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LAMINAR BURNING VELOCITIES OF METHANE-
OXYGEN-DILUENT GAS MIXTURES

by

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ABSTRACT

The burning velocities of methane-oxygen-diluent mixtures were determined at atmospheric pressure by use of bunsen flames under laminar flow conditions. The fuel-lean region was investigated at theoretical flame temperatures of 2000, 2250 and 2500°K employing nitrogen, helium and argon as diluents. By maintaining constant calculated flame temperatures for the different mixtures, evaluation of the effects of the properties of the diluent and of O_2/CH_4 ratio was greatly simplified. The dependence of burning velocity on O_2/CH_4 ratio was not in accord with the approximate theories of combustion. Therefore, an empirical expression was developed to describe the effect of diluent, O_2/CH_4 ratio, and flame temperature on burning velocity. The evaluation of this expression is discussed in terms of effective transport properties and concentrations.

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INTRODUCTION

The laminar burning velocity of a fuel-oxidant mixture is an important parameter in the expressions which have been developed to describe such combustion phenomena as ignition energies and flame stability.⁴ Many relationships for turbulent flames have been developed which require knowledge of the laminar burning velocity of the same reaction system. Therefore, it would be extremely useful if the laminar burning velocity of a given fuel-oxidant system could be predicted with good accuracy from knowledge of the composition of the reactant system and quantities calculable from the composition and conditions of the system.

The prediction of burning velocities by use of the approximate theories of flame propagation developed by Semenov⁸, and Tanford and Pease¹⁰ has been only partially successful^{2,9,11}. These relationships adequately describe experimental data only in those cases in which flame temperature is the major variation in flame conditions. The effects of variations in unburned gas compositions on burning velocity cannot as yet be defined with accuracy², presumably because of the approximations of mechanism and transport properties which are necessary to obtain analytical expressions. The more rigorous theories of combustion in flames do not readily lend themselves to prediction of the functional dependence of burning velocity upon composition. The ability of rigorous theories to predict absolute burning velocities for the few cases that have been treated, is not as valuable as their potential ability to describe the internal structure of the flame from knowledge of the burning velocity.

An analytical expression would best suit the practical purpose of predicting burning velocities, even if it were determined empirically. As its contribution to a better understanding of the combustion process, an empirical expression could be used as a basis for speculation on kinetic expressions that might be used in the integration of the fundamental equations of flame propagation.

To achieve such a goal, an empirical equation must show the functional dependence of burning velocity on unburned gas composition. If the data, upon which the expression is based, are restricted to fuel-air systems in which fuel concentration is the only variable, the dependence of burning velocity on composition cannot be distinguished from its far greater dependence on flame temperature. In divariant systems consisting of fuel, oxygen and diluent, the equivalence ratio (actual fuel/oxygen ratio divided by stoichiometric ratio) can be varied while the theoretical flame temperature is held constant. In this manner the functional

relationship between burning velocity and equivalence ratio can be investigated with temperature effects being held to a minimum. Because of the significant effect of the surrounding atmosphere on the chemistry of the combustion of fuel-rich bunsen flames, and the additional complexities introduced thereby, this initial study was limited to fuel-lean flames.

EQUIPMENT AND PROCEDURES

Burning velocities were determined by the total area method using shadowgraphs of bunsen flames. It is recognized that burning velocities determined in this manner are not absolute values. The literature shows that no experimental method is free of some question in this respect, and that the absolute burning velocities obtained may show a strong dependence upon the experimental method used.⁵ However, in some instances, radically different methods have yielded the same absolute values.⁶ It is of major significance that different methods indicate the same functional dependence upon the composition variables.² The method used in this work does yield reproducible relative values that can be used to establish the functional dependence of burning velocity on unburned mixture composition. It is this dependence that is of greatest value in efforts to determine the mechanism of flame propagation from experimental results. Furthermore, the method used is simple and duplicates conditions encountered in many practical applications.

