents are responsible for uncertainties in the results, as compared to true air, when employing vitiated air for high temperature combustion research. On the other hand, facilities employing vitiated air are able to operate at temperatures up to approximately 4500°F and pressures up to 2-3000 psi together with high flow rate capabilities.

This research program is a theoretical and experimental investigation of the effects of nonequilibrium conditions -- thermal and chemical -- upon the performance of combustors employing supersonic flows. Specifically, the effects of free radicals, atoms and third-body species such as water vapor (a contaminating constituent in vitiated air), upon the ignition process are determined. Calculations and experiments are made regarding the effects on the ignition of heated hydrogen of the nonequilibrium species obtained using vitiated air.

*Number in bracket indicates references in the Bibliography.
A. SCOPE OF PRESENT INVESTIGATION

In the supersonic burning of gaseous hydrogen with air, there is a finite distance required for the mixing and subsequent ignition and combustion of the two gas streams. It is a well-documented fact that the reaction rate of a chemical reaction is directly proportional to the temperature and pressure, with the effect of temperature the more prominent (1). Thus, with a high initial static temperature (T > 1200°K), reaction times are almost negligible and the mixing phenomenon of the two streams becomes the rate controlling mechanisms. At low static temperatures corresponding to low flight Mach numbers, the chemical reaction times are slow and the reaction rate determines the combustor length. The present investigation is in the latter regime where the chemical reaction times are assumed significantly longer than the corresponding mixing times.

In the ground testing of a SCRAMJET engine combustor, the working fluid must be expanded from an initially high stagnation temperature regime (T ≈ 2200°K) to the relatively low temperature regime at the combustor entrance. Local conditions at this station are usually a fairly high velocity (6000 ft/sec) stream at this relatively low static temperature (T ≈ 1000°K). During expansion process free radical concentrations formed at the upstream stagnation conditions are frozen due to the fact that the gas residence times are much shorter than the reaction times at local temperatures throughout the expansion nozzle. This phenomenon known as "recombination freezing" results in nonequilibrium free-radical concentrations in the incoming stream at the combustor inlet. Initial free-radical concentrations in the incoming free stream have been found in previous analytical investigations to have an influential effect on the ignition delay distance (3).

In the formulation of the analytical portion of this investigation, papers of several authors who investigated the ignition delay mechanism of the hydrogen-air reaction systems were reviewed. The results which were felt to be germane to the unique vitiated air-hydrogen reaction were incorporated into the analytical model. Of particular concern were the effects of water vapor (an inherent constituent in vitiated air), a substantial initial concentration of free radicals and relatively low static temperatures, on the combustion of heated gaseous hydrogen in a supersonic flowing stream. R. P. Rhodes (3) found that the initial free-radical concentration of the incoming stream reduced the ignition delay time by a factor of two for an order of magnitude increase in initial free-radical content. Vitiated air, inherently in its generation has a substantial concentration of free radicals as will be shown later. Snyder et al (4) found that H₂O reduced the ignition delay time an order of magnitude below that for dry air. Vitiated air has an appreciable amount of water vapor, \( \approx 19\% \) by weight.

R. S. Brokaw (5) considered the importance of including H₂O₂ and H₂O reactions in low temperature ignition delay studies. These species, which were not included in any of the other investigations, were found analytically to increase the ignition delay in the temperature regime of interest in this investigation.
III. ANALYTICAL PROGRAM

The complete combustion process is assumed to occur in two steps as illustrated on Figure 1, which describes schematically the ignition model. During the induction period free radicals are exponentially produced by: (1) the initial chain initiating steps, and (2) the chain branching mechanisms of the bimolecular reactions. Throughout this period the temperature remains essentially constant because certain reactions are slightly exothermic and others endothermic. Towards the end of the induction period, the free radicals begin to recombine exothermically and the temperature starts to rise exponentially. The period of rapid temperature increase is termed the reaction time. In the temperature regime of interest in this investigation, reaction times are much shorter than the induction or ignition delay period. Therefore, the overall effort is directed toward evaluating the effects of vitiation on the induction period. The ignition model assumption of constant temperature along with the simplification of constant pressure allows one to neglect the fluid-dynamic equations and to solve only the differential equations describing the reaction kinetics.

Most of the theoretical and experimental work concerning the combustion of hydrogen-air, is based on the assumption that complete thermal and chemical equilibrium exists at the combustor inlet. In the shock tube work of Schott and Kinsey (6) and the standing detonation wave work of Nichols (7), the combined streams are assumed in equilibrium immediately downstream of the shock front. With this type of experiment, the assumption of equilibrium starting conditions may be justified on the basis that the agreement between analysis and experiment is reasonably good. For certain cases of shock induced combustion, Rubins (8) has shown the possible existence of deviation from thermal equilibrium. In these cases, the different modes of energy storage are not in equilibrium thereby resulting in a translational temperature overshoot. By accounting for the overshoot in the reaction rate expression, he was still able to predict ignition delays for shock-induced combustion of hydrogen-air using the assumption of chemical equilibrium as a starting condition for the finite-rate calculation. Therefore, most ignition delay data for hydrogen-air can be described theoretically by assuming either, that the premixed gases are in chemical equilibrium just prior to ignition or that each stream exists at its corresponding equilibrium state prior to mixing. The present analytical investigation falls under the latter category.

The analytical effort employs a Finite Rate Reacting Gas (FRRG) computer program for generating the species concentration for the vitiated air. This program was developed by TRW Systems for the NASA (MSC). The program solves the equations for inviscid one-dimensional flow of a reacting gas mixture and calculates the nonequilibrium species concentration at specified area ratios in the gas generator nozzle, assuming finite rate or equilibrium chemistry. The specie concentration as a function of temperature ratio in the nozzle is presented as shown in Figure 2.

