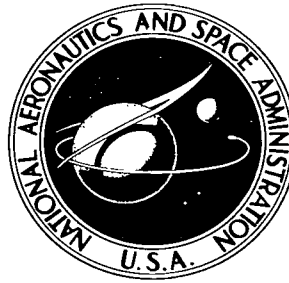


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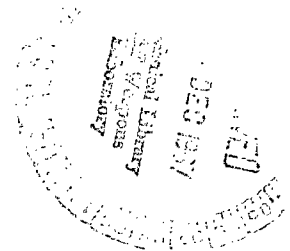
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HYDROGEN-OXYGEN CHEMICAL REACTION KINETICS IN ROCKET ENGINE COMBUSTION

by Martin Hersch
Lewis Research Center
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IN ROCKET ENGINE COMBUSTION**

By Martin Hersch

**Lewis Research Center
Cleveland, Ohio**

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HYDROGEN-OXYGEN CHEMICAL REACTION KINETICS IN ROCKET ENGINE COMBUSTION

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SUMMARY

Hydrogen-oxygen reaction times and concentration histories of chemical species during reaction were calculated for rocket combustor conditions. Calculations were made for oxidant-fuel weight ratios of 1 and 10 and initial reactant temperatures of 1200^o to 2500^o K at a chamber pressure of 20 atmospheres (2.0×10^6 N/sq m). The reaction time varied from about 0.01 second at 1200^o K to a few microseconds at 2500^o K. Calculations were made by using a numerical integration program and an analytical solution. The reaction mechanism used included five chain branching reactions and three recombination-type reactions.

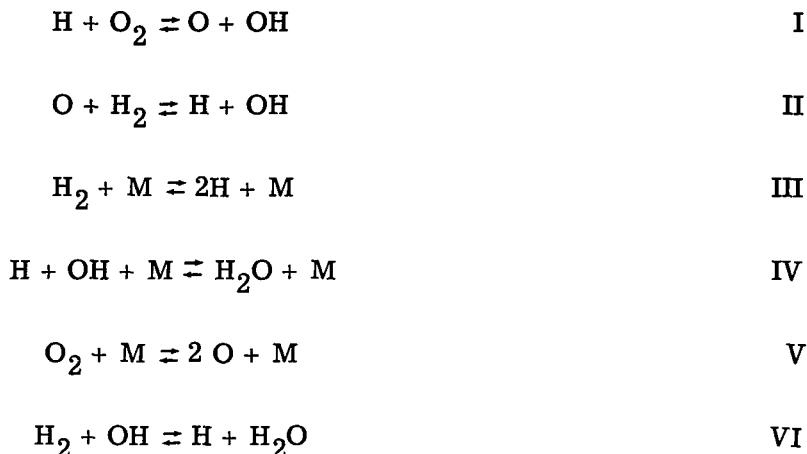
INTRODUCTION

Various steps in the rocket combustion process are atomization, vaporization, gas phase mixing, and chemical reaction. The slower steps control combustion efficiency. Some steps may also drive combustion instability, if their completion time approximates the wave time. Therefore, it is important to know the completion time for the various steps, the overall combustion time, and instability wave times. Combustion times for a typical rocket engine are reported in reference 1. High-frequency instability times are treated in reference 2.

Vaporization is often the slowest and, therefore, rate controlling step. Accordingly, it has been well studied (ref. 3). Gas phase mixing may also be a slow step and is considered in reference 4. Less information is available for chemical reaction rates in rocket combustion. Often, it is simply assumed that the reactions take place infinitely fast in comparison with other steps and thus may be neglected. This assumption may not always be valid. For example, if the slower steps, such as vaporization and mixing, are made highly efficient, then it would be important to know the chemical reaction times. Furthermore, combustion instabilities are known to cause large temperature and pressure gradients. Since chemical reaction rates are sensitive to temperature and pressure,

the assumption of infinitely fast reaction rates under these conditions may also be invalid.

Calculations of hydrogen-oxygen (H-O) kinetics for reaction limited ram-jet and rocket combustor conditions are presented in references 5 to 7. In these studies the following reaction mechanism was used:



where M is any third body.

In references 5 to 8 the initial temperature was varied from approximately 1000⁰ to 3000⁰ K at pressures of 1 to 65 atmospheres (1×10^5 to 6.5×10^6 N/sq m). Brokaw, using an analytical solution for reactions I, II, VI, VII, and VIII (ref. 9), shows that the following reactions are also important, particularly at low temperatures:



The importance of these two reactions increases rapidly as the initial temperature decreases. It would therefore be desirable to consider a mechanism which includes all eight reactions.

