Shock-Induced Combustion with Oblique Shocks: Comparison of Experiment and Kinetic Calculations

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Theoretical analyses made recently have shown the advantages of supersonic combustion for a hypersonic ramjet. The present work deals with experimental investigations of shock-induced H_2-air combustion in the constant pressure region aft of an oblique shock and with kinetic calculations for the H_2-air reaction. The concept of shock-induced combustion is defined and compared with the usual conditions where detonations are observed, and it is concluded that detonations are a special case of shock-induced combustion. Quenched gaseous components for the early parts of the chemical reaction were analyzed to determine hydrogen molecule reaction rate, and a comparison was made of the experimental H_2 reaction rate, with the current chemical kinetic computations. One-dimensional flow was assumed for the analysis. Agreement with one kinetic calculation for the H_2-air reaction at constant pressure was within the experimental error, indicating the possible use of kinetics to predict hypersonic ramjet combustion performance when shock-induced combustion is used.

Nomenclature

C_p' = specific heat at constant pressure neglecting vibrational energy
E_{vib} = vibrational energy for a diatomic molecule
$E.R.$ = equivalence ratio = fuel concentration/stoichiometric fuel concentration
p = static pressure
R = universal gas constant
T = static temperature
T_0 = static temperature at reaction time = 0
t = relaxation time
$X_{1,0}$ = mole fraction of component 1 at time = 0, etc.
θ = characteristic temperature for vibration = $h\nu/k$, where h = Boltzmann constant, k = Planck constant, and ν = frequency of vibration
τ = reaction time
τ_d = "ignition delay" time

Subscripts

0 = condition at time = 0, or reaction starting time
1,2 = condition 1,2, etc.
vib = pertaining to the vibrational energy mode

1. Introduction

THE hypersonic ramjet, flying at altitudes near 200,000 ft, has been the subject of many investigations.[1–6] Feasibility can be shown on paper, but experimental information for realistic flight evaluation is far from complete. The purpose of this exposition is to describe experimental work in shock-induced combustion, to show the relation between experimental results and kinetic calculations, and to interpret the results in terms of possible use of the phenomenon for hypersonic air-breathing propulsion.

Currently, two concepts of supersonic combustion exist: 1) injection of gaseous fuel into a supersonic flow where the air and fuel are at a reacting temperature, or "diffusional" burning,[7] and 2) employing shock waves to increase the temperature of premixed fuel and air to a point where chemical reaction will start, or "shock-induced" combustion.[8, 9] The present work at the Arnold Center is concerned primarily with shock-induced combustion.

Shock-induced combustion differs from "detonations" in that the chemical reaction behind the shock does not necessarily affect the shock. The usual concept of detonations is exemplified by those observed in constant area tubes, where the steady-state condition is known as the Chapman-Jouguet wave. Shock-induced combustion implies that the fuel and air mixture is heated to a reacting temperature by shock compression, and, thus, the term can apply to all conditions where shock is followed by chemical heat release. Theoretically, the chemical reaction could proceed in an environment at constant static pressure, where, since pressure behind the shock wave will not be affected, it can be inferred that the shock wave will not be displaced. Other boundary conditions may be imposed on the reaction, such as constant Mach number or static temperature.

Ignition temperature may be generated by a shock wave; consequently, it follows that geometrical contour downstream of the shock is an intimate function of both the rate of the exothermic reactions and the imposed boundary conditions. Conversely, the exothermic reactions require a finite time and will generate pressure and velocity fields depending on the geometry of the walls. Through this reasoning, one is led to the conclusion that the constant area detonation, or Chapman-Jouguet wave, is a special case of shock-induced combustion.

Several investigators have been concerned with the chemical kinetics of the H_2-air reaction.[10–13] The kinetic models for the calculations made by Libby, et al.,[10] and Momtchiloff, et al.,[12] may be interpreted to behave as if they were separate streams of hydrogen and air, at a reacting temperature, that are suddenly mixed, and then start to react. This is very nearly what happens experimentally when premixed fuel and air is suddenly raised to a reacting temperature upon passing

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through an oblique shock wave. Because of the similarities between these experimental and kinetic models, it was found that they can be correlated if due consideration is given to their obvious differences.

2. Experimental Program

2.1 Tunnel

The experiments were carried out in a water-cooled, Mach number 3 tunnel, which has been fully described in Refs. 8 and 9. Air was preheated to 1500°R with an indirect-fired heat exchanger, and further heated to a maximum of 3800°R with a hydrogen burning preheater (Fig. 1). The vitiated air entered the tunnel throat, expanded into the supersonic section, mixed with injected fuel, and then passed through an oblique shock wave formed by a 28° wedge.