In order to calculate the ignition delay, the approach taken was to write the equations governing the rate of growth of free radicals and to solve these
INITIAL TEMP. OF REACTANTS

STEADY STATE COMBUSTION TEMPERATURE

INDUCTION TIME

REACTION TIME

MIXING PLANE

TEMPERATURE

TIME

Figure 1: Schematic of the Ignition Model
FIGURE 2 EQUILIBRIUM AND FINITE-RATE COMPOSITIONS FOR G.G. PRODUCTS
for species concentrations as a function of time using standard matrix techniques. The computer program used was a modified program supplied by the NASA Lewis Research Center. It is a simplification of a more general program developed by the General Applied Science Laboratory (GASL). The modification included a constant ignition temperature simplification and the inclusion of several hydrogen-oxygen reactions (H2O2, H2O previously mentioned) which were analytically found to be important to the ignition kinetics at temperature below 1200°K. Inclusion of these species in the induction analysis should result in a more favorable comparison of experimental and theoretical ignition delays at low ignition temperatures. This computer program is labelled the Simplified Ignition Lag (SIL) program.

Figure 3 is a schematic diagram for the analytical mechanism of the investigation. Fifteen reactions are included in the SIL program. The main limitation of this program is that it fails to include any NO or NO2 reactions which were experimentally found to catalytically reduce the ignition delay time. Since the mechanism which accelerates the reaction is not understood at this time, it was not possible to include any NO or NO2 reactions in a manner that would be meaningful. Results from the SIL program are species concentrations as a function of time. The ignition delay time is determined from the semi-log plot of the hydroxyl radical concentration against time. This delay period is characterized by the region of essentially constant exponential growth of free-radical concentration. The end of the ignition delay period is considered to be the time when the hydroxyl concentration deviates from constant exponential growth as shown in the Figure 4. This definition for the end of the delay period is recommended by Hersch (9) who investigated the effect on ignition delay times for different methods of identifying the termination of the delay period.

Before applying the SIL program to the analysis of ignition delay with vitiated air, it was first necessary to establish the validity of the program's results. Since most of the theoretical and analytical work on ignition delay is concerned with real air and hydrogen, the verification was made for these gases. Initially a base case was selected for making this comparison and is shown with the base conditions also in Figure 4. As a starting condition both streams are assumed to be in chemical equilibrium at the assigned temperature and pressure prior to mixing. The combined stream is no longer in chemical equilibrium. In the bottom right hand corner are delay calculations of three other investigators for the base case conditions. In the next three figures are shown the effects of temperature, equivalence ratio and water vapor on the delay time under base case conditions.

The effect of temperature on induction time is shown in Figure 5, in which hydroxyl radical concentrations as a function of time are presented at three separate temperature levels. The results show that a 100°K reduction in temperature over the base case just about doubles the ignition lag time. However, a further reduction of the same magnitude, results in a factor of six difference in lag time over the base case.

Figure 6, illustrates the effect of equivalence ratio on the ignition lag
FIGURE 3  ANALYTICAL MECHANISM FOR INVESTIGATION
BASIS:
(1) HYDROGEN-AIR
(2) $T = 1200^\circ K, P = 1.0$ ATM., $\phi = 1.0$
(3) EQUILIBRIUM INITIAL CONDITIONS

\[ \tau_{10} = 39 \mu \text{SEC} \]

**FIGURE 4** IGNITION DELAY FOR BASE CASE SPECIES

**APPROXIMATE DELAY CALCULATIONS**

<table>
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<tr>
<th>SOURCE</th>
<th>$\tau_{10}$</th>
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<tr>
<td>FERRI</td>
<td>24 $\mu$SEC</td>
</tr>
<tr>
<td>NICHOLLS</td>
<td>34 $\mu$SEC</td>
</tr>
<tr>
<td>RHODES</td>
<td>75 $\mu$SEC</td>
</tr>
</tbody>
</table>
FIGURE 5 EFFECT OF TEMPERATURE ON INDUCTION TIMES
Basis:

1. Hydrogen-air
2. $T = 1200^\circ \text{K}, P = 1.0 \text{ ATM.}$
3. Equil. initial conditions

Figure 6: Effect of equivalence ratio on delay times.
for the base case species concentration. Three values for the equivalence ratio are presented on this figure. The results show that equivalence ratio has a very small effect on induction times in the range \(0.2 < \phi < 1.0\). MONTCHILOFF (10) obtained similar results in a theoretical and experimental comparison of the effects of equivalence ratio on delay times. The analytical model of the ignition process assumes instantaneous mixing for the two streams. This assumption is justified on the basis that local mixture ratio variations during the actual mixing process have essentially no affect on induction time.

Production of vitiated air usually implies the presence of water vapor as a contaminant in the gas stream. The effect of water vapor on the ignition lag time is illustrated in Figure 7, which presents OH concentrations with time for the base case species and for a 10 percent water vapor additive. The 10 percent replaces an equal volume of air. Temperature, pressure, and equivalence ratio are fixed in this comparison. Water vapor has previously been shown to reduce the ignition time by a small amount as substantiated by Snyder et.al. (4)

Several authors have derived either analytical or empirical relations for predicting the ignition delay times for the hydrogen-air system. A comparison of their results with those obtained from the SIL program are presented in Figure 8. The correlations shown here are terminated at their respective temperature range of application. Note, that most of the correlations presented are plotted as straight lines. However, the SIL program results deviate considerably from straight line behavior especially at low temperatures. The reason is that the SIL program includes the effect of the \(\text{HO}_2\) specie on the ignition times. This specie has often been observed in hydrogen-oxygen flames as an intermediate product that disappears rapidly as the reaction progresses. BROKAW (5) is the only other investigator to include this specie and the point shown includes his postulated mechanism by which the \(\text{HO}_2\) specie influences the delay time.

