REPORT 1264

A THERMAL EQUATION FOR FLAME QUENCHING[1]

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SUMMARY

An approximate thermal equation was derived for quenching distance based on a previously proposed diffusional treatment. The quenching distance was expressed in terms of the thermal conductivity, the fuel mole fraction, the heat capacity, the rate of the rate-controlling chemical reaction, a constant that depends on the geometry of the quenching surface, and one empirical constant.

The effect of pressure on quenching distance was shown to be inversely proportional to the pressure dependence of the flame reaction, with a small correction necessitated by the effect of pressure on flame temperature.

The equation was used with the Semenov equation for burning velocity to show that the quenching distance was inversely proportional to burning velocity and pressure at any given initial temperature and equivalence ratio.

In order to use the equation with experimental data, the rate-controlling reaction must be specified. Two choices were made: (1) the reaction between active particles and fuel (corresponding to the diffusional equation), and (2) the reaction between oxygen and fuel.

The two equations, one based on the active-particle—fuel reaction, the other based on the oxygen-fuel reaction, were tested using published data for the quenching of propane-oxygen-nitrogen flames. The data selected included the effect on quenching distance of oxygen-nitrogen ratio, propane concentration, and initial mixture temperature and pressure. A correlation of these data was obtained using each of the two equations, although both possessed shortcomings. The equation using the active-particle—fuel reaction did not correlate data for all rich mixtures; the equation using the oxygen-fuel reaction, while correlating data for both rich and lean mixtures, showed a larger deviation from the predicted linear relation than the other equation.

INTRODUCTION

The process of flame quenching is of interest, since it may be related to other combustion phenomena of engineering importance, such as flame stabilization, flammability limits, and the general behavior of flames near cold walls. Flame-quenching processes become especially important in turbojet combustion systems when operation at low pressure (i. e., high altitudes) is considered. Flame quenching is usually studied experimentally in terms of the quenching distance, which is defined as the minimum channel size that will allow a given flame to propagate. Most of the available quenching data have been obtained either by (1) observation of the minimum tube diameter or rectangular slot width that will allow a flame to flash back, or by (2) determination of the minimum distance between plane-parallel plates that will allow a flame to propagate from a spark of minimum ignition energy.

A rigorous theoretical treatment of the quenching process appears to be a most difficult task (refs. 1 and 2). Consequently, present theoretical treatments of quenching are necessarily approximate and seek primarily to correlate the quenching process with the variables that affect it, such as pressure, temperature, fuel type and concentration, inert-gas concentration, and quenching-surface geometry.

Approximate treatments of quenching have been based either on a thermal or a diffusional mechanism for the process (refs. 3 and 4). Such apparently different outlooks yield results consistent with experimental data because the equations for heat and mass transfer are formally identical, and because the thermodiffusivity and the molecular diffusivity are numerically nearly equal (ref. 5). Thus, each purely thermal approach to a combustion process has a diffusional analog.

A quenching-distance equation that successfully correlates lean quenching data (refs. 4, 6, and 7) is the diffusional equation given in reference 4. Unfortunately, the use of this equation in its present form is limited to stoichiometric or lean mixtures. The source of this difficulty appears to be the choice of the reaction kinetics.

Essentially, the objective of this paper is to extend, if possible, the useful range of the quenching concepts of reference 4 to include hydrocarbon-rich mixtures. Because of the similarities between heat and mass flow, it is to be expected that a thermal analog equation can be derived that will correlate data equally as well as the diffusional equation of reference 4. It is felt that such an alternate thermal equation is more susceptible to changes in the reaction kinetics than the original diffusional model. The result of such changes is an extension of the useful range of fuels and oxidants that may be treated.

[1] Supersedes NACA TN 3398, "A Thermal Equation for Flame Quenching," by A. E. Potter, Jr., and A. L. Berlad, 1955.