Previous work included experiments with normal shocks, generated at the intersection of two oblique shocks (Fig. 2a, Ref. 9). The present effort is concerned with oblique shocks generated by a single wedge (Fig. 2b, Ref. 8). The wedge angle was chosen so as to create a temperature rise that would be favorable to the reaction chemistry. Fuel was injected from the trailing edge of a thin, 12° half-angle, double-wedge strut (Figs. 1 and 2). Fuel temperature was estimated to be between 600° and 800°R.

Typical schlieren and emission photographs of the shock pattern, and shock with reaction mixture, are shown in Figs. 3a and 3b.

2.2 Gas Sampling

The total pressure probe was also used for gas sampling. Gas entered a small orifice, was cooled by sudden expansion to about $\frac{1}{6}$ of its original pressure, and then further cooled by contact with the cooled walls (Fig. 4). An analysis of the quenching process indicates that 1) the gas passes through the space between the normal shock and the probe tip in less than one μsec, and that 2) the combination of sudden expansion and wall cooling renders the gas substantially quenched in an additional one to three μsec. Experimental results exhibited evidence that quenching of the chemical reaction did, in fact, occur within a time interval approaching that predicted by this analysis. Gases were analyzed in continuous flow.

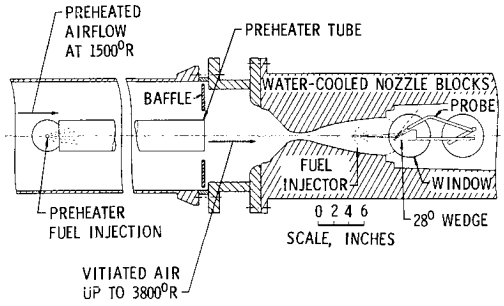

Fig. 1 Supersonic combustion tunnel and oblique shock experiment.

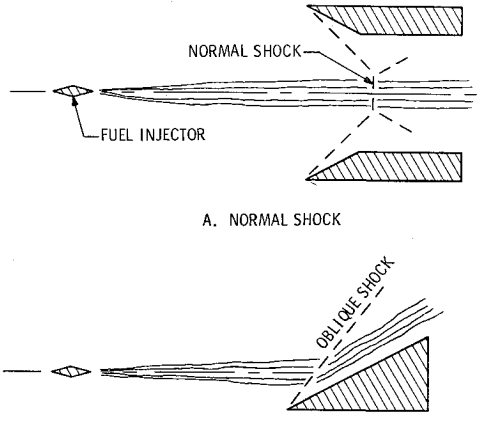

Fig. 2 Configurations for supersonic combustion experiments.

2.3 Calculation of Reaction Time

Reaction time was calculated by measuring the distance from the oblique shock wave to the probe and calculating velocity in the region downstream of the oblique shock;

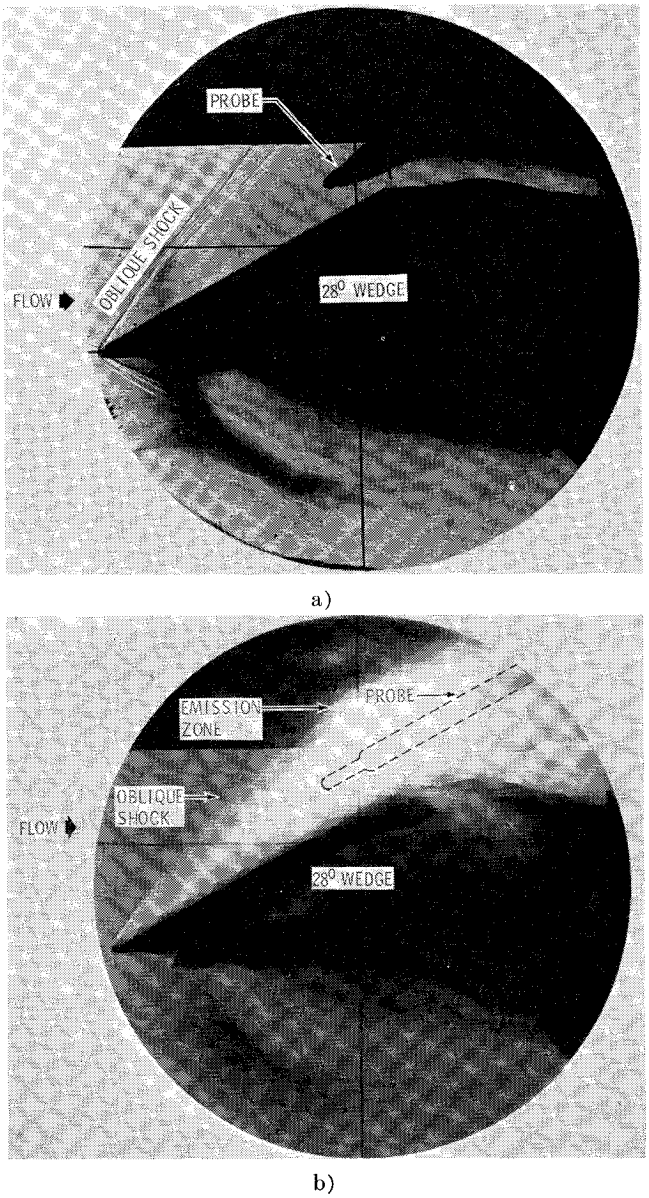

Fig. 3 Shock-induced combustion aft of an oblique shock in a Mach number 3 stream; a) schlieren photograph of the flow without combustion, and b) combined schlieren and emission photo with combustion.