A gas metering and mixing panel was used to obtain the desired burner feed compositions, and contained four separate metering systems, each of which utilized a carefully calibrated critical-flow jewel orifice. From its cylinder, each gas passed through activated alumina driers and sintered bronze filters to Nullmatic pressure regulators, capable of accurately maintaining the desired pressure upstream of the orifice. Inlet pressure to the orifices was measured precisely by means of ten-foot mercury manometers. The orifice assemblies consisted of special companion flanges supporting removable brass plates; each plate contained a synthetic sapphire drilled to closely conform to the standard configuration for a round-edged orifice. Downstream from the orifices the individual gas streams were joined and valves were provided so that it was possible to direct the mixture to a wet test meter or the burner. Each orifice was calibrated before each run, and with this system measurements of the relative flow of the gas streams were accurate to within $\pm 0.2\%$, the absolute flows to within $\pm 0.5\%$.

The optical system used for photographing the flame was of the "Z" type, incorporating two paraboloidal mirrors.³ A 2-watt zirconium concentrated arc lamp was used as a light source. The two front-surfaced parabolic mirrors were 6 inches in diameter, with a focal length of 48 inches. A flat front-surfaced mirror, used to increase the angle of reflection and enable easier recording of the image, was placed at the same angle with respect to the second mirror as the light source was placed with respect to the first. The image was recorded by means of a camera, with the lens removed, on Contrast Process Panchromatic film.

The burners, for obtaining the bunsen flames used in this study, were cylindrical tubes at least 30 inches long. The upper portion was surrounded with a waterjacket connected to a constant temperature bath and circulating pump. Three burners were necessary for the range of burning velocities encountered. The coolest flames

(2000°K) were studied using a burner of 0.375-inch inside diameter. For the 2250°K and 2500°K temperature levels, burners with inside diameters of 0.250 and 0.188 inches, respectively, were used.

The relative amounts of fuel, oxygen, and diluent gas fed to the burner tube were adjusted to obtain the desired adiabatic reaction temperature. The necessary quantities of these gases were calculated with the usual assumptions of chemical equilibria using the thermodynamic properties as tabulated in the National Bureau of Standards Circular C461.⁷ The results of these calculations are presented in Table 1.

Representative mass spectrometer analyses of the gases used in these studies are given in Table 2.

TABLE 2
ANALYSES OF GASES

<u>Methane</u>		<u>Propane</u>		<u>Oxygen</u>	
CH ₄	99.2%	C ₃ H ₈	99.4%	O ₂	99.5%
C ₂ H ₆	0.4	C ₂ H ₆	0.6	N ₂	0.4
C ₃ H ₈	0.2			CO ₂	0.1
N ₂	0.1				
CO ₂	0.1				
 <u>Nitrogen</u>		 <u>Argon</u>		 <u>Helium</u>	
N ₂	99.9%	A	99.9%	He	100%
A	0.1	N ₂	0.1		
O ₂	0.05				

EXPERIMENTAL RESULTS

The observed values of burning velocity of fuel-lean methane flames are presented in Figs. 1-3. The diluents used to control calculated flame temperatures were nitrogen, argon and helium. The abscissa is the reciprocal of the more generally used equivalence ratio since the fuel-lean region is of greatest interest.

In these plots, the maximum amount of diluent occurs at the stoichiometric point, and the extreme lean limits represent binary mixtures of fuel and oxygen. The general behavior of these curves in the leaner regions is reasonable in that the slopes agree with expectations based on the simplest consideration of transport phenomena. Thus with increasing (O_2/CH_4), the transport coefficients would be expected to decrease in the helium system, increase in the argon system and remain essentially constant in the nitrogen system.

The maximum in the 2000°K curve of Fig. 1 is based on differences slightly greater than the reproducibility of the data. Contention that the burning velocity is approaching a limiting value cannot be rejected on the basis of these data. From the appearance of the curves in Fig. 1 it would appear that at a lower temperature level, with the greater range of (O_2/CH_4) to be encountered, a maximum would be accentuated. Attempts to achieve this at 1800°K failed due to formation of cellular flames.