This report contains the derivation of a thermal quenching equation in which the rate-controlling reaction is not specified. Two possible rate-controlling reactions are postulated, and the resulting two quenching equations are tested using published quenching data. It is shown that, if the oxygen-fuel reaction is assumed to be rate-controlling, quenching data are satisfactorily correlated for both rich and lean propane-oxygen-nitrogen flames. The pressure dependence of the quenching distance and the relation of quenching distance to burning velocity are also discussed.

THEORY

It is possible to obtain the thermal equation directly from the diffusional equation (if the initial assumption is slightly altered), but it is felt that the derivation presented below is more understandable and allows the assumptions involved to be clearly seen.

For a quenching equation, an initial postulate concerning the conditions under which a flame can exist inside a tube is necessary. A reasonable postulate is as follows: If the heat lost by the flame to the tube walls exceeds a critical value, the flame will be quenched. Since a definite amount of heat is produced in a given flame, an equivalent statement is that, in order for a flame to exist, the heat retained by it must be greater than a critical amount. It will be assumed that this critical amount is a constant fraction of the total heat produced in the flame. The criterion may be written as

$$H_r \geq FH_T \tag{1}$$

(Symbols are defined in the appendix.)

For the combustion of hydrocarbons, complete combustion of the fuel is assumed to occur for all mixture compositions. Consequently, the total heat produced by the process may be written as the product of the mole fraction of fuel in the unburned gas X_f and the heat produced upon the disappearance of 1 mole of fuel ΔH (as defined herein, ΔH is constant and equal to the heat of combustion to CO_2 and H_2O only for stoichiometric or lean mixtures; for rich mixtures, ΔH decreases as the oxygen concentration decreases). Thus, for hydrocarbon flames, equation (1) may be written as

$$H_r \geq FX_f\Delta H \tag{2}$$

The heat retained by the flame can be written in terms of heat capacity and average temperature of the flame in the tube

$$H_r = \overline{C}_p(\overline{T}-T_o) \tag{3}$$

Equation (2) can then be written as

$$\overline{C}_p(\overline{T}-T_o) \geq FX_f\Delta H \tag{4}$$

The average temperature of the flame may be found if it is assumed that heat flow to the walls follows a differential equation of the form

$$(T-T_o)''=\frac{q}{\chi} \tag{5}$$

Here, q is the rate of temperature rise caused by chemical reaction, χ is the thermodiffusivity and primes refer to differentiation with respect to x.

The result of integrating and averaging equation (5) with the boundary condition $T=T_o$ at the wall is the equation

$$\overline{T}-T_o=\frac{d^2q}{G_t\chi} \tag{6}$$

In this equation d is a characteristic dimension of the tube, and G_t is a constant associated with the geometric shape of the tube. Derivations of G_t values for various geometries are given in reference 7. Equations (4) and (6) may be combined to give

$$\frac{d^2}{G_t} \geq \frac{FX_f\Delta H\chi}{q} \tag{7}$$

In order for a flame to pass down a tube, the tube size, given by d^2/G_t, must be equal to or greater than the right side of equation (7). Since the quenching distance is the characteristic tube dimension that just quenches a given flame, equation (7) can be written as a quenching-distance equation by removing the inequality sign;

$$d^2=\frac{FG_tX_f\Delta H\chi}{q} \tag{8}$$

Then the thermodiffusivity χ can be written as

$$\chi=\frac{\kappa_r RT}{PC_{p,r}} \tag{9}$$

The rate of temperature rise q can be written as

$$q=\frac{RT\Delta Hw}{P\overline{C}_pN} \tag{10}$$

Here, w is the rate of disappearance of fuel, molecules per cubic centimeter per second.

Equations (8), (9), and (10) may be combined to give for the thermal quenching equation

$$d^2=\frac{FG_tN\kappa_rX_f}{C_{p,r}w} \tag{11}$$

The most important fact concerning this equation is that the form for w is not specified; any reaction may be chosen as rate-controlling.

Equation (11) may be derived from the diffusional equation if it is assumed that the thermodiffusivity and diffusion

coefficient are equivalent, and if the criterion for flame propagation is assumed to be that the number of reaction events per cubic centimeter must be a critical fraction of the total number of events that normally occur in the unconfined flame.