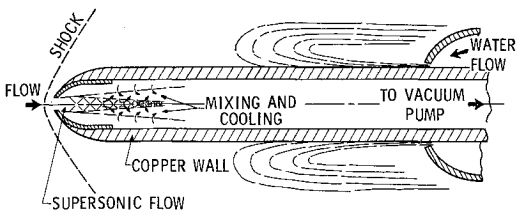

Fig. 4 Schematic of gas sampling probe.

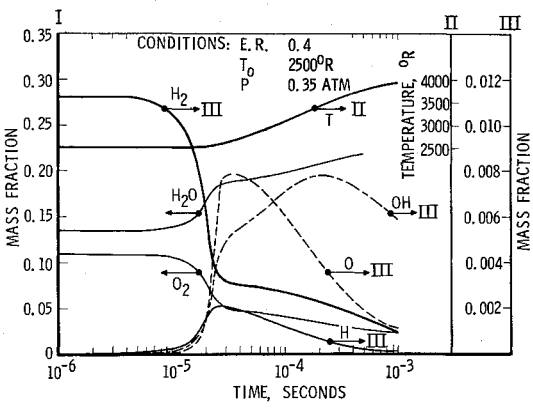

Fig. 5 Constant pressure reaction history for the H_2-air reaction simulating a typical experiment.

thus, time was determined from the shock to the probe. Mach number and the speed of sound were determined from 1) gas composition at the shock wave, or time "zero," 2) wedge surface pressure, and 3) temperature computed from the composition of preheater vitiated air and diffused, partially reacted fuel. Some error may be introduced into this calculation because of continued diffusion of the reacting fuel after time "zero," or because of expansion of the gases if significant quantities of heat are produced; however, this error is thought to be quite small.

3. Results and Discussion

3.1 Application of Kinetic Model to the Experiment

The conversion to O, OH, and H free radicals represents the initiating reactions of hydrogen and air.[10–13] It has been suggested by Schott and Kinsey[14] and Duff[15] that the process of the H_2-air reaction depends on the rates of the following reactions:

$$H_2 + OH = H_2O + H \quad (1)$$

$$H_2 + O = OH + H \quad (2)$$

$$O_2 + H = OH + O \quad (3)$$

$$O + H_2O = 2OH \quad (4)$$

$$H_2O + M = H + OH + M \quad (5)$$

$$H_2 + M = 2H + M \quad (6)$$

$$OH + M = O + H + M \quad (7)$$

$$O_2 + M = 2O + M \quad (8)$$

where M is any third body.

Machine computations which have been developed for analyzing the reactions of hydrogen and air have been attributed to the aforementioned authors.[10–13] A constant pressure reaction history for hydrogen and air is shown in Fig. 5 for conditions characteristic of the Arnold Center experiments, and was calculated using the computer program developed by Libby, Pergament, and Bloom.[10]

The model for the constant pressure reaction most nearly fits the experimental conditions because the region downstream of an oblique shock is both theoretically, for small quantities of heat release, and experimentally, one of constant static pressure. However, there are significant differences between model and experiment which must be accounted for in order to make a valid comparison. The model of Ref. 10 was interpreted to include two flowing streams of hydrogen and air, both in chemical equilibrium at a preset temperature, instantaneously mixed, and then proceeding to react. In the experiment, fuel is injected into preheated, vitiated air, which flows at a static temperature *less* than ignition temperature, and the mixture is then passed through a shock wave where the temperature is suddenly raised to a value somewhat higher than necessary for chemical reaction initiation (Fig. 6).

Four areas of possible divergences between the application of this model and the shock-induced combustion experiment are discussed as follows:

1) Effect of free radicals: Free radical concentrations were calculated for the experiment by assuming that the gas composition and temperature at the preheater discharge were in an equilibrium state, that the gas flowed to the throat of the nozzle at equilibrium, and that frozen flow existed from the tunnel throat to the test section (Fig. 1). These computed free radical concentrations were used to establish the initial conditions in the computer program at time "zero."