A comparison of the SIL program with experimental data for ignition delay at low temperatures is shown in Figure 9. Above 1000°K the SIL program predicts delays close to those of MONTCHILOFF (10) and SCHOTT (6) who is indicated on the graph by the square symbols. The SIL also tends to follow the basic trends of Snyder's shock tube data (4) which is identified by the circles and triangles at the lower temperatures. All in all, the general agreement of the SIL program with experimental data tends to support the argument that ignition delays increase significantly at low temperatures due to the self-inhibition of the reactions by \(\text{HO}_2\) formations.

Examination of the ignition model employed in the SIL program has shown that most of the relevant parameters influencing induction times are predicted with a reasonable degree of accuracy. The assumption of starting conditions where both streams are in chemical equilibrium is an essential part of this effort because (1) the experimental data used for comparison with the program results were obtained under conditions where equilibrium species concentrations at the combustor inlet is a reasonable approximation and (2) most of the theoretical approaches for calculating ignition delays employ the assumption of chemical equilibrium as a starting condition. In the next section, the effect of non-equilibrium starting conditions is examined using the SIL program.
FIGURE 7 EFFECT OF WATER VAPOR ON INDUCTION TIMES

BASE CASE SPECIES

BASIS:
(1) $\text{H}_2\text{O}$ REPLACES EQUAL VOLUME OF AIR
(2) $T = 1200^\circ \text{K}, P = 1.0 \text{atm.}, \phi = 1.0$
(3) EQUIL. INITIAL CONDITIONS
FIGURE 8  IGNITION DELAY TIMES FOR DIFFERENT CORRELATIONS
FIGURE 9 COMPARISON OF ANALYTICAL AND EXPERIMENTAL RESULTS
In the analysis for vitiated air, three simplifying assumptions were incorporated into the existing model in order to obtain qualitative answers to the effect of vitiation on ignition lag. These are:

1. All viscous effects are neglected.
2. Instantaneous mixing of the mainstream gases and hydrogen occur at the point of injection.
3. Constant temperature and pressure are assumed to exist throughout the induction period.

Of these four assumptions only the second and the fourth are believed to be of significance. The SIL program results of vitiated air and H₂ are shown in Figure 10. Non-equilibrium compositions of the vitiated air at the entrance to the supersonic test section are computed utilizing the FRRG program previously described. In addition, the hydrogen stream at the test section entrance is assumed to have a free-radical composition corresponding to stagnation conditions in the H₂ manifold. The vitiated air and hydrogen streams are mixed at constant pressure and the resulting species concentration are input as initial conditions into the simplified ignition lag program. As shown, the results indicate that the delay period may be as short as 20 μsec. Under the prescribed experimental conditions of this investigation, ignition should occur at less than two inches downstream of the test section entrance. However, mixing times are neglected in this specific analysis, therefore the actual delay distance may be somewhat longer than predicted. This figure also includes the delay time calculated assuming chemical equilibrium for both the vitiated air and hydrogen prior to mixing. The delay time of 158 μsec corresponds to a delay distance of approximately 12 inches.

A comparison was also made to show the effects of employing heated air to perform the same type of experiment. The results of the calculation is as shown in Figure 11. For finite rate reactions, the 30 μsec corresponds to a delay of 2.5 inches whereas 150 μsec for equilibrium corresponds to 11 inches. The vitiated air and heated air analytical results indicate that the ignition delays are almost equal. For non-equilibrium inlet conditions, the free-radical content of the vitiated air and hydrogen is present in sufficient quantity to cause the ignition to be very rapid. Heated air contains no hydroxyl radical at the test section inlet but the non-equilibrium concentrations of atomic hydrogen and oxygen are sufficient to cause rapid buildup of hydroxyl radicals through the bimolecular reactions. Again, ignition is very rapid.
Figure 10 Ignition delay for vitiated air/hydrogen

Basis:
1. $T_0^{\text{AIR}} = 2220^\circ\text{K}$
2. Inlet Mach no. = 3.0
3. $P = 1.0$ atm., $t = 1.0$
Finite-rate composition, $T = 1000^\circ$K

Equilibrium initial composition, $T = 1030^\circ$K

RASIS:
(1) $(T_0)_{\text{AIR}} = 2430^\circ$K
(2) Inlet Mach No. = 3.0
(3) $P = 1.0$ ATM., $\phi = 1.0$

FIGURE 11 IGNITION DELAY FOR HEATED AIR/HYDROGEN
IV. EXPERIMENTAL PROGRAM

Sizing of the experimental apparatus was based on the criteria that the supersonic ignition test section should be large enough to minimize scale effects but small enough to reduce run costs. It was also desirable that the design conditions simulate a flight regime in which the SCRAMJET vehicle might be expected to operate. This regime was chosen so that ignition delay times would be relatively long so that they could be measured.

Design conditions selected for the gas generator were: chamber pressure = 600 psia, chamber temperature = 2200°K and flow rate = 9 lb/sec. The design operating conditions for the gas generator and supersonic ignition test section are shown schematically in Figure 12. These conditions simulate approximately a flight Mach number of 6.5 at an altitude of 80,000 ft., a regime that should be of interest for SCRAMJET application.

In these experiments, the gas generator pressure and hydrogen pressure were selected so that when they expanded to atmospheric pressure the design supersonic test section conditions were realized. The combined streams were permitted to mix and burn in an unconfined manner as a free jet. The absence of a confining wall made it possible to visually observe the flame front and determine the ignition delay distance. A solid wall near the reaction zone often catalyzes the combustion process by either surface reactions or through a temperature increase due to viscous dissipation. In addition, the flow pattern (recirculation) becomes more complex. By examining ignition in a free jet, the ignition delay effects should be theoretically predictable providing the reaction mechanism is selected properly.