Table 1.-OBSERVED BURNING VELOCITIES AND CALCULATED REACTANT COMPOSITIONS FOR THE SYSTEMS INVESTIGATED

METHANE-OXYGEN-NITROGEN 2000°K Burner Diam = 0.375 in.					METHANE-OXYGEN-HELIUM and METHANE-OXYGEN-ARGON 2000°K Burner Diam = 0.375 in.					
O ₂ /CH ₄ Stoich. Ratio	Reactant Composition			Burning Velocity ft/sec	O ₂ /CH ₄ Stoich. Ratio	Reactant Composition			Burning Velocity (ft/sec)	
	CH ₄	O ₂	N ₂			CH ₄	O ₂	Inert	CH ₄ -O ₂ -He	CH ₄ -O ₂ -A
1.00	7.82	15.63	76.55	0.65*	1.50	5.51	16.52	77.79	1.14*	0.96*
				0.74*						0.98*
1.50	7.81	23.43	68.76	1.03*	2.00	5.74	22.96	71.30	1.07*	1.29*
				1.00*						1.29*
2.00	7.87	31.48	60.65	1.10	2.50	5.97	29.84	64.19	2.53	1.09*
2.50	7.91	39.59	52.49	1.21						1.24*
				1.22	3.00	6.22	37.34	56.44	2.37	1.27*
				1.17						1.15
				1.20	3.50	6.50	45.48	48.02	2.23	1.12
				1.21	4.00	6.30	54.38	38.83	1.91	1.18
3.00	7.96	47.75	44.29	1.21						1.25
3.50	7.99	55.97	36.04	1.30						1.17
				1.36						1.19
4.00	8.09	64.70	27.21	1.37	4.50	7.12	64.03	28.85	1.84	1.22
				1.41						1.26
				1.36						1.20
4.50	8.04	72.32	19.64	1.37						1.22
				1.35						1.22
				1.39						1.23
5.25	8.05	84.50	7.45	1.30	5.00	7.49	74.86	17.65	1.55	1.32
				1.30						1.23
5.70	8.05	91.95	0	1.21	5.70	8.06	91.94	0	1.22	1.26
				1.24						
				1.21						
				1.28						
2250°K Burner Diam = 0.25 in.					2250°K Burner Diam = 0.25 in.					
1.00	9.72	19.45	70.83	1.43	1.00	6.92	13.84	79.24	2.50†	1.29
				1.49	1.50	6.85	20.57	72.58	3.74	1.38
				2.03					3.57	1.47
1.50	9.54	28.63	61.82	2.00	2.00	7.21	28.85	63.94	3.70	1.51
				2.38					3.75†	1.92
2.00	9.61	38.46	51.93	2.64	2.50	7.61	38.04	54.36	3.91	1.91
2.50	9.70	48.49	41.82	2.83					3.66	2.11
3.00	9.76	58.57	31.67	2.75	3.00	8.02	48.14	43.83	3.57	2.23
				2.95					3.78	2.28
3.50	9.81	68.67	21.52	2.94	3.50	8.51	59.58	31.91	3.34	2.40
4.00	9.85	78.77	11.38	2.81					3.62	2.42
4.60	9.81	90.19	0		4.00	9.04	72.34	18.62	3.18	2.44
					4.60	9.81	90.19	0	2.90	2.38
										2.54
										2.52
										2.94
2500°K Burner Diam = 0.188 in.					2500°K Burner Diam = 0.188 in.					
1.00	12.61	25.22	62.16	2.50	1.00	9.17	18.33	72.50	5.02	2.42
1.50	12.04	36.14	51.82	3.31	1.50	9.07	27.21	63.72	6.09	3.09
				3.34					5.76	3.13
2.00	12.13	48.52	39.35	4.08	2.00	9.61	38.45	51.94	5.57	3.50
2.50	12.24	61.18	26.58	4.13	2.50	10.31	51.55	38.14	5.36	3.94
				4.31	3.00	11.12	66.74	22.14	5.17	3.94
3.00	12.31	73.88	13.80	4.60					5.03	3.62
				4.52						3.75
3.58	12.25	87.75	0	4.56						
				4.37						

* These values were obtained from flames showing a large degree of instability and are of doubtful validity.