Calculations for this system of eight reactions are presented in this study. Numerical and analytical techniques are used. The effects of the initiation reaction $\text{H}_2 + \text{O}_2 \rightarrow 2 \text{OH}$ is also considered with the use of the analytical technique.

The numerical method considers reversible reactions, concentration changes of all species, and temperature change during reaction. The analytical method is restricted to constant H_2 and O_2 concentrations, nonreversible reactions, and constant temperature. Reaction times are presented as functions of initial temperature and oxidant-fuel weight ratios O/F at a constant pressure. Histories of atom and radical concentrations during

the reaction are also shown. Calculations were made for initial temperatures ranging from 1200⁰ to 2500⁰ K and O/F values of 1 and 10 at a chamber pressure of 20 atmospheres (2.0×10^6 N/sq m).

CALCULATIONS

Numerical Solution

The numerical integration program used for this study is that developed under NASA contract and described in reference 8. The assumptions made in this program are

- (1) The mixture is one of thermally perfect gases.
- (2) Flow is inviscid throughout.
- (3) Transport properties can be neglected.
- (4) Internal energies are in thermal equilibrium.
- (5) The law of mass action applies throughout.

Backward rates are calculated, with microscopic reversibility assumed, from the ratio of forward rate constant to equilibrium constant. Equations of mass, energy, and state are solved simultaneously with the differential equations for specie concentration changes due to chemical reactions.

Input data for the numerical program are temperature, pressure, species concentrations, forward rate constants, and equilibrium constants. Parameters for the forward rate constants are given in table I, and parameters for the equilibrium constants in table II. The equilibrium constants were calculated from the data of reference 10 and similar unpublished Lewis data.

The reactions, including those proposed by Brokaw (ref. 9), used for the numerical integration program are

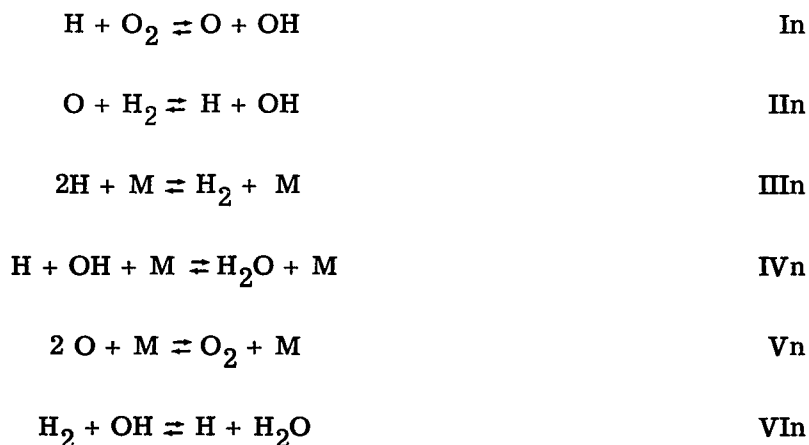


TABLE I. - FORWARD REACTION RATE PARAMETERS IN EXPRESSION^a

$$K = DT^n \exp(-E_A/RT)$$

Reaction number	D, cm ³ , mole, sec	n	Activation energy, E _A , cal/mole	Reference	Reaction
I	5.64×10 ¹³	0	15 100	5	H + O ₂ ⇌ O + OH
II	1.2×10 ¹³	0	9 200	5	O + H ₂ ⇌ H + OH
III	2.0×10 ¹⁸	-1.0	0	7	2H + M ⇌ H ₂ + M
IV	3.0×10 ¹⁹	-1.0	0	11	H + OH + M ⇌ H ₂ O + M
V	1.85×10 ¹⁷	-.5	0	5	2 O + M ⇌ O ₂ + M
VI	4.2×10 ¹²	.5	10 000	5	H ₂ + OH ⇌ H + H ₂ O
VII	5.4×10 ¹¹	0	24 000	9	HO ₂ + H ₂ ⇌ H + H ₂ O ₂
^b VIII	^c (5F _{H₂} + 1.75 F _{O₂}) 8.6×10 ¹⁴	0	-1 280	12	H + O ₂ + M ⇌ HO ₂ + M
IX	2.50×10 ¹²	0	39 000	13	H ₂ + O ₂ ⇌ 2 OH

^aWhere K is rate constant, D is preexponential constant, T is temperature in °K, n is temperature exponent, E_A is activation energy, and R is universal gas constant.

^bThird-body efficiencies obtained from unpublished Lewis data.

^cWhere F is mole fraction.

 TABLE II. - EQUILIBRIUM CONSTANT PARAMETERS IN EXPRESSION^a

$$K_c = D_c T^{E_c} \exp(F_c/T)$$

[Temperature range, 500° to 4600° K.]

