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THE OVERALL REACTION CONCEPT IN PREMIXED LAMINAR
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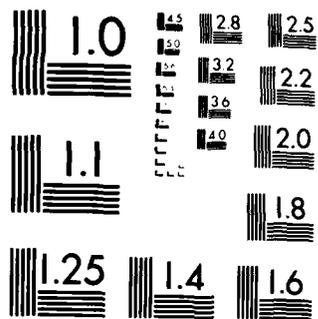
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TECHNICAL REPORT BRL-TR-2605

**THE OVERALL REACTION CONCEPT IN
PREMIXED, LAMINAR, STEADY-STATE FLAMES.
II. INITIAL TEMPERATURES AND PRESSURES**

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**Terence P. Coffee
Anthony J. Kotlar
Martin S. Miller**

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November 1984

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**US ARMY BALLISTIC RESEARCH LABORATORY
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20. ABSTRACT (Cont'd):

In this report, the adequacy of an overall reaction model for premixed, laminar, one-dimensional, steady-state flames is examined for variations in initial temperature and pressure. The single reaction model gives quite accurate results for burning velocity; temperature and heat release profiles are also generally accurate; major species profiles are reproduced with fair to good accuracy. The optimal overall parameters change with initial temperature and pressure. However, a single set of parameters are found to be accurate over a limited range of initial temperatures and pressures.

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I. INTRODUCTION

In a previous paper ¹ (hereafter referred to as I), we investigated the feasibility of representing a premixed, laminar, steady-state flame by a single overall or "global" reaction. The procedure used was to first solve the governing equations numerically for a detailed chemistry network, and then to find the appropriate overall reaction parameters, taken to be of simple Arrhenius form, by least squares fitting the heat release profile obtained from the detailed model. The resulting single equation is solved numerically. We found that the simplified system accurately represented the burning velocity and the temperature and heat release profiles of the detailed mechanism. In addition, the major species profiles were reproduced with fair precision. We considered three different flame systems (O_3 , H_2 /air, and CH_4 /air) over a range of equivalence ratios. The overall parameters generated depended on the stoichiometry as well as the detailed kinetics network. In particular, the overall activation energy was lower for a near-stoichiometric flame. This can be explained by examining the detailed model. Near stoichiometry, the burning velocity is higher, and more radicals are produced. These radicals diffuse ahead of the flame front and attack the fuel and oxidizer. These radical attack reactions have a low activation energy. The very lean or very rich flames, on the other hand, produce few radicals. The burning velocity is governed by high activation energy reactions in the flame front. The overall reaction reflects a compromise between these processes. As a result, a single overall reaction for a range of equivalence ratios does a poor job of representing burning velocities and thermal structure (temperature and heat release profiles).

In this paper, we consider the validity of the global reaction model under variations in initial temperature and pressure. Again, the overall reaction produced a faithful representation of the flame under any single set of initial conditions, but the optimum single reaction parameters generally vary (more or less, depending on the flame) when a wide range of temperatures or pressures is considered.

The use of an overall reaction leads to a vast simplification of practical problems. The analysis to follow, combined with I, is intended to provide a rational basis for judging the tradeoff between this desirable tractability and the consequent sacrifices in the accuracy with which real flames may be described.

II. GOVERNING EQUATIONS

In a previous paper,² the equations governing a one-dimensional, laminar, premixed, steady-state flame were derived for different approximations to the

¹T. P. Coffee, A. J. Kotlar, and M. S. Miller, "The Overall Reaction Concept in Premixed, Laminar, Steady-State Flames. I. Stoichiometries," *Combustion and Flame*, 54, 155-169 (1983).

²T. P. Coffee and J. M. Heimerl, "Transport Algorithms for Premixed, Laminar, Steady-State Flames," *Combustion and Flame* 43, 273-289 (1981).