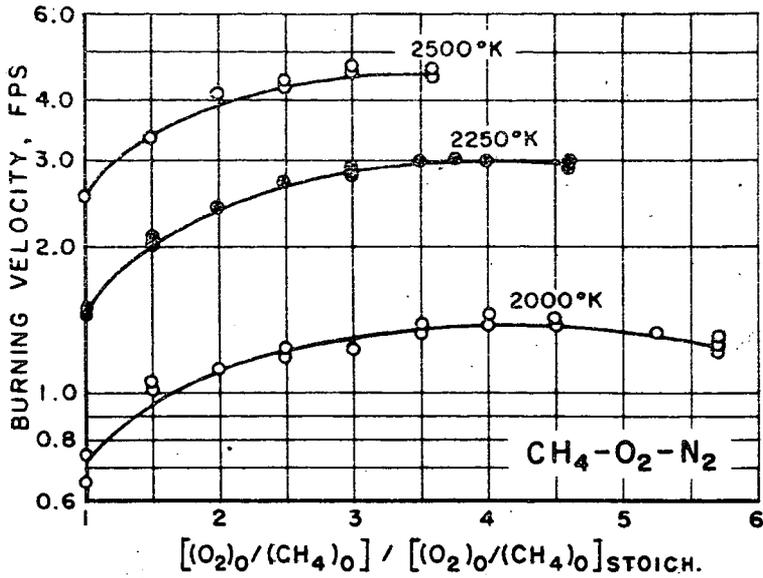


Fig. 1.-BURNING VELOCITIES OF THE METHANE-OXYGEN-NITROGEN SYSTEM AT ATMOSPHERIC PRESSURE AND THEORETICAL FLAME TEMPERATURES OF 2000°, 2250° AND 2500°K

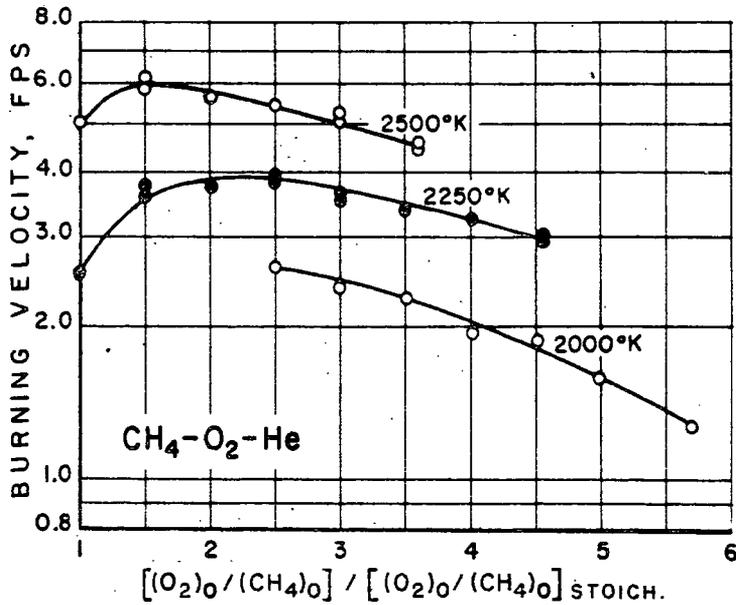


Fig. 2.-BURNING VELOCITIES OF THE METHANE-OXYGEN-HELIUM SYSTEM AT ATMOSPHERIC PRESSURE AND THEORETICAL FLAME TEMPERATURES OF 2000°, 2250° AND 2500°K