2) Effect of premixed shock heating vs mixing of preheated gas: It was assumed that mixed fuel and air, suddenly heated, reacts chemically in an identical manner as heated, un-mixed fuel and air, which are suddenly mixed. The mixed fuel and air, suddenly heated, is an experimentally realizable condition, whereas the instantaneously mixed reaction is not.

3) Shock wave translational and rotational temperature overshoot: The effective static temperature overshoot, caused when vibrational energy is slow to reach equilibrium as the gas passes through the shock wave, and the time associated with this lag were calculated for a typical test condition. The overshoot was assumed to affect gas temperature only at the shock wave and for a short distance downstream, and not to affect free radical composition entering the shock.

Vibrational energy of a gas may be calculated from the relationship[16]

$$E_{vib} = RT[\theta/T]/[\exp(\theta/T) - 1] \quad (9)$$

where θ (characteristic temperature for vibration) $= h\nu/k$. For oxygen, $\theta = 6000°R$, but for mixtures at lower temperatures, the gas behaves as if the effect of nitrogen is negligible.

At equilibrium, the difference in vibrational energy between two states is then,

$$E_{vib2} - E_{vib1} = R\left[T_2\left(\frac{\theta/T_2}{\exp(\theta/T_2) - 1}\right) - T_1\left(\frac{\theta/T_1}{\exp(\theta/T_1) - 1}\right)\right] \quad (10)$$

For the case where vibrational energy equilibrium is slow

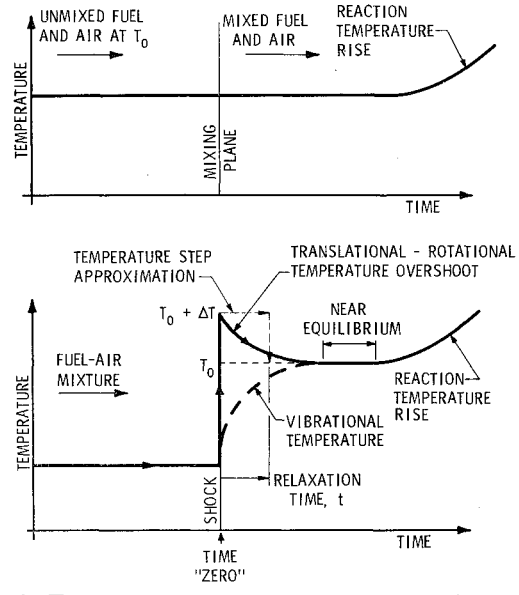

Fig. 6 Temperature-time curves comparing shock-induced combustion model with kinetic models of Libby and Momtchiloff.

to arrive, the energy is temporarily absorbed by the rotational and translational modes, so that

$$dE_{\text{vib}} = -C_p' dT \tag{11}$$

where C_p' does not include vibrational energy and equals $(7R/2)$.

If it is assumed that the chemical reaction rate depends on the translational temperature (that is, the energy with which the molecules collide), the cumulative effect of translational temperature overshoot is to increase the rate of reaction for all the components, so that the momentarily effective temperature rise at the shock will be

$$\Delta T = (E_{\text{vib2}} - E_{\text{vib1}})/C_p' \tag{12}$$

The reaction kinetics computer program can be modified by using a temperature step function to approximate the overshoot, as shown in Fig. 6. Free radical concentrations recalculated at the end of the step may be used as a starting point for a second computation, which is then continued to the point of completed reaction.

In Fig. 7, the hydrogen molecule reaction rate for a typical experimental condition is compared with the effect of a step function temperature overshoot of 150°R imposed for the duration of the vibrational relaxation. Vibrational relaxation time is defined as the time required for the temperature to reach $(1 - 1/e)$ of the difference between the initial and equilibrium value (Fig. 6, Ref. 17).

4) Assumption of one-dimensional flow: Since this assumption potentially simplifies the analysis of the experimental data, it was examined further. It has been noted from Refs. 9, 10, and 12 that the initial kinetic reactions have been treated as being relatively insensitive to variations of fuel concentration which may be caused by turbulent mixing. Although some reservations exist, the logic of the arguments by these authors establishes that a one-dimensional flow model with superimposed kinetic reactions may be used with reasonable confidence.

3.2 Comparison of Kinetic Models

Three of the kinetic calculations—constant pressure,[10] constant density,[11] and constant area[12]—have been compared for the early portion of the kinetic processes. During this time interval, the chemistry consists mainly of shuffling reactions, with little or no rise in temperature or pressure. Therefore, all three calculations can be treated as occurring at constant pressure until such time as the reaction becomes exothermic. The kinetic products can then be compared on a common basis. Some differences were found in the predicted rate of H_2 molecule reaction, as shown in Fig. 8. These variations have been attributed to the choice of reaction kinetic rates, which imposes a factor of individual judgment and accessibility of information at the instance of the investigation. The agreement on the rates is by no means universal. Momtchiloff used rate constants suggested by Bray,[18] Duff,[15] and others; Fowler used rates suggested by Schott[14, 19] and Duff[15]; and Libby employed those of Schott.[19] Westenberg's results were not compared here since his reaction assumed zero free radicals at initiation, although the reaction rates appear to be similar to those used by Libby.