Reaction number	D _c , cm, mole, sec	E _c	F _c , °K	Reaction
I	4.0748×10 ²	-0.4103	-8.6625×10 ³	H + O ₂ ⇌ O + OH
II	1.8764	.02212	-9.1499×10 ²	O + H ₂ ⇌ H + OH
III	.27658	-6.444×10 ⁻³	5.2421×10 ⁴	2H + M ⇌ H ₂ + M
IV	5.9727×10 ⁻³	.2779	6.0344×10 ⁴	H + OH + M ⇌ H ₂ O + M
V	1.2722×10 ⁻³	.4262	6.0169×10 ⁴	2 O + M ⇌ O ₂ + M
VI	2.1570×10 ⁻²	.2845	7.9234×10 ³	H ₂ + OH ⇌ H + H ₂ O
VII	.6614	-.0464	-7.6370×10 ³	HO ₂ + H ₂ ⇌ H + H ₂ O ₂
VIII	6.9973×10 ⁻²	.2799	2.3858×10 ⁴	H + O ₂ + M ⇌ HO ₂ + M

^aWhere K_c is equilibrium constant, and D_c, E_c, and F_c are equilibrium constant coefficients.



Analytical Solution

An analytical solution is obtained with isothermal conditions and constant reactant (H_2 and O_2) concentration for the system assumed:



Reactions IIIa and Va are written in the direction of dissociation because during initiation and induction radical formation is of concern. Reaction IVn is omitted from the analytical solution because it results in a term containing products of radicals, which results in a nonlinear equation. This reaction, however, is of little importance during the induction period. Reaction VIII is considered in both directions in the analytic solution.

The differential equations expressing the growth rate of intermediate species are (for the case of reaction VIII in the forward direction)

$$\frac{dC_O}{dt} = K_1 C_{O_2} C_H - K_2 C_{H_2} C_O + 2K_5 C_{O_2} C_M \quad (1a)$$

$$\begin{aligned} \frac{dC_H}{dt} = & -K_1 C_{O_2} C_H + K_2 C_{H_2} C_O + 2K_3 C_{H_2} C_M \\ & + K_7 C_{H_2} C_{HO_2} - K_8 C_{O_2} C_M C_H + K_6 C_{H_2} C_{OH} \end{aligned} \quad (1b)$$

$$\frac{dC_{HO_2}}{dt} = -K_7 C_{H_2} C_{HO_2} + K_8 C_{O_2} C_M C_H \quad (1c)$$

$$\frac{dC_{OH}}{dt} = K_1 C_{O_2} C_H + K_2 C_{H_2} C_O - K_6 C_{H_2} C_{OH} \quad (1d)$$

where the C is concentration, K is a reaction rate constant, and t is time.

The general solution for these differential equations is (ref. 9)

$$C_O = \sum_{i=1}^4 A_i e^{\lambda_i t} + a_o \quad (2a)$$

$$C_H = \sum_{i=1}^4 B_i e^{\lambda_i t} + b_H \quad (2b)$$

$$C_{HO_2} = \sum_{i=1}^4 C_i e^{\lambda_i t} + c_{HO_2} \quad (2c)$$

$$C_{OH} = \sum_{i=1}^4 D_i e^{\lambda_i t} + d_{OH} \quad (2d)$$

where A_i , B_i , C_i , and D_i are constants with dimensions of concentration, and λ is a constant with dimensions of second⁻¹.

The terms a_o , b_H , c_{HO_2} , and d_{OH} , which result from initiation reactions, are given in reference 9. For example,

$$a_o = \frac{K_3 C_{H_2} C_M + K_5 C_{O_2} C_M}{K_6 C_{H_2}} \quad (3)$$

Standard matrix techniques may then be used to determine the values of λ . Then, with the boundary condition that the radical concentrations are zero at time zero, the pre-exponential constants may be calculated. It then remains to calculate the growth of H_2O and H_2O_2 . The differential equations for these are

$$\frac{dC_{H_2O}}{dt} = K_6 C_{H_2} C_{OH} \quad (4a)$$

$$\frac{dC_{H_2O_2}}{dt} = K_7 C_{H_2} C_{HO_2} \quad (4b)$$

Since equations (4) contain terms which are now known, they may be integrated directly, with the boundary conditions of zero concentration at time zero. The detailed analytic solution was prepared and programmed for a high-speed digital computer by David A. Bittker of the Lewis Research Center.

RESULTS AND DISCUSSION

Results are presented for an oxidant-fuel weight ratio O/F of 1 and 10, a chamber pressure of 20 atmospheres (2.0×10^6 N/sq m), and initial temperatures ranging from 1200° to 2500° K.