In terms of the active-particle—fuel reaction (the reaction specified as rate-controlling in the diffusional equation of ref. 4), the reaction rate w is given by

$$w=k_t a_r \sum_i c_{i,r} \tag{12}$$

If a collision-theory-type temperature dependence is assigned to k_t, equation (12) becomes

$$w=B_t a_r \sum_i c_{i,r}\, T_r^{1/2} \exp\left(\frac{-E_t}{RT_r}\right) \tag{13}$$

Combination of equations (10) and (13) gives

$$d^2=\frac{F G_t N \kappa_r X_f}{C_{p,r} B_t a_r \sum_i c_{i,r}\, T_r^{1/2} \exp\left(\frac{-E_t}{RT_r}\right)}=\frac{F}{B_t}\psi_t \tag{14}$$

The choice of rate-controlling reaction is not limited to the active-particle—fuel reaction. For example, it is possible to follow Semenov (ref. 8) and assume the reaction in the combustion zone to be bimolecular and first-order with respect to fuel and oxygen. For this assumption, the real reacting species are obviously not fuel and oxygen molecules, but such an assumption is satisfactory if the concentrations of the reacting species are proportional to the fuel and oxygen concentrations. In this case, then, the rate of the reaction is given by

$$w=k_s a_r b_r \tag{15}$$

If a collision-theory-type temperature dependence is assigned to k_s, equation (15) becomes

$$w=B_s a_r b_r T_r^{1/2} \exp\left(\frac{-E_s}{RT_r}\right) \tag{16}$$

The combination of equations (10) and (16) gives

$$d^2=\frac{F G_t N \kappa_r X_f}{C_{p,r} B_s a_r b_r T_r^{1/2} \exp\left(\frac{-E_s}{RT_r}\right)}=\frac{F}{B_s}\psi_s \tag{17}$$

In order to test equations (14) and (17), data for the quenching of propane-oxygen-nitrogen flames were used. These data points include the effect on quenching distance of oxygen-nitrogen ratio, fuel concentration and pressure (ref. 6), and initial temperature (ref. 9).

In order to use equation (14), the assumptions made in reference 4 concerning the active-particle—fuel reaction were followed: T_r was taken to be 0.7 T_F, the partial pressure of fuel in the reaction zone was assumed to be one-half the partial pressure of fuel in the unburned gas, the partial pressures of the active particles (H atoms, OH radicals, and O atoms) in the reaction zone were assumed to be 0.7 times their partial pressure in the adiabatic equilibrium flame, and E_t was assumed to be 7 kilocalories per mole (ref. 10).

In order to use equation (17), the partial pressures of fuel and oxygen in the reaction zone were taken as equal to their values in the unburned gas and E_s was assumed to be 38 kilocalories per mole (ref. 11). In keeping with previous usage (refs. 4 and 6), T_r was chosen to be equal to 0.7 T_F.

The thermal conductivities were calculated for the unburned gas mixture at 0.7 T_F. The conductivities for oxygen, nitrogen, and propane were calculated at 1553° K, using the tables and data given in chapter 8 of reference 12. Conductivities at other temperatures were computed on the assumption of a ¾-power temperature dependence. The thermal conductivities of the gas mixtures were calculated as the sum of the mole fraction times the thermal conductivity of each component, or,

$$\kappa=\sum_i X_i \kappa_i \tag{18}$$

It was felt that this simple linear mixing rule was adequate for systems largely composed of nitrogen and oxygen because of their similar conductivities.

The heat capacities were calculated for the unburned gas mixture at 0.7 T_F by the same linear mixing rule used for the thermal conductivities. Individual heat capacities for oxygen, nitrogen, and propane were calculated from data given in reference 13.

The equilibrium adiabatic flame temperatures and product compositions were calculated by the matrix method of reference 14 using the thermodynamic constants of reference 14 and the heat of formation of propane given in reference 13.