multicomponent transport coefficients. We will use the simplest approximation (Method V), which still gives accurate results. To derive the overall reaction equation, we assume a single irreversible reaction of the form fuel plus oxidizer goes to product, that is,



where v_F , v_O , and v_P are the stoichiometric coefficients. The reaction rate k is expressed as an unmodified Arrhenius rate

$$k = A_r \exp(-E/RT), \quad (2)$$

where E is the activation energy and R is the gas constant. After some manipulation, the heat release q can be expressed as

$$q = Q (\rho Y_F)^{v_F} (\rho Y_O)^{v_O} A \exp(-E/RT), \quad (3)$$

where Q is the heat of reaction per unit mass of fuel. The further assumption is made that the Lewis numbers for all the species are unity. The general equations for the detailed chemistry model then reduce to the single equation

$$\frac{d}{dx} \left(\lambda \frac{dT}{dx} \right) - c_p m_o \frac{dT}{dx} = -q \quad (4)$$

The constant Q is chosen so as to obtain the proper adiabatic temperature. The constants $\rho\lambda$ and c_p are the same as for the detailed model equations. A and E are obtained from a fit of q for the detailed chemistry model. Equation (4) can then be solved numerically. Details are in I.

III. FLAMES TO BE CONSIDERED

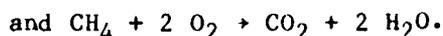
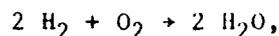
To increase the generality of our conclusions, we consider three different types of flame, which were also used in I. The simplest flame mechanism is that of an ozone decomposition flame.³ There are only 3 species and 3 reversible reactions. A more complicated case is hydrogen/air flames.⁴ The mechanism used consists of 8 species and 17 reversible

³J. M. Heimerl and T. P. Coffee, "The Detailed Modeling of Premixed, Laminar, Steady-State Flames. I. Ozone," *Combustion and Flame*, 39, 301-315 (1980).

⁴G. Dixon-Lewis, "Kinetic Mechanism, Structure and Properties of Premixed Flames in Hydrogen-Oxygen-Nitrogen Mixtures," *Proc. R. Soc. Lond. A*, 292, 45-99, (1979).

reactions. Last, we consider methane/air flames,⁵ with 20 species and 63 reversible reactions; however, the mechanism used here is different than that used in I and is more accurate for rich flames.

We will consider a 50% O₃, 50% O₂ flame, a 40% H₂, 60% air flame (slightly rich), and a stoichiometric CH₄/air flame. The overall reactions are taken to be



IV. INITIAL TEMPERATURE DEPENDENCE

In this section, we consider the effects of varying the initial (unburned) temperature T_u on the overall reaction parameters A and E. Since this involves solving the detailed chemistry model for the various initial temperatures, we shall also consider the dependence of the burning velocity on initial temperature. Andrews and Bradley⁶ have investigated this problem experimentally for CH₄/air flames. They derived the formula

$$S = 10 + .000371 T_u^2, \quad 150K < T < 1000K, \quad (5)$$

where S is the burning velocity. Tsatsaronis⁷ derived a similar formula based on his detailed chemistry model for the same flame. We shall fit our model results by the slightly more general form

$$S = a + b T_u^c. \quad (6)$$

A multi-parameter fit is made to determine a, b, and c, with a relative error criterion.

A. Ozone

The parameters for a set of ozone flames are given in Table 1. The constants $c_p, \rho\lambda$, and Q show very little change. However, A and E do increase

⁵T. P. Coffee, "Kinetic Mechanisms for Premixed, Laminar, Steady State Methane/Air Flames," *Combustion and Flame*, Vol. 55, pp. 161-170, (1984).

⁶G. E. Andrews and D. Bradley, "The Burning Velocity of Methane-Air Mixtures," *Combustion and Flame*, 19, 275-288 (1972).

⁷G. Tsatsaronis, "Prediction of Propagating Laminar Flames in Methane, Oxygen, Nitrogen Mixtures," *Combustion and Flame*, 33, 217-239, (1978).

as the initial temperature is increased. This can be explained by studying the detailed model. For a preheated mixture, more of the O_3 breaks up through the high activation energy reaction $O_3 + M \rightarrow O + O_2 + M$. For a cooler mixture, diffusion of O ahead of the flame front and the low activation energy reaction $O + O_3 \rightarrow O_2 + O_2$ is more important. This is reflected in the effective values of E .

TABLE 1. ONE-REACTION-MODEL PARAMETERS FOR A 50% O_3 , 50% O_2 FLAME AS A FUNCTION OF T_u .