The principal reason for studying chemical kinetics in rocket combustion is to determine the reaction time. This time is the time required to convert vaporized and mixed propellants to burned combustion products. For the hydrogen-oxygen system the reaction takes place in three distinct stages. The first stage is a very short initiation period during which small concentrations of atoms and radicals, such as H, O, and OH, are produced. This period is followed by the induction period, characterized by constant exponential growth of atoms and radicals, under nearly isothermal conditions. During induction a plot of the logarithm of concentration against time is a straight line.

Very late in the induction period atom and radical concentrations become large enough to cause self-heating due to three-body recombination reactions. The temperature then greatly increases, atom and radical growths depart from constant exponential

increase, and the system attains equilibrium. This final period may be referred to as the postinduction period.

The numerical and analytical methods are somewhat complementary. The numerical calculations cannot be started at the true beginning of the reaction, with the assumption that the reaction starts with zero atom and radical concentrations. On the other hand, the analytical program can be started for the conditions of zero atom and radical concentrations at zero time. The analytic method, however, gives no information which can be used to define the end of induction.

The OH concentration plotted against time during initiation and induction for various initial temperatures is shown in figure 1. Temperature change is also shown. The solid and dashed curves represent numerical and analytical results, respectively. The lower left ends of the curves show the very short period of initial formation of atoms and radicals. The curves then show the relatively long induction period characterized by constant exponential growth under isothermal conditions. The numerical results show that the induction period, as defined by constant exponential growth, continues, in some

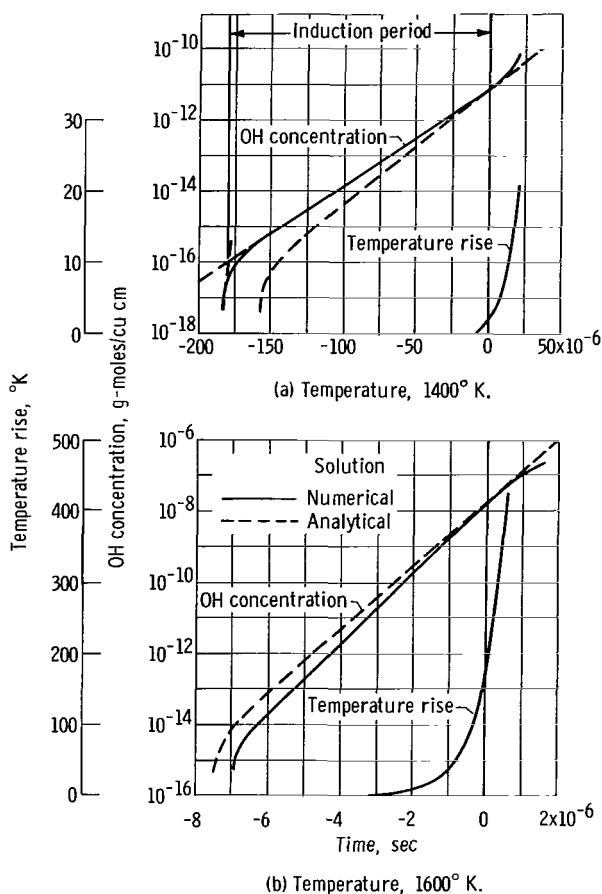


Figure 1. - OH growth and temperature rise.

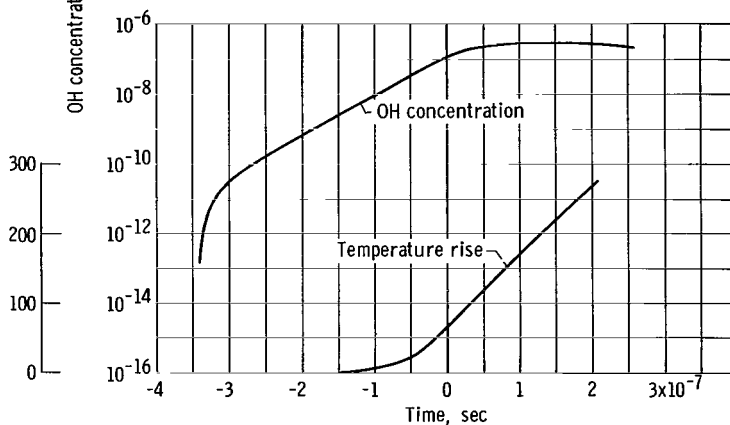
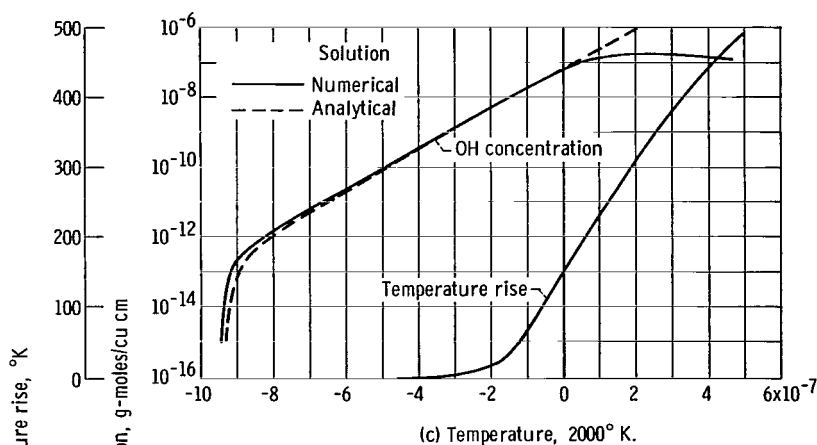