3.3 Effect of Variation in Free Radical Concentration

In order to comprehend the effect of error in computing preheater free radical concentration on the shock-induced chemistry, the H_2-air reaction computer program was rerun with varying amounts of free radicals at the reaction starting time, time "zero." Figures 9–11 show the effect of varying the total concentration of free radicals from 0.1 to 10 times that calculated for a typical experiment in the Arnold Center research tunnel.

It is interesting to note that a large change in initial free radical concentration entering the reaction produces very little change in the over-all reaction speed. For example, Figs. 9 and 10 show that, in the region of rapid reaction, a reduction by a factor of 10 in initial free radicals slows the H_2 and H reaction by about 10 μsec, and in the later part of the reactions also by about 10 μsec. If the initial free radicals are increased by a factor of 10, H_2 and H reaction is speeded up by 10 μsec at first, and somewhat more as the reaction progresses. The temperature history (Fig. 11) is similar.

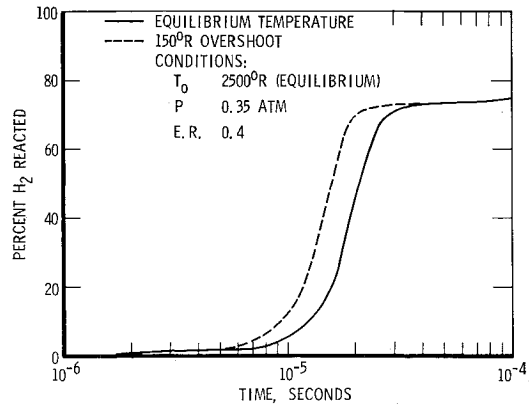

Fig. 7 Effect of translational and rotational temperature overshoot on H_2 reaction rate.

A second computer calculation was made using the same data but eliminating all the free radicals at the reaction starting point except one. The purpose of this computation was to compare the relative effects of the initial OH, H, and O radicals on the reaction progress. Figure 12 shows the effect on the H and H_2 reaction for a typical tunnel experiment, and for the hypothetical cases where no initial free radicals are present. The OH radical is shown to be most effective, as might be expected from the kinetic relations, and is also the one present in the greatest quantity. The initial OH radical, when present singly, has nearly the same effect on the reaction rate as the sum of the combined OH, H, and O components.

A further examination of Figs. 9–12 shows a large change in the reaction rate if free radicals are completely absent at time "zero." The inference is that free radicals must be present before the reaction can proceed. This being the case, new free radicals must be sought from dissociations. Since dissociation reactions are much slower than the bimolecular reactions, the combustion process during the initial period, or, as it is often called, "ignition delay" period, is considerably slower.

3.4 Factors Used in Correlating Experimental Data with Kinetics

The experimental data were recorded at various inlet temperatures and pressures so that a means of adjusting the

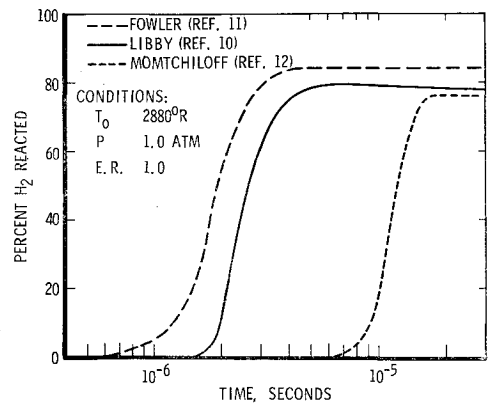

Fig. 8 Comparison of H_2 reaction rates for various kinetic calculations.

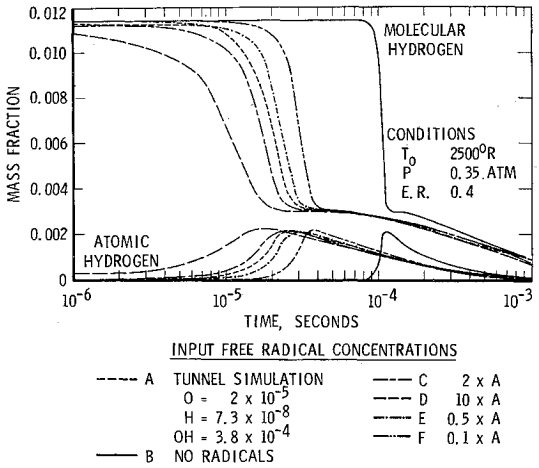

Fig. 9 Calculated mass fraction of H_2 and H vs time with large changes in input free radical concentration.