RESULTS AND DISCUSSION

CORRELATION OF QUENCHING-DISTANCE DATA

Inasmuch as the square of the quenching distance is predicted to be directly proportional to ψ_t (eq. (14)) or to ψ_s (eq. (17)), values of ψ_t and ψ_s were calculated for the pressure, equilibrium adiabatic flame composition and temperature, unburned gas composition and temperature, and tube geometry (assumed to be plane-parallel plates) which correspond to the quenching distances reported in references 6 and 9 for propane-oxygen-nitrogen flames. Plots of d^2 against ψ_t and ψ_s are shown in figures 1 (a) and (b), respectively. The ranges of the variables included were: pressure, 0.1 to 1.0 atmosphere; equivalence ratio, 0.33 to 1.90; mole fraction oxygen in the oxidant mixture, 0.21 to 0.70; and unburned gas temperature, 300° to 558° K. In figure 1 (a), it is shown that ψ_t (which is entirely analogous to the diffusional equation of ref. 4 except for the choice of quenching criterion) satisfactorily correlates the effect of these variables except for propane-rich mixtures where $\varphi>1.2$.

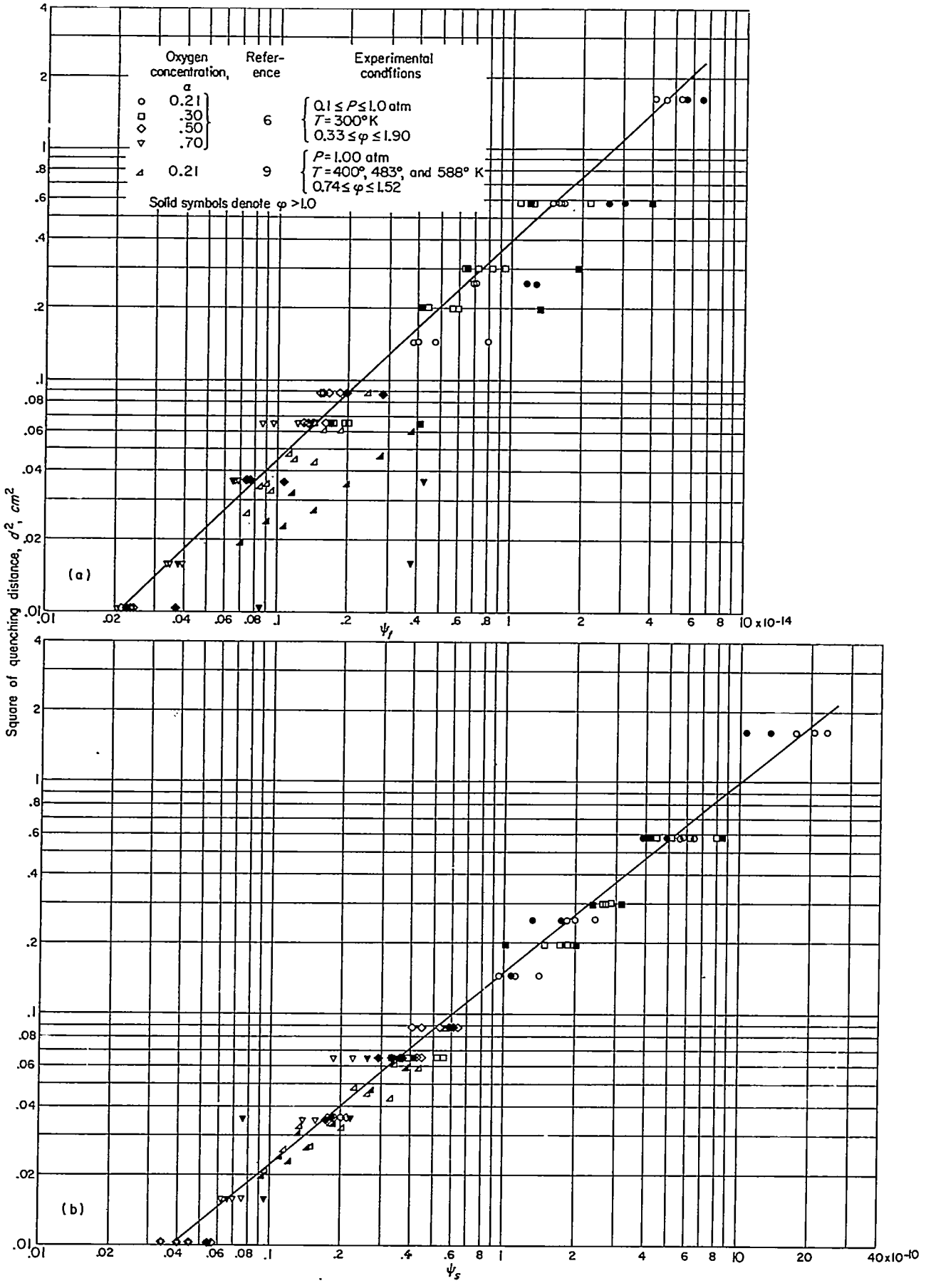

FIGURE 1.—Summary correlations of quenching data by thermal quenching equations.

Inspection of figure 1(b) reveals that ψ_s (which includes the oxygen-fuel reaction as the rate-determining step, rather than the active-particle—fuel reaction) correlates not only the propane-lean, but also all the propane-rich quenching data. Thus, it appears that by proper choice of the rate-controlling reaction, the quenching concepts of reference 4 may be extended in such a way as to include rich mixtures for the propane-oxygen-nitrogen system.

The same reaction-rate constants were used for mixtures both rich and lean in propane. This may not be possible for all hydrocarbons; it may be necessary to use different rate constants for rich and lean mixtures, since it is generally believed that the reaction mechanism changes in going from a lean to a rich hydrocarbon flame.