Figure 1. - Concluded.

cases, well into the region of temperature increase. In this study the induction period is considered ended when the atom and radical concentrations deviate from constant exponential growth. This deviation can be determined, in this study, by the numerical method only. Although the analytical assumptions are violated when the system is nonisothermal, the limit to which the analytical results can be continued is the point at which the concentration curves deviate from a straight line, as shown in figure 1. This point has therefore been selected as the end of induction and is plotted at zero time.

Numerical computation times increased drastically with decreasing temperature. Therefore, at 1200° K numerical results were obtained only near the end of induction and thus are not shown in figure 1.

The OH concentration at the start and end of induction are shown in figure 2. The concentration at the start of induction was determined by the intersection of the extrapolated slopes of the initiation and induction portions of the curves, such as shown in figure 1(a). The induction period is often considered ended when the OH concentration reaches the threshold of experimental detectibility, 10^{-9} gram-mole per cubic centi-

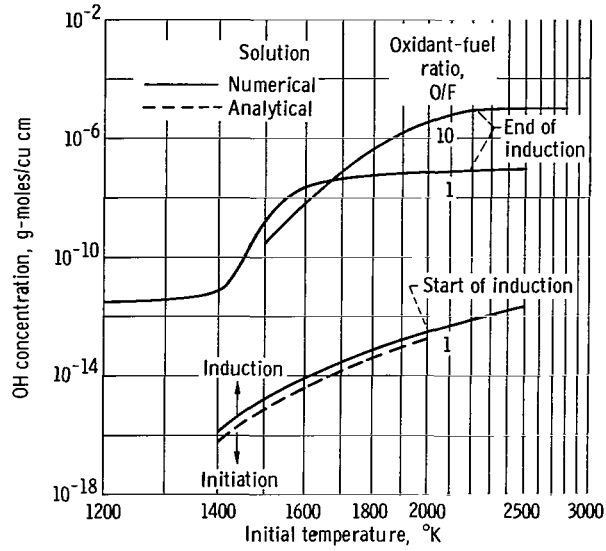


Figure 2. - OH concentration at start and end of induction.