T_u	c_p	T_b	Q	$\rho\lambda$	A	E
200	.254	1874	710	6.96E-8	1.20E7	7.6
300	.259	1943	709	6.86E-8	1.40E7	7.9
400	.263	2016	708	6.78E-8	1.69E7	8.2
500	.266	2093	706	6.69E-8	2.24E7	8.9
600	.269	2171	704	6.62E-8	3.12E7	9.7

The burning velocities from the detailed chemistry model (S_D) are given in Table 2. These can be fitted quite accurately (within 1.1%) by the formula

$$S = 56.8 + .00162 T_u^{2.039} \quad (7)$$

TABLE 2. A COMPARISON OF BURNING VELOCITIES FOR A 50% O_3 , 50% O_2 FLAME AS A FUNCTION OF T_u .

T_u	S_D	S_0	% error	S_1	% error
200	136.1	142.8	5	145.3	7
300	241.6	254.1	5	252.9	5
400	382.1	404.2	6	398.2	4
500	567.5	608.4	7	609.4	7
600	812.4	889.7	10	994.4	22

Table 2 also shows the burning velocities from the overall reaction model (S_0). The agreement is good, although less satisfactory for the hotter flames.

It would be more convenient to have a single fit for the kinetics, instead of having A and E depend on T_u . So we tried fitting all five heat release profiles with a single set of values for A and E . We obtained $A = 1.90E7$ and $E = 8.6$. This is similar to the values in the middle of the range, for $T = 400K$. The corresponding burning velocities S_1 are also given in Table 2. The results are less accurate at the extremes.

The overall model also reproduces the profiles reasonably accurately. The quality of the fits is similar to the case given in I. Again, agreement is better for the slower flames, where fewer O radicals are produced.

B. Hydrogen/Air

Table 3 contains the parameters for a set of hydrogen/air flames. As before, A and E increase with the initial temperature. The high activation energy (16 kcal/mole) reaction $H + O_2 \rightarrow OH + O$ becomes more important at the higher temperatures.

TABLE 3. ONE-REACTION-MODEL PARAMETERS FOR A 40% H₂, 60% AIR FLAME AS A FUNCTION OF T_u.

T _u	c _p	T _b	Q	ρλ	A	E
200	.412	2140	28710	7.42E-8	1.23E15	5.4
300	.416	2215	28599	7.32E-8	2.20E15	6.5
400	.420	2287	28442	7.23E-8	4.00E15	7.8
500	.423	2356	28228	7.15E-8	7.52E15	9.4
600	.427	2422	27948	7.08E-8	1.45E16	11.2

Table 4 contains the burning velocities. The detailed model speeds can be fitted accurately (within .3%) by the formula

$$S = 44.2 + .01906 T_u^{1.655} \quad (8)$$

The exponent is no longer near two, as reported by Andrews and Bradley and by Tsatsaronis. So the optimal exponent does depend on the flame.

TABLE 4. A COMPARISON OF BURNING VELOCITIES FOR A 40% H₂, 60% AIR FLAME AS A FUNCTION OF T_u.

T _u	S _D	S _O	% error	S _I	% error
200	166.5	158.8	-5	99.5	-40
300	284.4	269.8	-5	181.7	-36
400	429.0	406.1	-5	313.5	-27
500	600.8	580.3	-3	604.6	1
600	799.8	800.8	0	1235.1	54

The overall reaction model does an excellent job of reproducing the burning velocities. We also fit all five profiles simultaneously and obtained

$A = 7.24E15$ and $E = 9.3$. The corresponding burning velocities S_1 suffer considerable loss in accuracy, especially at the extremes.

The overall model reproduces the temperature and heat release profiles fairly well, but the species profiles are inaccurate due to the large radical concentrations (see I). As before, agreement is better for the slower flames, where fewer radicals are produced.

C. Methane/Air

The parameters of the methane/air flame are in Table 5. A and E remain almost constant between 200K and 500K before increasing with the initial temperature as before. There is no obvious reason for this behavior. The burning velocities from the detailed model in Table 6 can be fitted accurately (within 4%) by the formula

$$S = 10 + 5.532E-5 T_u^{2.290}. \quad (9)$$

The exponent is slightly different than that determined experimentally by Andrews and Bradley (Eq.(5)), but the values for the burning velocity are in good agreement with the experimental data.

TABLE 5. ONE-REACTION-MODEL PARAMETERS FOR A STOICHIOMETRIC CH₄/AIR FLAME AS A FUNCTION OF T_u.