A mean line drawn through the data presented in figure 1 reveals that the predicted linear relation between d^2 and ψ_t and ψ_s does not hold exactly. Rather, the data are best correlated if $d^2 \propto (\psi_t)^{0.94} \propto (\psi_s)^{0.84}$. This difficulty is related, at least in part, to the choice of reaction mechanism, since the "best correlation" exponent is changed by a change in reaction mechanism.

PRESSURE DEPENDENCE OF QUENCHING DISTANCE

Examination of the equation for quenching distance (eq. (11)) reveals that all terms on the right side are independent of pressure except the reaction rate w. However, hydrocarbon flame temperatures change slightly with pressure because of changes in the extent of dissociation. Consequently, a pressure change indirectly affects temperature- and composition-dependent terms in the equation. This effect is quite small, so that the pressure dependence of the quenching distance may be regarded as primarily dependent on the pressure dependence of the initial chemical processes in the combustion wave.

The calculated pressure dependence of the quenching distance (including the indirect effect of pressure on flame temperature and composition) is compared in table I with the experimental values of reference 6. The average deviation of the calculated values from the observed values was found to be 10 percent for ψ_t and 17 percent for ψ_s. Reference 6 gives a value of 7 percent for the average deviation of values computed for the diffusional equation.

It is seen that ψ_s does not predict the pressure dependence as well as does ψ_t, except for the high-velocity flames at an oxygen fraction of 0.70. This is probably because the assumption of a simple second-order reaction between fuel and oxygen is a poor one. Concerning prediction of pressure dependence, a better choice of over-all order would be 1.7 rather than 2.0; this is in agreement with theoretical studies of the pressure dependence of burning velocity (ref. 12, p. 765), which indicate that the global reaction in most hydrocarbon flames ranges between first and second order. Such a choice would also tend to improve the linearity of the relation between d^2 (observed) and ψ_s (calculated).