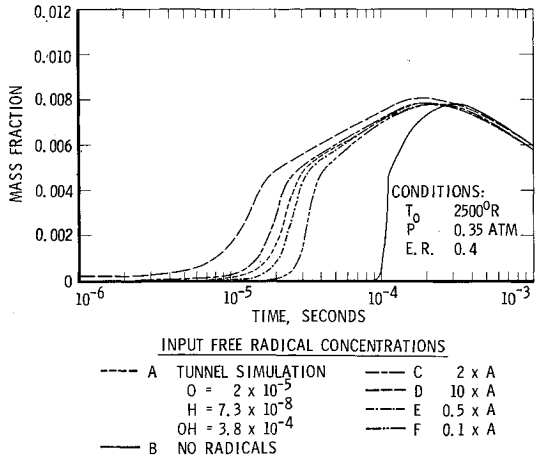

Fig. 10 Calculated mass fraction of OH vs time for large changes in input free radical concentration.

results to a common reference level was needed. Nicholls's equation[20] for "ignition delay" time, as modified by Libby,[10] contains initial temperature, pressure, and concentrations:

$$\tau_d = \left[\frac{RT_0}{6X_{1,0}p(10^{14})}\right] \exp\left[\frac{15{,}860}{T_0}\right]\left[\ln\frac{X_{1,0}}{X_{2,0}} + \ln 3 + \frac{47{,}410}{T_0}\right] \quad (13)$$

Since the third bracketed term is roughly constant, the equation may be written

$$\tau_d \sim \frac{T_0 \exp[15{,}860/T_0]}{X_{1,0}p} \quad (14)$$

where $X_{1,0}$ represents oxygen and $X_{2,0}$ hydrogen concentration at time "zero."

An assumption was made that Nicholls's equation for "ignition delay" (τ_d) can be used to describe the reaction time (τ) for any one component (i.e., hydrogen molecule) to reach a specified stage of reaction completion. This assumption would seem to apply for the early reactions and before the recombination reactions become important; so that the equation for selected conditions 1 and 2 can be written

$$\frac{\tau_1}{\tau_2} = \left[\frac{T_{01}}{T_{02}}\right]\left[\frac{P_2}{P_1}\right] \exp\left[\frac{15{,}860}{T_{01}} - \frac{15{,}860}{T_{02}}\right] \quad (15)$$

In Eq. (15), it was assumed that there is no change in component concentration ($X_{1,0}$), which therefore cancels. The equation was found to be useful in adjusting experimental

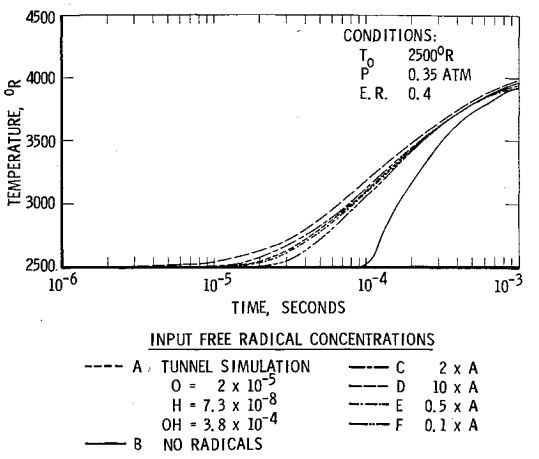

Fig. 11 Calculated temperature vs time for large changes in input free radical concentration.

data so that a comparison could be made with the kinetic calculations at similar initial conditions of temperature and pressure. It was used exclusively for the early parts of the reaction before significant change in temperature had occurred.

The validity of Eq. (15) and the assumptions used to derive it were checked by comparing the 50% hydrogen reaction point in the chemically "fast" phase, from kinetic calculations, with the adjusted values obtained from the equation. The results are shown in Fig. 13. Point *A* on the kinetically calculated curve was used as a starting point to apply the equation. Only the temperature adjustment was applied because the kinetic calculation is for a single pressure. The agreement between the kinetically calculated curve and the "adjusted" curve demonstrates that errors introduced by the equation are relatively small.