T _u	c _p	T _b	Q	ρλ	A	E
200	.319	2181	11483	5.68E-8	2.21E18	31.6
300	.323	2232	11352	5.62E-8	1.92E18	31.2
400	.328	2281	11201	5.57E-8	1.89E18	31.2
500	.332	2329	11029	5.53E-8	2.17E18	31.7
600	.336	2376	10834	5.48E-8	2.71E18	32.6
700	.339	2422	10615	5.45E-8	3.66E18	33.8
800	.343	2466	10372	5.41E-8	5.02E18	35.1
900	.345	2510	10105	5.38E-8	8.33E18	37.1
1000	.348	2552	9815	5.35E-8	1.55E19	39.7

The overall reaction model does extremely well in reproducing the burning velocities. If all the profiles are fitted simultaneously, we obtain $A = 2.56E18$ and $E = 32.7$. This single set of parameters does a reasonable job of reproducing the burning velocities except at extremely high T_u.

The overall model reproduces the profiles fairly well. The main source of error is in neglecting the intermediates H₂ and CO (see I). Again, agreement is better for the slower flames, where less H₂ and CO is produced.

TABLE 6. A COMPARISON OF BURNING VELOCITIES FOR A STOICHIOMETRIC CH₄/AIR FLAME AS A FUNCTION OF T_u.

T _u	S _D	S _O	% error	S _I	% error
200	20.0	20.2	1	18.0	-10
300	37.4	37.4	0	33.4	-11
400	61.2	60.7	-1	54.9	-10
500	93.0	91.7	-1	84.6	-9
600	134.6	131.6	-2	125.6	-7
700	187.6	183.0	-2	182.1	-3
800	253.9	247.7	-2	261.7	3
900	335.2	328.8	-2	380.0	13
1000	432.7	429.0	-1	584.3	35

V. PRESSURE DEPENDENCE

Unlike initial temperature, an analytic expression can be derived for the dependence of burning velocity on pressure, under certain assumptions. Consider the equation governing the overall reaction, that is,

$$\frac{d}{dx} \left(\lambda \frac{dT}{dx} \right) - c_p m_o \frac{dT}{dx} = Q (\rho Y_F)^{\nu_F} (\rho Y_O)^{\nu_O} A \exp(-E/RT), \quad (10)$$

based on Eqs. (4) and (5). The order of the reaction is $n = \nu_F + \nu_O$. Now increase the pressure by a factor α . That is, if p is the original pressure, the new pressure is αp . Eq. (10) can then be written as

$$\frac{d}{dx} \left(\lambda \frac{dT}{dx} \right) - c_p m_o \frac{dT}{dx} = \alpha^n Q (\rho Y_F)^{\nu_F} (\rho Y_O)^{\nu_O} A \exp(-E/RT). \quad (11)$$

Apply the transformation $x' = \alpha^{n/2} x$ and then divide by α^n , resulting in

$$\frac{d}{dx'} \left(\lambda \frac{dT}{dx'} \right) - c_p \frac{m_o}{\alpha^{n/2}} \frac{dT}{dx'} = Q (\rho Y_F)^{\nu_F} (\rho Y_O)^{\nu_O} A \exp(-E/RT). \quad (12)$$

This is formally identical to Eq. (10), except that the mass flux is divided by $\alpha^{n/2}$. Here we assume that λ , c_p , and Q are independent of pressure, which is an excellent approximation. The mass flux $m_o = \rho_o S$, where ρ_o is the density of the original mixture. Since ρ_o is proportional to p , the result is that

$$S \propto p^{n/2-1}. \quad (13)$$

This is similar to the relation derived by Williams.⁸ However, fewer assumptions have been made in the derivation, and the result also holds for a detailed chemistry model, if all the reactions have the same order.

Historically, Eq. (13) has been used to fit burning velocities as a function of pressure. Andrews and Bradley⁶ derived the formula

$$S = 43 p^{-0.5}, \quad p > 5 \text{ atm.} \quad (14)$$

from experimental data for CH₄/air flames. However, this fit does not hold for lower pressures. The change of the exponent with pressure has been explained by Westbrook and Dryer.⁹ Most of the important reactions in the mechanism are second order. But for higher pressure flames, the third order reaction $H + O_2 + M \rightarrow HO_2 + M$ becomes important. This reaction is a sink for H atoms and causes the flame to slow down. At higher pressures the burning velocity will fall below the predictions of a model based on Eq. (13).