1. Gas Generator The components of the gas generator include an injector, two chambers, a turbulence ring and a converging-diverging contoured (parallel flow) nozzle. All components of the gas generator except for the injector were fabricated out of oxygen free, electrolytic tough pitch copper. A preliminary heat transfer analysis indicated than an uncooled copper gas generator system subjected to the experimental chamber design conditions mentioned previously could theoretically operate for run times up to 14 sec. before failure. A maximum of 3.5 sec. steady state operation, which entails approximately 6.0 sec. of total gas generator run time was necessary for any of the prescribed experimental runs.

The two combustion chamber sections are separated by a turbulence ring. The purpose of the turbulence ring is to promote mixing in the hot gases in order to insure a relatively uniform exhaust stream.

The gas generator injector has three separate elements: The oxidizer inlet housing, fuel injection pintle and a porous metal stainless steel surface in the area exposed to the hot combustion gases. The diluent nitrogen is introduced into the combustion chambers through this porous metal surface and thus serves to cool the entire injector face. The fuel injection pintle serves to introduce the hydrazine into the combustion chamber and together with the nitrogen inlet housing forms an annulus for injecting the nitrogen tetroxide oxidizer. The pintle contains 10 hydrazine injection ports of 0.0156 in.
FIGURE 12 SCHEMATIC OF EXPERIMENTAL SET-UP
The gas generator nozzle was designed for uniform, parallel flow across the exit plane. Design of the exit contour was accomplished by utilizing a computer program which solves the two-dimensional, axisymmetric flow relations by the method of characteristics. The program assumes constant values for the ratio of specific heats and for the molecular weight of the mixture. This assumption yields a good approximation to the contour coordinates calculated for real gas conditions. The parallel flow nozzle has a throat diameter of 1.596 in. and an overall area ratio of 5.38. A photograph of the assembled gas generator apparatus is shown in Figure 13 with the hydrogen manifold removed.

2. Hydrogen Heater The hydrogen heater consists of a large stainless steel coil which is heated by convection and radiation from the combustion products of three propane burners. It has the capability of supplying continuously, 0.5 lb/sec of 900°K gaseous hydrogen at an operating pressure of 325 psig.

3. Supersonic Test Section The exit plane of the gas generator parallel flow nozzle together with the hydrogen manifold make up the supersonic test section.

The hydrogen manifold and nozzle exit section are mated so that an annulus is formed between them. The hydrogen gas is expanded to sonic conditions as flow through the annulus is choked. The lip separating H₂ and vitiated air flow is 0.050 in. thick and was purposely made as small as possible to minimize flow recirculation problems in this area, but still maintain some structural strength.

A. EXPERIMENTAL RESULTS

The experimental program was conducted essentially in two phases. In the first phase, the gas generator was developed to a level where it could be operated with a high degree of reliability. It was also necessary to demonstrate that the gas generator would operate repeatedly at high combustion efficiencies (n_c ≥ 93%) in order that the gas composition and state properties could be reasonably estimated by theoretical techniques. To attempt to achieve high combustion efficiency, the gas generator had a large characteristic length (L* ~ 120 in.) and a turbulence ring to promote mixing and complete combustion of the exhaust products. Subsequent to the achievement of satisfactory operation of the gas generator, the supersonic combustion ignition delay experiments were undertaken. This constituted the second phase of the experimental program. Results from each phase of this program are discussed separately in the following sections. A summary of representative performance results from the experimental effort, is presented in Table 1. The tabulation presents flow rates, chamber pressure and temperature when measured, hydrogen conditions, ignition delay data, a characteristic velocity (C*) determined from measured values, and a C* efficiency. The theoretical characteristic velocity employed to determine the C* efficiency was calculated from an equilibrium thermochemistry computer program.
**TABLE I**
IGNITION DELAY EXPERIMENTS

<table>
<thead>
<tr>
<th>Propellants: ( \text{N}_2\text{H}_4/\text{N}_2\text{O}_4/\text{N}_2 + \text{H}_2 )</th>
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<tr>
<td>Run No: 39 40 41 42 43 44 45</td>
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<tr>
<td>Oxidizer Flow Rate (lb/sec)</td>
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<td>( \text{N}_2 ) Flow Rate (lb/sec)</td>
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<td>Hydrogen Flow Rate (lb/sec)</td>
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<td>G.G. Chamber Conditions</td>
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<td>Chamber Pressure (psia)</td>
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<td>Measured Chamber Temp (°K)</td>
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<td>Total Flow Rate (lb/sec)</td>
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<td>( \text{C}^* ) (Measured) (ft/sec)</td>
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<td>( \text{C}^* ) Efficiency</td>
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<td>( \text{H}_2 ) Measured Manifold Temp (°K)</td>
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1. **Gas Generator Experiments** The objective of the initial series of runs was to balance the propellant flow rates and confirm the integrity of the system as a reliable apparatus. Line and injector resistances generated in the early series of experiments were used to set the propellant tank pressure for the desired propellant flow rates. 5.6 lbs/sec of liquid nitrogen tetroxide, 1.6 lbs/sec of liquid anhydrous hydrazine and 1.8 lbs/sec of gaseous diatomic nitrogen comprised the propellant combination utilized to generate 9 lbs/sec of vitiated air at 600 psia in the gas generator.