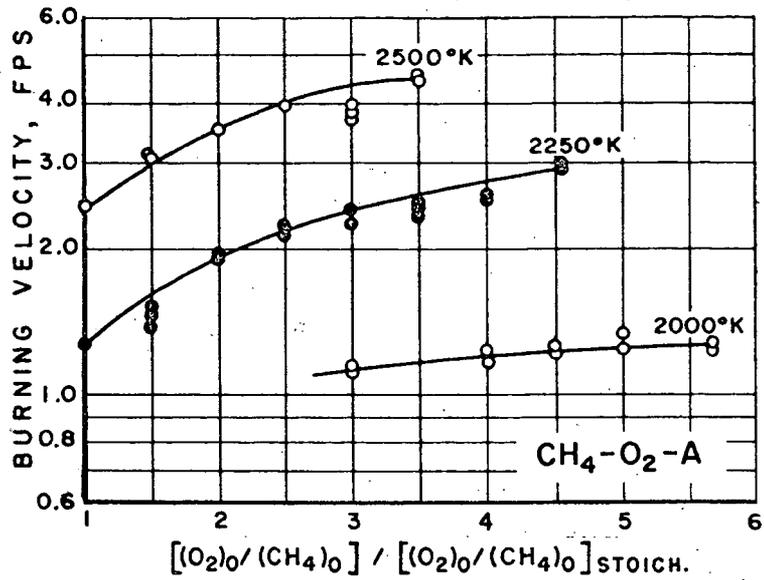


Fig. 3.-BURNING VELOCITIES OF THE METHANE-OXYGEN-ARGON SYSTEM AT ATMOSPHERIC PRESSURE AND THEORETICAL FLAME TEMPERATURES OF 2000°, 2250° AND 2500°K

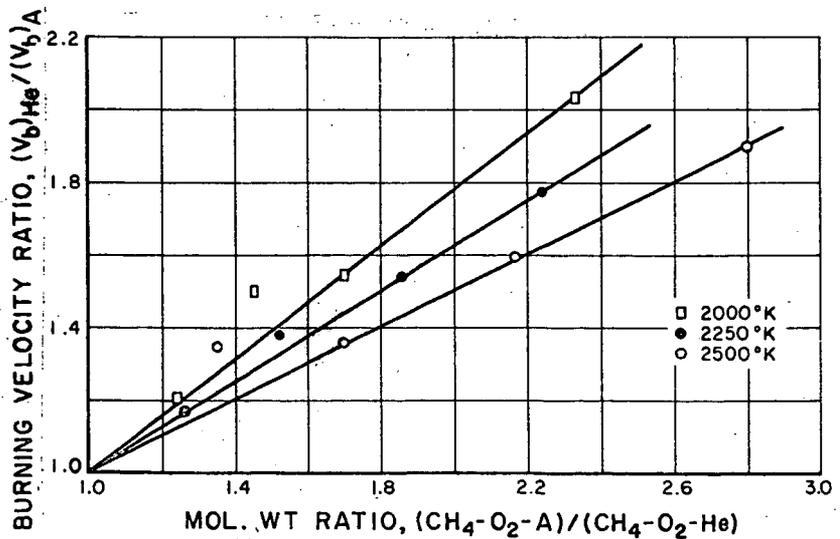


Fig. 4.-COMPARISON OF BURNING VELOCITIES OF METHANE-OXYGEN-HELIUM AND METHANE-OXYGEN-ARGON SYSTEMS AT THE SAME THEORETICAL FLAME TEMPERATURE AND OXYGEN/FUEL MOLE RATIO

DISCUSSION

To develop a method for the prediction of burning velocities based on the properties of the primary gases, it is necessary to determine "effective" values of the flame properties in terms of the primary gas properties. An obvious initial attempt was to correlate the effect of the diluent in terms of its effect on the transport properties. If it is assumed that the chemical processes are independent of the diluent, comparison of the helium and argon systems offers the most direct level to the contribution of transport phenomena. For this type of investigation, the simplest method of computing the transport properties of the mixtures can be used. Therefore, the reciprocal of the mean molecular weight was used as a measure of transport properties. In Fig. 4, where results are compared at the same temperature and O_2 /fuel ratio, distinct curves for the different temperature levels appear. Superficially none of the properties of the primary gases except those affecting the transport properties differ between the two systems being compared and one would expect no temperature dependence if this simple treatment were correct. The separation of these curves may be traced to the oversimplification in the use of density alone, or it may be attributed to differences in the microscopic description of the flame front. Since something in the nature of a mean or integrated transport property is desired, such differences in the internal flame structure may be sufficient to prevent the use of the simplest expressions for the transport property. However, unless the variation of the transport properties through the flame is considered, using the greatest rigor possible, there is little justification for a more complex expression. Thus, an empirical expression for prediction of burning velocity appears adequate.