Consequently if a kinetics system includes H₂/O₂ reactions, Eq. (13) generally will not hold over a large pressure range near atmospheric pressure, and a single set of overall reaction parameters will not be a good approximation. This is verified in the data given below. Fits over a range of high pressures or low pressures will probably be more accurate.

A. Ozone

The results for an ozone flame are in Tables 7 and 8. The burning velocity based on the detailed chemistry model (S_D) is practically constant and decreases only slowly with pressure. This is because the flame is primarily governed by the second order reactions $O_3 + M \rightarrow O_2 + O + M$ and $O_3 + O \rightarrow O_2 + O_2$. By Eq. (13), this implies that S is constant with respect to p . The three body reaction $O + O + M \rightarrow O_2 + M$ is much less important. However, at higher pressures, it will remove more radicals and slow down the flame.

The quantities c_p , T_b , Q , and λ are almost constant with respect to pressure. The parameters A_1 and E_1 are based on the usual overall reaction $O_3 \rightarrow 1.5 O_2$ ($n=1$). The activation energy is essentially constant, but the pre-exponential factor increases with pressure. Since the order n should equal 2, we also computed parameters A_2 and E_2 based on the overall reaction $2 O_3 \rightarrow 3 O_2$. Both A_2 and E_2 are almost constant, reflecting the lack of change of the detailed kinetics mechanism with pressure. This fit is also somewhat more accurate (Table 8).

⁸F. A. Williams, *Combustion Theory*, Addison-Wesley Co. Inc., Reading, Massachusetts (1965), p. 99.

⁹C. K. Westbrook and F. L. Dryer, "Prediction of Laminar Flame Properties of Methanol-Air Mixtures," *Combustion and Flame* 37, 171-192 (1980).

TABLE 7. ONE-REACTION-MODEL PARAMETERS FOR A 50% O₃, 50% O₂ FLAME AS A FUNCTION OF p.

<u>p</u>	<u>c_p</u>	<u>T_b</u>	<u>Q</u>	<u>ρλ</u>	<u>A₁</u>	<u>E₁</u>	<u>A₂</u>	<u>E₂</u>
0.1	.259	1937	707	6.87E-9	1.42E6	7.9	1.16E12	12.7
0.2	.259	1940	708	1.37E-8	2.82E6	7.9	1.14E12	12.7
0.5	.259	1942	709	3.43E-8	6.95E6	7.9	1.12E12	12.7
1.0	.259	1943	709	6.86E-8	1.40E7	7.9	1.12E12	12.7
2.0	.259	1944	709	1.37E-7	2.78E7	7.9	1.11E12	12.7
5.0	.259	1944	710	3.43E-7	7.05E7	7.9	1.13E12	12.7
10.0	.259	1945	710	6.86E-7	1.42E8	8.0	1.12E12	12.7

TABLE 8. A COMPARISON OF BURNING VELOCITIES FOR A 50% O₃, 50% O₂ FLAME AS A FUNCTION OF p.

<u>p</u>	<u>S_D</u>	<u>S_{o1}</u>	<u>% error</u>	<u>S_{o2}</u>	<u>% error</u>
0.1	242.1	254.2	5	240.8	-1
0.2	242.0	254.1	5	240.7	-1
0.5	241.9	253.9	5	240.6	-1
1.0	241.6	254.1	5	240.6	-1
2.0	241.2	253.4	5	240.1	-0
5.0	240.1	253.0	5	238.9	-0
10.0	238.7	251.6	5	240.3	-1

B. Hydrogen/Air

Tables 9 and 10 contain the results for a 40% H₂, 60% air flame. If the burning velocity S_D is fit using Eq. (13), the result is

$$S = 261.3 p^{.0976} \quad (15)$$

The corresponding order $n = 2.20$ ($v_F = 1.47$ and $v_o = .73$). However, this is only an approximation, and the effective order does decrease with pressure. Since the three body reaction $H + O_2 + M \rightarrow HO_2 + M$ is important as well as various two body reactions, there is no reason to believe that the effective order will be constant over a pressure range.