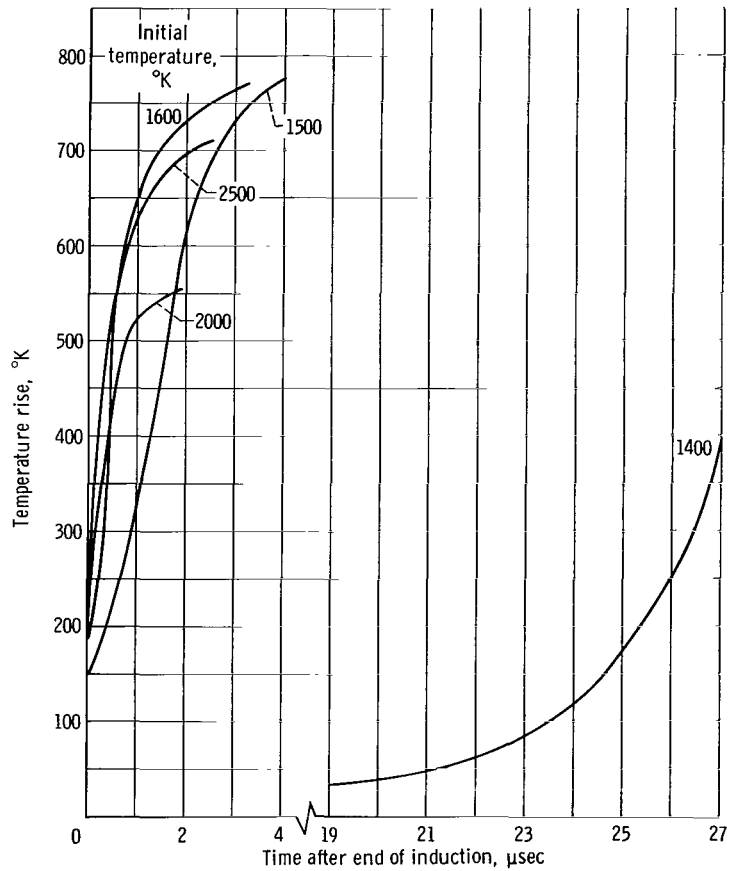


Figure 3. - Temperature rise during postinduction period (numerical solution).

meter (ref. 9). This criterion appears to be suitable only for an initial temperature near 1500° K. The induction time may now be determined by using these limits of OH concentration shown in figure 2.

Since in this study the total reaction time, rather than only induction time, is important, the postinduction time must also be investigated. Some idea of the postinduction time may be obtained from the curves of figure 3, which shows the temperature rise for various initial temperatures as a function of time. These results show that for the temperature range of about 1500° to 2500° K the postinduction time is on the order of a few microseconds. Though only partial results are shown for 1400° K, apparently at this temperature, the postinduction time may be on the order of perhaps 30 microseconds.

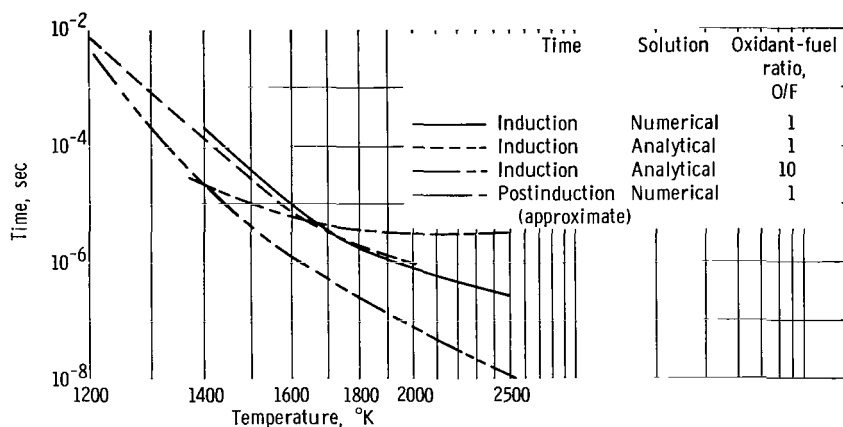


Figure 4. - Effect of initial temperature on induction and postinduction time.

The induction and approximate postinduction times are shown in figure 4. Numerical and analytical methods show nearly identical results for the induction time. For an initial temperature higher than about 1700° K, the induction time is small in comparison with the postinduction time and therefore may be neglected. At lower temperatures the postinduction time, though, becomes increasingly small in comparison with the induction time. The induction time increases rapidly with decreasing temperature for temperatures below 1600° K. In this region the induction time increases exponentially with the reciprocal of initial temperature.

The effect of O/F on induction time was investigated by determining the OH concentration at the end of induction for an O/F of 10 by using the numerical program. Calculations at an O/F of 10 were made for temperatures between 1500° and 2500° K. Detailed calculations at this O/F for the entire induction period or for lower temperatures were not made because of the long computational times. The OH concentration at the end of induction for an O/F of 10 is shown in figure 2. At high temperatures the OH at the end of induction at an O/F of 10 is about 10 times that at an O/F of 1. At

temperatures lower than 1700°K the OH concentration at the end of induction is approximately the same for both O/F conditions.

The induction time based on these numerically calculated OH concentrations at the end of induction for an O/F of 10 is shown in figure 4. The curve was extended to 1200°K by assuming that the OH concentration at the end of induction for an O/F of 10 was the same as for an O/F of 1. These results indicate that at higher temperatures the induction time decreases when the O/F is increased. At lower temperatures the effect of O/F on induction time appears to be small.

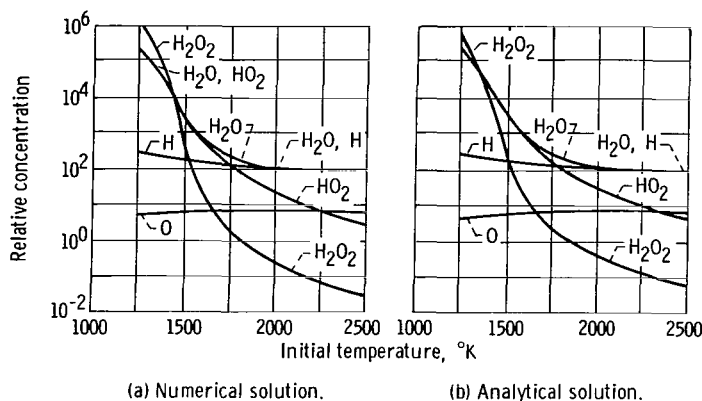


Figure 5. - Relative product species during induction period (normalized to OH concentration).