A simple empirical correlation showing the effect of diluent is shown in Figs. 5 and 6 where the systems of different diluents are compared at the same theoretical flame temperature and oxygen-fuel ratio. The equations represented by the lines in these figures are:

$$\frac{V_{b,He}}{V_{b,A}} = \exp (0.0859 M_{He}) \quad (1)$$

$$\frac{\left[\frac{V_b}{(CH_4)_0} \right]_{He}}{\left[\frac{V_b}{(CH_4)_0} \right]_{N_2}} = \exp (0.0930 M_{He}) \quad (2)$$

At the present time, no theoretical explanation is offered to account for the interdependence of $\log V_b$ and diluent/fuel ratio at constant O_2 /fuel ratio and temperature. However, since the diluent content accounts for the major variation in transport properties, some form of dependence on the diluent content is to be expected. The particular form of that dependence, shown in Figs. 5 and 6, must be considered as the expression of a mean or effective evaluation of the effect of transport properties. Some contribution to the particular form obtained probably comes from certain mathematical relationships among the reactant compositions, resulting from the temperature calculations. Thus for any given temperature level at a given oxygen-to-fuel ratio, the following approximate relations hold in the fuel-lean region:

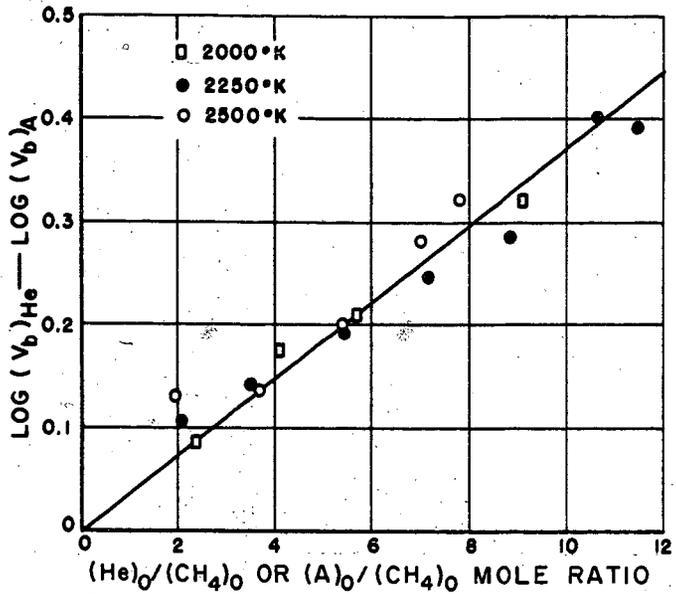


Fig. 5.-CORRELATION OF BURNING VELOCITIES OF THE METHANE-OXYGEN-HELIUM AND METHANE-OXYGEN-ARGON SYSTEMS AT ATMOSPHERIC PRESSURE AND THEORETICAL FLAME TEMPERATURES OF 2000°, 2250° AND 2500°K