2. **Supersonic Combustion Experiments** The supersonic combustion experiments were directed towards determining the effect of vitiated air on the ignition delay times when burning hydrogen. Mixing and subsequent combustion of the heated hydrogen took place in the gases flowing as a free jet without any confining walls. Both streams were operated with a static pressure of 14.7 psia at the injection station in an attempt to minimize shock interactions in the exhaust gases due to the surrounding atmospheric conditions. By permitting the gases to mix and burn in an unconfined exhaust stream, the flame front could be visually observed and photographically recorded. A solid wall in the vicinity of the flame front can often have a catalytic effect on the ignition process. The absence of a confining duct should permit the experimentally determined flame front to be a function of the chemical kinetics of the reacting system of gases. There is also a possibility of induction of ambient air into the mixing region of the hydrogen and vitiated air which could reduce the effective temperature of the mixture (longer delay times). The problem stems from the entrainment effect that the hydrogen injection stream's velocity has on the quiescent ambient air. The hydrogen stream flowing at sonic velocity (~6000 ft/sec) entrains the cool ambient air and mixes it with the hydrogen. This mixture may be, in turn, mixed with the synthetic air. The overall result could be a decrease in the rate of free radical production due to the lower temperature environment and a lengthening of the ignition delay length. The fact that the ambient air does have an effect on an experimental apparatus of this nature is substantiated in Reference 11. It was obvious that the physical characteristics of the existing hydrogen injection system and combustion apparatus might generate this type of false ignition data, therefore, a short series of experiments were undertaken and the resulting effects were found to be apparent although minimal.

The vitiated air enters the supersonic test section at Mach 2.9 with an average computed static temperature of 1000°K and a static pressure of 14.7 psia. Injection conditions for the hydrogen are Mach 1.0 with a static temperature of 500°K to 800°K (depending on objectives of the run) and a static pressure of 14.7 psia. The relative velocity ratio for the two streams at injection conditions is 1.08 (hydrogen/vitiated-air). These experiments, however, had relatively large differences in static temperatures for the hydrogen (500°K - 800°K) and vitiated air system (1000°K) at test section conditions. The relative temperature difference for these two streams made it difficult to establish an "effective" mean temperature of the gases prior to ignition. Therefore, an analytical attempt to establish such a temperature was made in order to correlate analytical and experimental data. The results of this analytical investigation are discussed in the section entitled, "Discussion of Analytical and Experimental Results". Injection of both streams at essentially the same
temperature would eliminate this problem. However, if the hydrogen system was designed for a static temperature at injection equivalent to the vitiated air temperature, the heater would have to be designed to operate at 1200°K. The 867°K limit on the total temperature for the hydrogen was selected as a compromise to allow the use of relatively inexpensive materials in the construction of the hydrogen heater and downstream hardware.

After system integrity was confirmed, a subsequent series of experiments was initiated to demonstrate the phenomenon of hydrogen ignition in a supersonic (M = 2.9) vitiated air stream. This phenomenon was photographically recorded on black and white 16mm motion picture film taken at 550 frames per second. Upon reviewing the pictures, the film velocity (frames per second) was sufficient to slow down the ignition process so that an approximate ignition delay length could be measured. This measurement is based on the position where the flame front (first emitted light) initially appears. A photographic history of the ignition phenomenon is shown in Figure 14. These six consecutive frames show the instant just prior and subsequent to ignition. Frame 1 (upper left) depicts the transparent nature of the gas generator products (vitiated air). Flow is from left to right. In the next frame (lower left) hydrogen ignition is assumed to have occurred because of the presence of the visible luminous flame front. At this stage in the investigation no deliberate effort was made to accurately measure the actual ignition delay length, although it was possible to estimate this distance by comparison with known component dimensions in the photograph. A rough comparison of the annular hydrogen injector dimensions (10 in. in diameter) with the distance from it to the hydrogen flame front, measures an ignition delay length of approximately 10 in. In the three runs made during this short exploratory series, the same approximate distance was realized. As shown in frames 3, 4, 5, and 6, the hydrogen flame front propagates towards the point of injection and stabilizes approximately 2 in. downstream. This phenomenon occurred in all of the ignition delay investigations, but at slightly longer distances. This stabilized flame front can also be considered as a steady state ignition delay length, but it is felt that its position is highly influenced by the geometry of the hydrogen injector lip (recirculation effects).

In order to completely describe the vitiated air effects on ignition delay, as many properties of the gas generator and its products of combustion (vitiated air) as possible must be measured. High temperature 40 percent iridium, 60 percent rhodium/iridium thermocouples and water-cooled gas sampling probes were fabricated, for use in the measurement of the gas generator chamber temperature and the sampling of the vitiated air stream. The purpose of obtaining a vitiated air gas analysis was to attempt to determine the amount of nitric oxide (NO) and nitrogen dioxide (NO₂) in the vitiated air, and to compare the actual gas composition with that which was theoretically calculated by the Finite Rate Reacting Gas (FRRG) program. This specific computer program was discussed in the analytical section of this report. Theory and experiments indicate that nitric oxide and nitrogen dioxide have a catalyzing effect on the ignition process (4). If this is so, then knowing the mass fraction or existence of these two constituents in the vitiated air stream should help justify experimental results. Also, the presence of nitrogen dioxide in the gas is a measure of the incompleteness of the gas generator combustion process. Nitrogen
dioxide is not one of the products of the complete reaction of nitrogen tetroxide and hydrazine.

The analysis of the vitiated air gas sample collected indicated that no nitrogen dioxide was present in the air stream. This implies that the combustion process in the gas generator must have gone to completion and that the inefficiency as manifested in the less than 100 percent C* efficiency was due to heat transfer and boundary layer effects. A heat transfer analysis across the chamber walls was done and was found to account for at least a 2 percent decrease in C* efficiency.

The heat transfer rate through the gas generator chamber walls was measured simultaneously with the chamber temperature and ignition delay lengths. This was accomplished by mounting in the chamber wall two chromel alumel thermocouples at a depth of 0.813 and 1.563 in. respectively. The distance from the deeper thermocouple (1.563 in.) to the hot side of the chamber wall is 0.54 in. After each run the slope of the temperature increase for each thermocouple was compared for a quasi-steady temperature difference. The averaged difference was 88°K. Knowing the thermal conductivity of oxygen free electrolytic copper and the distance between the thermocouples, it was a relatively simple matter of applying Fourier's one-dimensional law of heat conduction for a hollow cylinder to get an approximate heat transfer flux rate of 1.54 BTU/in²-sec. This efflux of heat was then analytically accounted for in the measured C* combustion efficiency of the gas generator.