3.5 Comparison of Experimental Oblique Shock Data with the Kinetic Calculation

After the foregoing implications of theory and experiment were considered, 1) the data were adjusted to a common reference inlet temperature and pressure; 2) computer kinetic program results were obtained using inlet conditions typical of the tunnel operating condition; and 3) both were plotted in Fig. 14. The kinetic calculation was made for an equivalence ratio 0.4 but it was found, as previously noted, that the

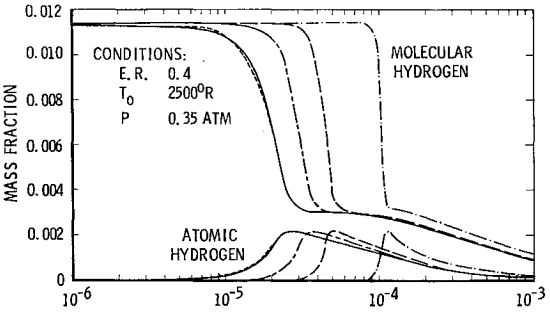

Fig. 12 Effect of single free radical concentration on calculated mass fractions of H_2 and H vs time for conditions simulating tunnel experiments.

reaction rate is relatively insensitive to fuel concentration. In fact, it is apparent that fuel concentrations below the usually accepted combustible limit will react when the initial temperature is higher than reaction temperature.

The hydrogen molecule reaction data in Fig. 14 pepper the kinetically calculated curve within the experimental error in most cases and are much closer than the agreement of all three kinetic calculations with each other (Fig. 8). Curve *A* was plotted using the digital computer program developed by Libby, et al.,[10] and the results for initial conditions typical of the Arnold Center experimental combustion tunnel. Curve *B* was obtained by assuming that all H radicals present, at the time the reacting gas entered the sampling probe, recombined to form H_2.

This recombined increment of H_2 was then added to the unburned H_2 fraction already existing to find the total amount of H_2. In the experiment, one would expect full recombination of H that enters the sampling probe to form H_2 by the time the gas reaches the analyzer equipment. Therefore, the data should be close to curve *B* if sampling and quenching were ideal, and the kinetically computed values were unimpeachable.

It is possible that the reaction is affected by the presence of the probe more than would be expected from flow analysis of the sampled gas. In this event, the H_2 reaction rate may be slower than the results show, but not faster (Fig. 8). The experimental results would then lean toward the computed values of Momtchiloff.

The inference of Fig. 14 is that the reaction history for the early portions of the H_2-air reaction can be predicted with the kinetic calculations, where kinetic rate constants have been judiciously selected. It does not decisively justify any particular method of selecting the rate constants, but it does give impetus to further experiments of this nature to verify the rate values for the heat producing reactions. These later reactions are of more direct interest to those engaged in hypersonic ramjet evaluations.

4. Conclusions

Some conclusions may be drawn from an analysis of the foregoing:

1) Shock-induced combustion experiments and reaction kinetic computations for the H_2-air reactions show that the one-dimensional flow treatment can be applied to arrive at good correlation between the kinetically computed and the experimental gas composition for quenched reactions during the early, or shuffling, reactions.

2) As a consequence of this agreement, it appears reasonable that the complete chemistry, including heat release and recombination reactions, should be predictable. Further work is needed to verify reaction rates for the important exothermic reactions.

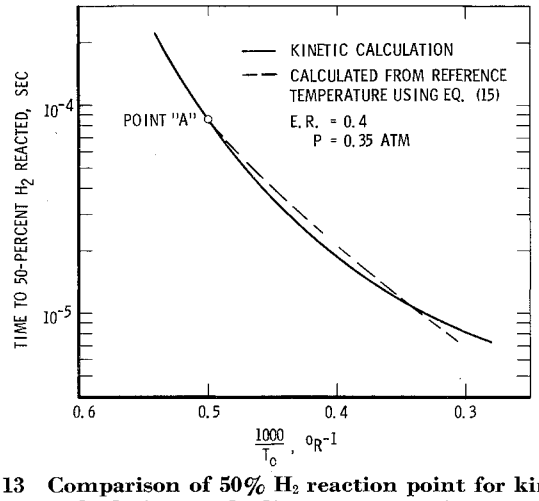

Fig. 13 Comparison of 50% H_2 reaction point for kinetic calculations and adjustment equation.

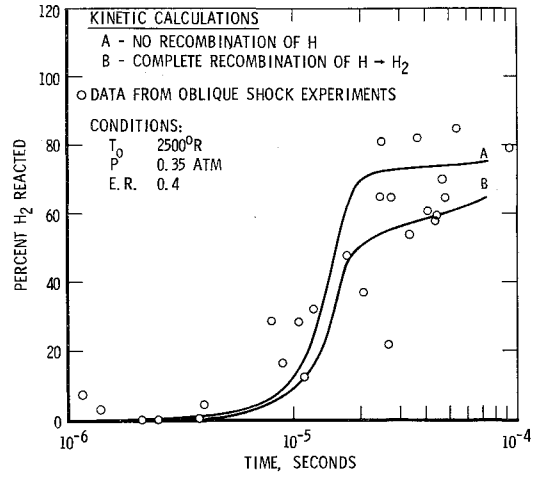

Fig. 14 Comparison of constant pressure kinetic calculations and experimental data for H_2 reaction in vitiated air.