Some idea of the composition during induction is given by figure 5. During initiation relative concentrations of the atom and radical species shift with time. During induction, however, the relative concentrations remain nearly constant with time. Relative concentrations of O, H, HO_2 , H_2O , and H_2O_2 are shown, relative to OH concentration, during induction as a function of temperature. Almost perfect agreement is obtained between the numerical and analytical results. The relative concentrations of O and H appear nearly constant during induction over most of the temperature range. Concentrations of HO_2 , H_2O , and H_2O_2 are much greater than OH at low temperatures. At higher temperatures the concentrations of O, H, HO_2 , and H_2O are one or two orders of magnitude greater than OH. At 2500°K the H_2O_2 concentration during induction is nearly two orders of magnitude less than that of OH.

All numerical results presented thus far were obtained by starting the computations with initial concentrations of atoms and radicals determined analytically, because the numerical program cannot be started with zero atom and radical concentrations. The effect of starting numerical calculations with arbitrary atom and radical concentrations is shown in figure 6. The dashed lines are concentration histories when the starting concentrations were calculated analytically. The solid curves are concentration histories for arbitrary initial concentrations. These results show that if the initial

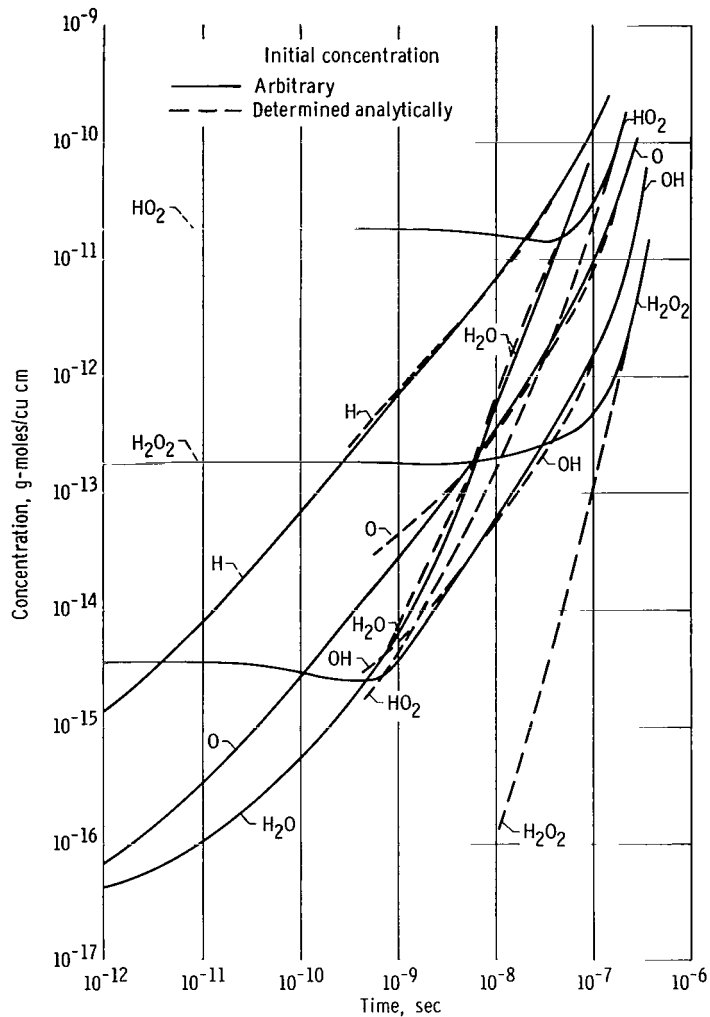
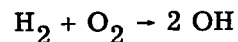


Figure 6. - Comparison of concentration histories for arbitrary with analytically determined initial concentrations. Temperature, 2000° K; pressure, 20 atmospheres (2.0×10^6 N/sq m); oxidant-fuel ratio, 1.

concentration of a species is too large, the concentration will remain essentially constant until the correct relative concentration is reached. It will then increase as it would have, had its initial concentration been correct.