To measure the gas generator chamber temperature two 40 percent iridium, 60 percent rhodium/iridium thermocouples mounted inside the chamber cavity at depths of 0.37 and 1.125 in. respectively from the hot wall, were employed. The components of the thermocouple consist of a 0.062 in. inconel sheath with a magnesium oxide layer which insulates the two 0.010 in. diameter 40 percent iridium, 60 percent rhodium/iridium wires. A bead joining the two exposed thermocouple wires at a distance of 0.25 in. from the sheath material, formed the hot junction of the instrument. The thermocouples held up satisfactorily during the course of this investigation. Measurements were taken on all seven runs and were relatively consistent as indicated in Table 1. Temperature differences between the two thermocouples during the runs when averaged were approximately 76°K. The values of chamber temperature as listed in Table 1 are the uncorrected readings from the thermocouple positioned closest to the hot gas core (1.125 in.). A radiation and conduction thermocouple heat transfer error analysis was carried out according to the method outlined in Reference 12.

Results of the analysis indicated an average heat transfer error of approximately 5°K and 80°K for radiation and conduction respectively. The primary objectives for measuring the chamber temperature was to verify the accuracy of the temperatures computed by the Finite Reaction Rate Gas (FRRG) program. Temperatures (~ 1700°K) measured and corrected for thermocouple losses did not compare favorably with corrected theoretical temperatures (~ 2100°K) calculated by the FRRG program. Reasons for the discrepancy were not resolved since the resulting effects of this discrepancy on the investigation were felt to be minimal. However, due to the many unknowns associated
with high temperature thermocouple measurements, the temperature computed by the FRRG program were considered to be the more accurate of the two. Therefore, the corrected theoretical temperatures were subsequently employed to generate an input temperature which is used in the Simplified Ignition Lag (SIL) program to calculate the theoretical ignition delay distance. The theoretical ignition length is compared with that experimentally measured for correlation. This correlation is discussed in the section "Discussion of Analytical and Experimental Results." An extensive treatise on this specific portion of the investigation can be found in Reference 13.

The actual ignition delay length data were photographically recorded on Kodak 4X-Reversal black and white film using a Fastex 16mm camera at a film framing rate of 550 frames per second. A photograph of the apparatus for observing and measuring the ignition phenomenon is presented as Figure 15. The graduated measuring scale is mounted on the side of the test stand for determining the ignition delay length. Since the vitiated air/hydrogen mixture burns with a diffuse consistency, the dark background horizontal slot in the measuring scale emphasized the ignition distance and subsequent reaction phenomena. Each white stripe is an inch apart. The left side of the horizontal slot is in the exit plane of the hydrogen annular injector and thereby forms the datum point from which the exact ignition delay length is measured. The distance measured from this datum to the point of first light apparition is defined to be the ignition delay length. Figures 16 and 17 are typical photographic data results of the experimental runs. Each figure depicts 14 consecutive frames showing the instant before and after the onset of ignition. In Figure 16, the fourth frame down shows the point of ignition or a distance of 10 in. The inked-in line highlights the line of demarcation between the dark background of the horizontal slot in the measuring scale and the vitiated air hydrogen combustion flame front. The white elongated marks on the extreme outer edge of the film strip are the film velocity timing marks which are generated at a frequency of 100 per sec. The film velocity as shown is approximately 550 frames per second. Careful review of the succeeding frames subsequent to the point of ignition, reveals that the flame front propagates towards the plane of hydrogen injection to about 3 in. and then oscillates between 3 and 7 in. This effect is believed to be a recirculation phenomenon and a function of the nozzle/injector-lip geometry.

In Run 43 (Figure 17), the temperature of the heated hydrogen ($T_{H_2} = 590K$) was just slightly above the autoignition temperature. It was demonstrated in previous experimental runs that hydrogen injected at a temperature of 506°C and 521°C would not ignite in this apparatus. Nevertheless, (Figure 17) ignition was realized but with a relatively long ignition delay length. Concentrating on the third and fourth frames down and in the immediate area to the extreme right side of the measuring scale, the onset of ignition is indicated by the illumination of this section. However, the flame front does not sharply appear until frame number 6. This indicates that the point of ignition is somewhere near the extremity of the measuring scale. By considering the flame front propagation speed (approximately 2 in. per frame) and assuming it constant, the point of ignition is calculated to be approximately 19 in. In this particular run the flame front stabilized at a distance of 8 in., which indicates hydro-
$L_{ID(EXP)} = 10$ INCHES

$T_{H_2} = 710^\circ K$

$\phi = 1.07$

FIGURE 16  IGNITION DELAY - RUN 41
\( L_{ID(EXP)} = 19 \text{ INCHES} \)

\( T_{H_2} = 590^\circ K \)

\( \phi = 1.12 \)

**FIGURE 17**  IGNITION DELAY - RUN 43
gen temperature as well as geometry may have a profound influence on the point of stabilization. No attempt was made to resolve this uncertainty.

Photographic data results of the other four runs are similar in nature to these presented. A compilation of the ignition delay data as a function of hydrogen manifold temperature and equivalence ratio is presented graphically as Figures 18 and 19. A comparison of these results with the analytical findings is discussed in the following section.
Figure 18: Graph Ignition Delay Versus H₂ Temperature
FIGURE 19  GRAPH IGNITION DELAY VERSUS EQUIVALENCE RATIO
IV. DISCUSSION OF ANALYTICAL AND EXPERIMENTAL RESULTS

The ignition delay for a reacting gas mixture is determined primarily by the number of free radicals initially present. At no time during the ignition and subsequent combustion process does the mass fraction of free radicals become a large portion of the total gas mixture. Under conditions of relatively long ignition delays (low static temperatures), the ignition process can be accelerated significantly by small amounts of free radicals. An extensive analysis was undertaken to determine the effect of vitiation on the ignition delay characteristics of hydrogen. The analysis showed that the nonequilibrium free-radical content of the vitiated air system as it enters the supersonic combustion chamber would result in relatively short ignition delay times. The effect is not entirely a function of the vitiated air composition but was shown to be related to the nonequilibrium expansion process by which the gas is accelerated to test section conditions (recombination freezing). In that sense real air exhibits the similar effect on the ignition delay characteristics with hydrogen when the air is expanded to the supersonic test section Mach number from a high temperature source.