3) Wind tunnel testing with air-breathing combustion engines, where partial vitiation of the air is a necessity, is now perceived to be on a more solid foundation where the entering gas free radical content is used with kinetic computations to calculate the effects of those free radicals on engine performance.

4) It is significant that, for combustion testing in a wind tunnel, small changes in free radical content from upstream combustion heating of the gas may be ignored. If the free radical concentration entering an engine is changed by a factor of 10, the reaction time and temperature history are changed by only a few percent.

5) The experiments have demonstrated that shock-induced chemical reaction can be experimentally produced in a constant pressure field. By analyzing the flow conditions for detonation waves, it is found that the usual detonation wave observed occurs in a constant area duct. Since shock-induced combustion can theoretically occur for constant area, static temperature, static pressure, or Mach number; the constant area detonation may be regarded as a special case of shock-induced combustion.

6) The experiments indicate that the standing oblique shock wave is a means by which kinetics of chemical reactions may be examined with relatively inelaborate instrumentation.

5. Application of Results

It has been clearly demonstrated that chemical reactions in premixed gases can be initiated by means of shock waves. A study of the present state of kinetic rate constants reveals that kinetic equations may be used to predetermine the geometry of the walls downstream of the shock, and that the wall shape may be selected so that the reaction will proceed at constant pressure, or Mach number, etc. However, it appears that further experimental verification is needed to establish the exothermic reaction rates within narrower limits than exist at the present time.

Potentially, shock-induced combustion employed in a hypersonic ramjet offers control of the combustion processes over a wide range of flight and diffuser Mach numbers. A simple calculation can be made by using one-dimensional flow aerodynamic equations and conventional Mach number tables with oblique shock functions (Fig. 15). The curves represent 1800°R static temperature isotherms, a nominal value for the lower ignition temperature for the hydrogen-air mixture. To the right of each curve, shock generated temperature is higher than 1800°R for any selected wedge angle. For a hypersonic ramjet, a diffuser Mach number

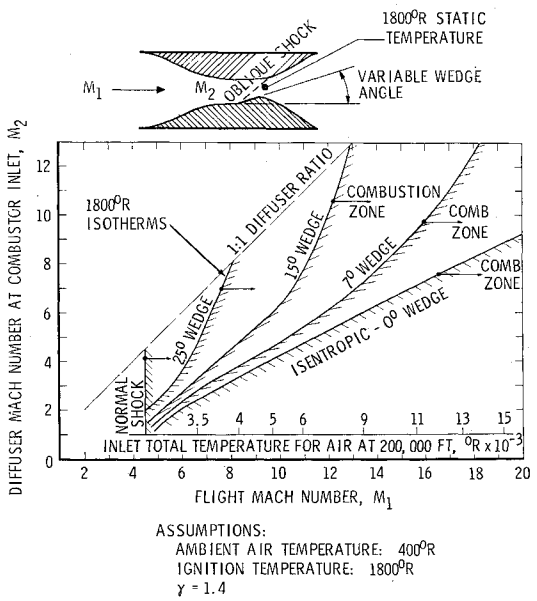

Fig. 15 1800°R isotherms to ignite H_2-air mixtures for various flight Mach numbers (M_1) and diffuser Mach numbers (M_2).

(M_2) may be selected for a fixed diffuser configuration, such that several flight Mach number values (M_1) may be compatible by changing the ignition generating wedge angle only. Excessive temperature, and the attending dissociation and recombination problems for a range of flight Mach numbers, thus can be more easily held in control by means of the variable wedge.

Since shock-induced combustion presupposes the presence of fuel and air mixture, the fuel obviously must be injected at a point somewhere upstream of the chemical reaction zone. It was not the intention of this work to develop fuel injectors for hypersonic ramjets; however, results obtained using a thin wedge for fuel injection may serve as a stimulus for further study into this problem. Fuel distribution from the wedge can be studied both experimentally and theoretically, wedge pressure losses from shocks can be calculated; and, as suggested by Roy,[3] the wedge could possibly be incorporated as a part of the inlet diffuser of a hypersonic ramjet. Also, the fuel itself acts as a coolant for the metal injector wedge.

Supersonic "diffusional burning" combines the processes of fuel injection, mixing, and combustion; each of which is a complex phenomenon. It is apparent that any complex reaction in aerothermochemistry can be more easily analyzed if broken down into physically separable processes. Thus far, investigations at the Arnold Center indicate that this separation is feasible in supersonic flow where shock-induced combustion is used to initiate the chemistry. Sufficient measurements have been obtained in the initial stages of the H_2-air reaction aft of the shock to compare favorably with calculations based on reaction kinetics for similar conditions.

On the basis of these initial results, it should be possible to proceed toward a more realistic prediction of the conditions necessary for heat release in supersonic combustion, for the H_2-air system, that will generate thrust in a hypersonic air-breathing engine.

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