QUENCHING DISTANCE AND BURNING VELOCITY

The relation of quenching distance to burning velocity has been discussed in references 3, 4, and 15. In reference 4, an equation relating the two variables is derived on a purely diffusional basis. The thermal analog to this equation may be derived as follows:

TABLE I.—COMPARISON OF OBSERVED PRESSURE DEPENDENCE OF QUENCHING DISTANCE WITH PREDICTED VALUES

Oxygen fraction, α	Equivalence ratio, φ	Exponent n describing pressure dependence, $d \propto p^{-n}$			
		Observed [a]	Predicted		
			Diffusional [a]	Thermal	
				ψ_t	ψ_s
0.17	0.943	0.90	0.88	0.83	1.03
	1.000	.89	.85	.76	1.01
	1.340	.84	.75	.75	1.00
	1.530	.71	.75	.75	1.00
0.21	0.738	0.85	0.86	0.80	1.00
	.864	.84	.89	.84	1.03
	1.000	.89	.93	.89	1.03
	1.240	.95	.78	.79	1.02
	1.490	.98	.76	.77	1.00
0.30	0.566	0.76	0.87	0.87	1.01
	.662	.93	.89	.88	1.04
	.773	1.06	.92	.90	1.07
	1.000	.98	.94	.90	1.09
	1.380	.93	.86	.84	1.06
	1.903	.74	.76	.76	1.01
0.50	0.476	1.01	0.91	0.89	1.06
	.544	1.01	.93	.91	1.07
	.680	.96	.96	.92	1.09
	1.000	.93	.97	.92	1.11
	1.358	.91	.95	.90	1.09
	1.940	.88	.82	.81	1.05
0.70	0.345	1.12	0.91	0.89	1.06
	.395	1.12	.93	.91	1.07
	.494	1.02	.96	.92	1.09
	1.000	1.01	.98	.93	1.09
	1.234	1.07	.98	.92	1.11
	1.829	.60	.89	.86	1.08

[a] Ref. 6.

According to reference 16, the Semenov equation for the burning velocity may be written as

$$U^2=\frac{2\kappa_F\overline{W}}{a_o\rho_o\overline{C}_{p,F}}\left(\frac{n_1}{n_2}\right)^m\left(\frac{\kappa}{DPc_p}\right)_F^m \tag{19}$$

Multiplying equation (11) by equation (19), converting specific heats to molar heat capacities, and writing a_o and ρ_o in the form

$$a_o=\frac{NX_fP}{RT_o} \tag{20a}$$

$$\rho_o=\frac{\overline{M}P}{RT_o} \tag{20b}$$

yield

$$d=\frac{RT_o}{UP}\sqrt{\frac{2\kappa_r\kappa_F FG_t\left(\frac{n_1}{n_2}\right)^m\left(\frac{\kappa RT}{DC_pP}\right)_F^m\overline{W}}{C_{p,r}\overline{C}_{p,F}w}} \tag{21}$$

If it is assumed that $\overline{W}$ and w have the same pressure dependence, it follows from equation (21) that, at a given equivalence ratio,

$$U\propto\frac{1}{dP}\propto P^{-(n+1)} \tag{22}$$

From reference 6, it is seen that n decreases from about -0.9 to -1.05 as the percent oxygen in the oxidant mixture is increased from 21 to 70. Simultaneously, the burning velocity increases from about 40 centimeters per second to the order of 300 centimeters per second. Consequently, equation (22) predicts that the exponent describing the pressure dependence of U should increase from about -0.1 to 0.05 as U increases from 40 centimeters per second to about 300 centimeters per second. This result may be compared with the experimental findings of Lewis (ref. 17) (which indicate an increase of about -0.05 to 0.07 as burning velocity changes from 40 to 300 cm/sec) to confirm equation (22) qualitatively.

CONCLUDING REMARKS

The fact that reasonable agreement exists between experiment and quenching concepts based on either heat or mass transfer indicates only that these processes have the same formal laws and that the thermal and molecular diffusivities are either equal or remain directly proportional for the data considered in this study. No evidence for the preponderance of either heat or mass transfer in quenching can be deduced from these results. The principal advantage gained by formulation of the quenching concepts of reference 4 in a "thermal" form is that a change in reaction kinetics is much simpler from a conceptual point of view for the "thermal" form than for the "diffusional" form. This conceptual advantage might be useful in a discussion of bizarre fuel-oxidant systems for example, CS_2–F_2 flames. In addition, the actual calculations may be simplified, since by the proper choice of reaction mechanism, it may become unnecessary to calculate the equilibrium flame composition.