The parameters A₁ and E₁ are based on the previous overall reaction $2 H_2 + O_2 \rightarrow 2 H_2O$ ($n = 3$). The activation energy increases with pressure. At higher pressures, there are fewer radicals produced, and the flame front is much thinner.

We also generated the parameters A₂ and E₂, based on the overall reaction $1.5 H_2 + .75 O_2 \rightarrow 1.5 H_2 O$ ($n = 2.25$). This is close to the average order. The parameters still change with pressure, since the effective order still

changes. The fits are slightly more accurate, however.

TABLE 9. ONE-REACTION-MODEL PARAMETERS FOR A 40% H₂, 60% AIR FLAME AS A FUNCTION OF p.

p	c _p	T _b	Q	ρλ	A ₁	E ₁	A ₂	E ₂
0.1	.415	2184	28105	7.36E-9	4.39E15	5.8	8.93E10	4.5
0.2	.415	2196	28297	1.47E-8	3.56E15	5.9	1.14E11	4.6
0.5	.416	2208	28491	3.67E-8	2.64E15	6.1	1.61E11	4.8
1.0	.416	2215	28599	7.32E-8	2.20E15	6.5	2.09E11	4.9
2.0	.416	2220	28681	1.46E-7	1.92E15	7.0	2.82E11	5.3
5.0	.416	2225	28757	3.66E-7	1.74E15	8.0	4.42E11	6.0
10.0	.416	2227	28797	7.31E-7	1.83E15	9.2	6.71E11	7.0

TABLE 10. A COMPARISON OF BURNING VELOCITIES FOR A 40% H₂, 60% AIR FLAME AS A FUNCTION OF p.

p	S _D	S _{O1}	% error	S _{O2}	% error	S ₁	% error
0.1	192.4	171.5	-11	180.0	-6	179.6	-7
0.2	221.4	198.7	-10	208.9	-6	196.6	-11
0.5	259.2	240.5	-7	257.1	-1	217.8	-16
1.0	284.4	269.8	-5	285.5	0	237.8	-16
2.0	302.9	290.2	-4	307.2	1	259.9	-14
5.0	310.2	303.4	-2	317.4	2	292.5	-6
10.0	296.7	294.0	-1	303.5	2	317.8	7

Finally, all seven profiles were fit simultaneously, with the order $n = 2.25$. We obtained $A = 2.02E11$ and $E = 5.3$. The corresponding burning velocities S_1 are still reasonable, though there is a considerable loss of accuracy.

All three models reproduce the profiles about as well as in I. Agreement is better for the high pressure flames, where fewer radicals are produced.

C. Methane/Air

The parameters for a stoichiometric CH₄/air flame are given in Tables 11 and 12. Fitting the burning velocities S_D , the result is

$$S = 30.25 p^{-.3685}. \quad (16)$$

The corresponding order is $n = 1.26$ ($v_F = .42$ and $v_O = .84$). The effective order, however, decreases rapidly with increasing pressure, and Eq. (16) is not particularly accurate.

TABLE 11. ONE-REACTION-MODEL PARAMETERS FOR A STOICHIOMETRIC CH₄ /AIR FLAME.

P	c _p	T _b	Q	ρλ	A ₁	E ₁	A ₂	E ₂
0.1	.322	2171	10950	5.66E-9	4.94E18	24.6	5.42E6	13.5
0.2	.323	2191	11084	1.13E-8	3.02E18	25.7	9.94E6	14.2
0.5	.323	2216	11244	2.82E-8	1.86E18	28.0	2.38E7	15.8
1.0	.323	2232	11352	5.62E-8	1.92E18	31.2	5.52E7	17.9
2.0	.324	2247	11449	1.12E-7	3.84E18	37.1	2.06E8	22.0
5.0	.324	2264	11560	2.80E-7	6.04E19	52.3	2.96E9	31.9
10.0	.324	2275	11631	5.60E-7	5.60E21	74.7	1.21E11	47.4

TABLE 12. A COMPARISON OF BURNING VELOCITIES FOR A STOICHIOMETRIC CH₄/AIR FLAME AS A FUNCTION OF p.