The nonequilibrium analysis for a test case typical of the experimental conditions resulted in a calculated ignition delay length of 1.5 in. for the vitiated air. This calculation assumes the hydrogen and vitiated air to be instantaneously and completely mixed at a constant mixture temperature of 1000°K and an equivalence ratio of unity. It also assumes nonequilibrium gas compositions for both streams prior to mixing. When equilibrium compositions were employed in these calculations, the ignition delay length was computed to be 11.5 in. This is approximately a factor of eight increase in distance over the nonequilibrium case. Therefore, should the experimentally observed ignition delay distance be significantly less than 11.5 in. for mixture temperatures of 1000°K or less, the effects of the nonequilibrium inlet conditions will have been demonstrated. The fact that ignition occurred in some cases at distances less than 11.5 in., does indicate the presence of nonequilibrium free radicals which have reduced the ignition delay phenomenon.

An additional theoretical analysis was undertaken to attempt to achieve better correlation between experimental and analytical program results. In the experimental program, the hydrogen and vitiated air enters the test section area at different static temperatures. This fact complicates the analysis of the delay time since a mean mixing temperature for the reacting gases must be defined before computing the ignition delay distance. The operational procedure for this specific analysis was executed as follows:

1. In an experimental ignition delay run note the hydrogen manifold temperature and flow rate, gas generator chamber pressure, propellant flow rates, and observed ignition delay distance.

2. Input the required gas generator properties and parameters into the Finite Rate Reacting Gas (FRRG) computer program and calculate velocity, specie concentration, static temperature and pressure at the gas generator nozzle exit plane, which also serves as the entrance to the supersonic test section area.
3. Input into the Simplified Ignition Lag (SIL) computer program the specie concentrations computed above, together with the additional hydrogen injected, the mixture mean temperature, pressure and equivalence ratio.

4. Determine from the computer-plotted output graph (OH radical) concentration versus time the point where the slope of constant OH radical growth starts to deviate from straight line behavior (end of ignition period).

5. Multiply the time indicated at this point of deviation by the velocity computed by the FRRG to determine the comparable ignition delay length.

In the initial attempt to compare the experimental and analytical results as outlined above, the mean temperature of the mixed stream (hydrogen and vitiated air) was determined by employing the gross over-simplification that the mean temperature can be obtained from the weighted heat constant of the combined streams. Since the flow rate of the vitiated air stream was measured to be 9.0 lb/sec in contrast to the flow rate of 0.08 lb/sec to 0.29 lb/sec of the hydrogen stream, the mean temperature of the mixture, assuming instantaneous and complete mixing, was for all practical purposes equal to that of the vitiated air stream (~980°K). A typical result of this preliminary effort is presented as Figure 20. A comparison of the analytical results with the experimentally measured ignition lengths were not satisfactory. Experimentally there was a factor of approximately 3 difference in the measured ignition delay distances for the different hydrogen inlet conditions. This discrepancy between the analytical and experimental results is believed to be entirely an effect of the over-simplified assumption of instantaneous and complete mixing of the two dissimilar streams.

A combined mixing and reaction kinetics analysis is beyond the scope of the present program, however, a simple rule of thumb used in free jet mixing studies states that mixing is usually complete in a distance of approximately 10 jet diameters from the injection plane. This distance would be about 45 in. in the existing system. The maximum ignition delay distance observed was 19 in.; therefore, it can be safely concluded that the onset of ignition occurs before the hydrogen gas has a chance to diffuse completely into the vitiated air core. Hence, the reaction zone of the mixture forms a conical sheath about the vitiated air stream.

In order to improve the correlation of the experimental and analytical data, a short analytical investigation of the effects of vitiated-air/hydrogen mixture temperature on ignition delay as calculated by the Simplified Ignition Lag (SIL) computer program was accomplished. Since it was shown previously that the hydrogen does not completely penetrate the vitiated air core before ignition, then, it is only logical to conclude that the mean mixture ignition temperature should be a direct function of penetration depth. The initial phase of this specific investigation employed the assumption that due to the limited penetration of the hydrogen into the air core, that ignition occurred at a mean temperature equal to the injection temperature of the hydrogen. The static temperature of the hydrogen subsequent to passage through the sonic annular injector is the value used in all of the analytical ignition delay efforts. Hydrogen temperatures as listed in the tables and graphs are mani-
$T_{H_2} = 710^\circ K$, $\phi = 1.07$, $T_{mix} = 976^\circ K$

VEL = 6265 ft/sec

$L_{ID(SIL)} = 4.09$ INCHES

$L_{ID(EXP)} = 10$ INCHES

$\tau_{ID} = 54 \mu sec$

FIGURE 20  IGNITION DELAY 100% MIXING - RUN 41
fold or total temperatures. In all cases considered, the OH radical concentration never increased. On the basis of this data it can be concluded that ignition never occurred. The experimental observation that ignition and sustained combustion did occur in Run 41 and other runs considered in this special analysis, invalidates the assumption of mixture temperatures equal to hydrogen injection temperatures. In order to approximate the depth of hydrogen penetration into the hot vitiated air core before ignition and, in turn, to be able to generate a realistic mean mixture temperature for analytical purposes, the GASL's "Finite Rate Supersonic Combustion Mixing Analysis" (14) computer program was employed. This program describes the turbulent mixing of axisymmetric hydrogen-air jets inside a duct. Even though the program was written for ducted flow, the initial portion of the output is applicable to the mixing and ignition processes. This is only true when the processes are confined to a region located a considerable distance upstream from the point where the reaction zone would theoretically contact the duct wall.