SUMMARY OF RESULTS

The results of an investigation into the possibility of extending quenching concepts previously proposed to include quenching of hydrocarbon-rich flames may be summarized as follows:

1. An equation for the critical quenching configuration of a channel was derived for hydrocarbon flames:

$$d^2=\frac{FG_tN\kappa_rX_f}{C_{p,r}w}$$

where

$C_{p,r}$	heat capacity in reaction zone, cal/(°K) (mole)
d	characteristic dimension of tube geometry; quenching distance, cm
F	constant that relates total heat produced by combustion to heat which must be retained by flame for it to exist
G_t	dimensionless factor, dependent only on channel geometry
N	Avogadro's number
w	rate of reaction in reaction zone, molecules/(cc) (sec)
X_f	mole fraction of fuel in unburned gas
κ_r	mean thermal conductivity in reaction zone, cal/(cm) (sec) (°K)

2. Two possible flame-initiating reactions were considered in the detailed formulation of w: (1) the reaction between active particles and fuel molecules, and (2) the reaction between oxygen and fuel molecules.

3. The two equations resulting from the two reaction mechanism choices were tested using published data that included the effect of oxygen-nitrogen ratio, fuel concentration, pressure, and unburned gas temperature on the quenching distance for propane-oxygen-nitrogen flames. These variables were correlated reasonably well for both rich and lean flames by the equation involving the oxygen-fuel reaction. The equation involving the active-particle—fuel reaction was satisfactory primarily for lean flames.

4. It was concluded that, by the proper choice of reaction kinetics, the quenching concepts previously proposed may be cast in a form useful for the prediction of both propane-rich and -lean quenching data.

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APPENDIX

SYMBOLS

The following symbols are used in this report:

a fuel concentration, molecules/cc
B Arrhenius constant
b oxygen concentration, molecules/cc
C_p heat capacity, cal/(°K) (mole)
$\overline{C}_{p,F}$ average heat capacity, T_o to T_F, cal/(°K) (mole)
c_i concentration of ith active particle, molecules/cc
c_p specific heat, cal/(°K) (g)
$\bar{c}_p$ average specific heat, T_o to T_F, cal/(°K) (g)
D diffusion coefficient, sq cm/sec
d characteristic dimension of tube geometry; quenching distance, cm
E activation energy, cal/mole
F constant that relates total heat produced by combustion to heat which must be retained by flame for it to exist
G_t dimensionless factor, dependent only on tube geometry
ΔH heat released upon consumption of 1 mole of fuel by combustion process, cal/mole
H_r heat produced by chemical reaction in primary reaction zone, cal/mole
H_T total heat produced by combustion of 1 mole of unburned gas, cal/mole
k rate constant, (cc) (molecules)/sec
M average molecular weight of unburned gas, g/mole
m molecularity of flame reaction
N Avogadro's number
n exponent describing pressure dependence of quenching distance
n_1/n_2 moles of reactant per moles of product from stoichiometric equation
P pressure, atm
q rate of temperature rise, °K/sec
R gas constant
T temperature, °K
U flame speed, cm/sec
$\overline{W}$ average reaction rate in flame front as defined by Semenov, molecules/(cc)(sec)
w reaction rate in reaction zone, molecules/(cc)(sec)
X_f mole fraction of fuel
x distance, cm
α mole fraction of oxidant in oxidant-inert mixture
κ thermal conductivity, cal/(cm)(sec)(°K)
ρ density, g/cc
φ equivalence ratio
χ thermodiffusivity, sq cm/sec

$$\psi_s \quad \frac{G_t N \kappa_r X_f}{C_{p,r} a_r b_r T_r^{1/2} \exp \frac{-E_s}{(RT_r)}}$$

$$\psi_t \quad \frac{G_t N \kappa_r X_f}{C_{p,r} a_r \sum c_{t,r} T_r^{1/2} \exp \frac{-E_t}{(RT_r)}}$$

Subscripts:

F flame
i active-particle species
o unburned gas
r reaction zone
s involves reaction of oxygen and fuel molecules
t involves reaction of active particles and fuel molecules

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