It is possible to include additional initiation reactions in the mechanism. The most important of these reactions is perhaps the reaction:



The effect of this reaction on the time to reach a given OH concentration is shown in figure 7. Including this reaction decreases the reaction time by about two-thirds. At higher temperatures, where the postinduction time is controlling, this shortening of the

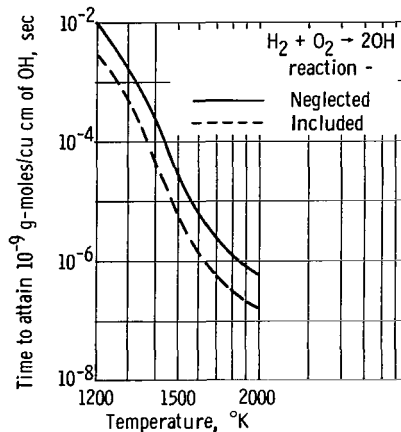


Figure 7. - Effect of reaction $H_2 + O_2 \rightarrow 2OH$ on time to attain arbitrary OH concentration (analytical calculations). Oxidant-fuel ratio, 1.

induction time would have no effect on the overall reaction time. This reaction might be of more importance at lower temperatures. It can be shown, however, that for rocket combustion studies this is not a significant effect.

SIGNIFICANCE OF CHEMICAL REACTION

TIMES IN ROCKET COMBUSTION

Liquid-propellant rocket-engine combustion involves many steps, atomization, vaporization, gas phase mixing, and chemical reaction. The importance of any steps in the combustion process may be related to its speed or completion time. The slower steps will be more important than the rapid ones. The combustion dead time, an indication of the time required for the overall process, is reported in reference 2 for a typical H_2-O_2 rocket engine. It varied from 0.25 to 2.25 milliseconds for a wide range of operating conditions. High-frequency instability must also be considered. Reference 1 shows that wave times for high-frequency instability range from about 0.1 to 1 millisecond. Thus, the time range of interest in liquid rocket combustion ranges from 0.1 to several milliseconds. Therefore, if chemical reaction times become greater than about 0.1 millisecond, they must be considered in studying rocket combustion.

This study shows that chemical reaction time increases very rapidly with decreasing reactant temperatures if the temperature is below about 1700° K. At 1600° K the reaction time is about 10 microseconds, which may be considered infinitely fast in comparison with other steps in rocket combustion. At about 1400° to 1500° K the reaction time increases to about 10⁻⁴ second and thus enters the region of interest in rocket

combustion. Above 1700° K the time decreases to several microseconds, and is fairly insensitive to temperature.

It is beyond the scope of this study to investigate the heating of unreacted propellants in the combustion chamber. Nor can it be determined here what the gas temperature is at the start of reaction. Presumably, though, the propellants in actual rocket combustors are substantially heated soon after injection by convection, radiation, and recirculation. The results of this study indicate that if the unreacted propellants are rapidly heated to about 1400° K, then chemical kinetics can be neglected as a rate controlling mechanism in steady-state combustion for a chamber pressure of 20 atmospheres (2.0×10^6 N/sq m).

Combustion instabilities, however, are known to cause large temperature and pressure gradients. If the instability wave reduces the temperature to below about 1500° K, then reaction times would greatly increase and become approximately equal to the wave period. Under these conditions the reaction kinetics could become an instability driving force.

SUMMARY OF RESULTS

Hydrogen-oxygen reaction times and concentration histories were calculated for rocket combustor conditions. Calculations were made for oxidant-fuel weight ratios of 1 and 10 and initial reactant temperatures ranging from 1200° to 2500° K at a chamber pressure of 20 atmospheres (2.0×10^6 N/sq m). A numerical integration program and a programmed analytical solution were used. The following principal results were obtained:

1. Hydrogen-oxygen reaction times varied inversely with initial reactant temperature, ranging from a few microseconds at 2500° K to approximately 0.01 second at 1200° K. Since processes requiring times less than milliseconds can be neglected in determining performance efficiencies or combustion characteristics this study indicates that chemical reactions can be neglected if the injected propellants are heated above approximately 1400° K.

2. When the initial propellant temperature was higher than about 1700° K, the reaction time was primarily the postinduction or heat-release period. At lower temperatures the reaction time was the isothermal induction time.

3. Induction time decreased with increasing oxidant-fuel mixture ratio. This effect of the oxidant-fuel mixture ratio on induction time increased with increasing temperature.

4. The analytical method presented permitted very rapid and accurate calculations of induction times and concentration histories for the hydrogen-oxygen system. However, in order to calculate induction times additional information must be available to define the end of the induction period. The numerical results presented provided this information.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, August 1, 1967,
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—NATIONAL AERONAUTICS AND SPACE ACT OF 1958

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