The approximation of a mean mixture temperature was obtained by using the calculated penetration depth at which the equivalence ratio output of unity occurred. This value was located at a grid point positioned 0.414 in. deep into the periphery of the vitiated air core at an axial distance of 1.2 in. from the plane of hydrogen injection. It was therefore concluded that in the distance from the hydrogen injection plane out to this station (1.2 in.) the hydrogen gas diffused into the vitiated air core to a depth of 0.414 in. The volume percentage of the vitiated air core penetrated by the hydrogen gas in this distance amounts to approximately 40 percent. Therefore, the mean temperature of the mixture was arrived at by using only 40 percent of the total vitiated air flow in a vitiated-air/hydrogen weighted mean temperature calculation. This effort reduced the mean mixture temperature to a more realistic value and resulted in a better correlation between analytical and experimental data. The static temperature of the vitiated air core at the hydrogen injection plane was calculated to be 850°K. This value was arrived at by subtracting the temperature drop (ΔT = 133°K) due to heat transfer effects in the combustion chamber and along the nozzle walls, from the theoretical static temperature (983°K) calculated by the FRRG program. In the heat transfer analysis, it was found that the chamber temperature decreased by 67°K for a quasi-static heat transfer rate through the walls of 1.55 BTU/in^2-sec. It was therefore assumed that the additional temperature decrease through the nozzle section should be at least equal to that lost in the combustion chamber. With a modified vitiated air static temperature of 850°K versus the original theoretical static temperature of 983°K, it is felt that the species concentrations as calculated by the FRRG program at the original temperature (983°K) would not vary excessively due to this temperature difference. Hence, the species concentration as calculated originally at 983°K was input into the SIL program along with the mean temperature generated by using the modified temperature (860°K). A typical result representative of this analysis is shown in Figure 21 which is manifested in the longer ignition delay time. It was also observed that the analytical and experimental ignition delay lengths agreed much better than in the previous comparison where it was assumed that the hydrogen and vitiated air streams mixed instantaneously and completely prior to ignition. This comparison would seem to indicate that the effect of different static temperatures for the hydrogen and vitiated air streams in
\[ T_{H_2} = 710^{\circ}K, \, \phi = 1.07, \, T_{mix} = 831^{\circ}K \]
\[ VEL = 6265 \, ft/sec \]
LID(SIL) = 7.9 INCHES
LID(EXP) = 10 INCHES

\[ \tau_{ID} = 105 \mu sec \]

FIGURE 21  IGNITION DELAY 40% MIXING - RUN 41
addition to mixing rates, have a profound effect on any analytical ignition delay model.

Figures 22 and 23 are graphical plots comparing the experimental and analytical data as a function of hydrogen manifold temperature and equivalence ratio. As can be observed in Figure 22, the differences in delay length diverges as the hydrogen temperatures decrease. This effect may be caused by the effect of temperature on the mixing or diffusion rate in the two streams. It is shown in Reference 15 that the mass diffusivity in gaseous mixtures does increase with temperature. Another noteworthy observation is the trend of decreasing ignition length with increasing hydrogen temperature as manifested in both the analytical and experimental data results. This fact tends to reconfirm the validity of the analytical model in that ignition delay length decreases with increasing temperatures. In Figure 23 the experimental data deviate from the analytical theory of relative independence of ignition delay distances on equivalence ratio. The observed fact that the experimental data points do straddle a straight line when drawn equidistant from each point, provides some encouragement. An explanation as to why there is such scatter cannot be given at this time. Analytically, the data points do indicate a relatively constant ignition delay distance irregardless of equivalence ratio variance.
FIGURE 22  GRAPHICAL COMPARISON OF ANALYTICAL AND EXPERIMENTAL IGNITION DELAY RESULTS AS FUNCTION OF H₂ TEMPERATURE
FIGURE 23  GRAPHICAL COMPARISON OF ANALYTICAL AND EXPERIMENTAL IGNITION DELAY RESULTS AS FUNCTION OF EQUIVALENCE RATIO
V. CONCLUSIONS

Detailed analytical and experimental measurements have been made in a mixing and reacting vitiated-air/hydrogen supersonic stream for the purpose of determining the effects of vitiation products (free radicals, third bodies, etc.) on the ignition delay phenomenon. On the basis of the experimental data and their subsequent correlation with the analytical model, it is concluded that:

1. The presence of water vapor in a combustor entrance flow decreases the ignition delay or induction time for the temperature regime investigated.

2. The analytical ignition delay model was not independent of the mixing process. Results predicted by the analytical model compared favorably with the experimental data when effects of mixing were considered. Therefore in practical systems, reaction and mixing are coupled and should be considered simultaneously.

3. Nonequilibrium free-radical content of the incoming stream accelerates the reaction process. This causes ignition to occur much sooner than would be expected if the incoming stream were near equilibrium as far as free-radical concentrations are concerned as occurs when atmospheric air is diffused to the supersonic combustor inlet conditions. This indicates that the use of vitiated air to simulate the ignition phenomenon for a SCRAMJET system will lead to data which differs from flight conditions. These conclusions apply in the low temperature supersonic regime where reaction times are rate controlling.

4. In the temperature regime investigated (~ 850°K), the experimentally measured content (4 percent) of nitric oxide (NO) in the vitiated air had little effect on the ignition delay length. The catalytic effect of NO on ignition was not observed.
VI. BIBLIOGRAPHY


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