P	S _D	S ₀₁	% error	S ₀₂	% error	S ₁	% error
0.1	59.6	57.2	-4	62.5	5	72.4	21
0.2	55.1	53.0	-4	58.4	6	57.4	4
0.5	45.6	44.9	-2	49.7	9	42.1	-8
1.0	37.4	37.4	0	41.6	11	33.2	-11
2.0	28.3	28.7	1	32.3	14	26.1	-8
5.0	17.6	18.0	2	20.5	16	19.0	8
10.0	10.7	10.9	2	12.5	17	14.8	38

The parameters A₁ and E₁ are based on the previous overall reaction CH₄ + 2 O₂ + CO₂ + 2 H₂O (n = 3). The activation energy increases rapidly with pressure. High activation energy seems to correlate with a small effective order, but we do not understand why.

The parameters A₂ and E₂ are based on the overall reaction .42 CH₄ + .84 O₂ + .42 CO₂ + .84 H₂O, consistent with Eq. (16). The parameters still change rapidly with pressure, since the effective order does vary with pressure, and the fitted order is not very accurate at low or high pressures. The burning velocities S₀₂ based on these parameters are actually less accurate than those based on the original overall reaction.

Again, all seven profiles were fit simultaneously with the order n = 1.26. The result is A = 4.69E7 and E = 18.7. The corresponding burning velocities S₁ are less accurate for low or high pressures, since the effective order changes so rapidly in this case.

The first two models reproduce the profiles about as well as in I. The single set of parameters is somewhat less accurate.

VI. CONCLUSIONS

In I, we concluded that a single reaction formalism, having a rate constant of simple Arrhenius form, could reproduce the burning velocity, thermal structure, and to some extent, species profiles obtained from multi-reaction network calculations for a range of stoichiometries. This result has been generalized to ranges of initial temperature and pressure.

Previously, we showed that the optimal values of the parameters vary with stoichiometry. Here we find the same is true of the temperature and pressure. A single set of fitting parameters, with a few exceptions, does a reasonable job only over a limited range of stoichiometries, temperatures, or pressures. Over a larger range, we feel that it is necessary to change the fitting parameters or to investigate more complicated schemes than a single overall reaction in order to achieve reasonable agreement with the detailed reaction model.

However, over moderate ranges of initial temperature or pressure, a single set of parameters may well be sufficient. When considering variation in the initial temperature, usually only changes of a few hundred K are considered. Over this range the optimal parameters change very little.

For actual problems, very large variations in pressure are often considered. In this paper, we discovered one case (ozone flame) where the overall order and the corresponding fitting parameters hardly change with pressure. For hydrogen/air or methane/air flames, there is a large change in the overall order near atmospheric pressure. Yet even over this range considered in this paper (0.1 to 10.0 atm.), a single set of parameters does a reasonable job of reproducing burning velocities. For ranges of pressure that are not near atmospheric pressure, the accuracy will probably be improved.

The structure of the flame changes less for changes in initial temperature and pressure than it does for changes in stoichiometry. Over a moderate range of initial conditions, or if only moderate accuracy is required, a single overall reaction is adequate. Even if the need for additional accuracy requires treating the fitting parameters A and E as a function of the initial conditions, simplification of the problem in terms of the overall reaction concept can be considerable.

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LIST OF SYMBOLS

- A = lumped parameter, cgs units.
- c_p = specific heat of the mixture, $\text{cal-g}^{-1}\text{-K}^{-1}$.
- E = activation energy, kcal-mole^{-1} .
- k = rate constant, in centimeter-mole-second units.
- m_o = mass flux, $\text{g-cm}^{-2}\text{-s}^{-1}$.
- p = pressure, atm.
- q = heat release per unit volume, $\text{cal-cm}^{-3}\text{-s}^{-1}$.
- Q = heat of reaction per unit mass of fuel, cal-g^{-1} .
- R = gas constant, $1.9872\text{E-3 kcal-mole}^{-1}\text{-K}^{-1}$.
- S = burning velocity, cm-s^{-1} .
- T = temperature, K.
- T_b = temperature of the burned mixture, K.
- T_u = temperature of the unburned mixture, K.
- x = spatial coordinate, cm.
- Y_F = mass fraction of fuel.
- Y_O = mass fraction of oxidizer.
- λ = thermal conductivity of the mixture, $\text{cal-cm}^{-1}\text{-s}^{-1}\text{-K}^{-1}$.
- ρ = density of the mixture, g-cm^{-3} .

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