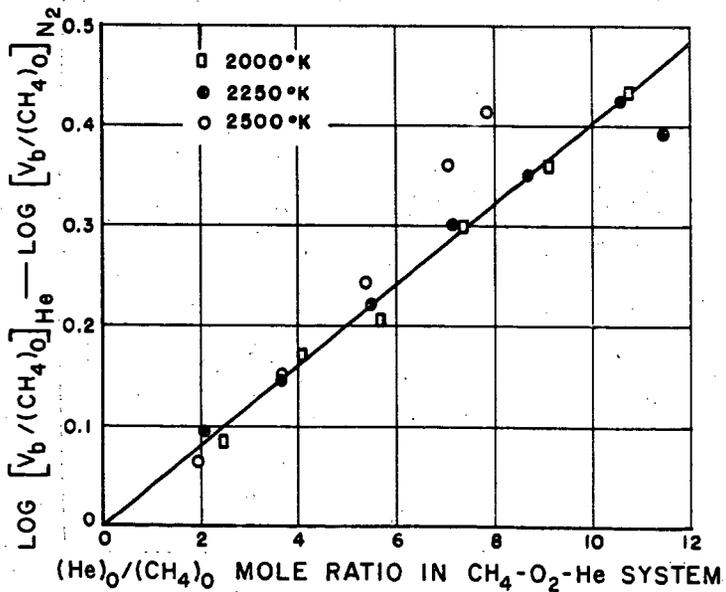


Fig. 6.-CORRELATION OF BURNING VELOCITIES OF THE METHANE-OXYGEN-NITROGEN AND METHANE-OXYGEN-HELIUM SYSTEMS AT ATMOSPHERIC PRESSURE AND THEORETICAL FLAME TEMPERATURES OF 2000°, 2250° AND 2500°K

$$M_{\text{He}} = M_A \quad (3)$$

$$M_{\text{He}} = 1.69 M_{\text{N}_2} \quad (4)$$

$$M_{\text{He}} = 1.71 (M_{\text{O}_2} - M_{\text{O}_2}^0) \quad (5)$$

Since $M_{\text{O}_2}^0$ is a function of temperature only, any empirical expression involving M_T cannot be distinguished from one involving a linear combination of M_T and M_{O_2} . The proportionality between the logarithm of the burning velocity and the diluent/fuel ratio, therefore, can also be expressed as one between $\log V_b$ and a linear combination of M_T and M_{O_2} . In this manner, the proposed effective transport term can be converted to one involving all components of the reactant mixture, which might be preferable. However, no particular linear combination was indicated and, therefore, the simplest expression was used.

To predict the dependence of burning velocity on reactant composition, an expression must also be found to describe the effect of oxygen/fuel ratio shown in Fig. 1. In the nitrogen system, there is little change in transport properties, and the system should be dependent only on the chemical effects of changing reactant composition. Examination of the curves of Fig. 1 indicates that they may be superimposed through the use of a proportionality constant. That is, the functional dependence of burning velocity on the oxygen/fuel ratio appears independent of temperature over the range investigated, which includes those usually encountered in fuel-air flames. Thus, an expression for the burning velocities of the systems investigated can be obtained which consists of only three factors. One is temperature dependent; one expresses the dependence on the oxygen/fuel ratio; and the last expresses the effect of transport properties in terms of the inert diluent. However, because only two of the three factors are independent, the mathematical expression relating the three factors must be arbitrary.

The following expression with four empirical parameters was used to describe the burning velocity of the $\text{CH}_4\text{-O}_2\text{-N}_2$ system:

$$\begin{aligned} [V_{b, \text{N}_2}]^2 &= [-1.75 + 1.99 M_{\text{O}_2} - 0.119 (M_{\text{O}_2})^2] \times 10^5 \\ &\times \left[\exp(-50.6 \times 10^3 / RT_f) \right] \end{aligned} \quad (6)$$

This equation can be used to describe systems of the other diluents, by means of Equations 1 and 2. To avoid the appearance of the CH_4 concentration in the N_2 system when applying the general equation to another diluent, it is necessary to introduce the approximate relationship:

$$(\text{CH}_4)_0 = 3.26 \times 10^{-7} (T_f - T_0)^{5/3} \quad (7)$$

for the systems containing nitrogen as diluent. This relationship was obtained by fitting a curve to the reactant methane concentration-flame temperature points in the theoretical flame temperature calculations. Combining Equations 1, 2, 6, and 7 yields an expression capable of describing the data of Figs. 1, 2, and 3.

$$\begin{aligned}
 [V_{b,I}]^2 &= 0.982 \left[(O_2)_O - 0.934(CH_4)_O \right] \left[(CH_4)_O - 0.0634(O_2)_O \right] \\
 &\times 10^{10} \times \left[\frac{T_O}{T_f - T_O} \right]^{10/3} \left[\exp(-50.6 \times 10^3 / RT_f) \right] \left[\exp(\alpha_I M_I) \right]
 \end{aligned}
 \tag{8}$$

with the values

$$\begin{aligned}
 \alpha_{He} &= 0.186 \\
 \alpha_A &= 0.0142 \\
 \alpha_{N_2} &= 0.0
 \end{aligned}$$

Similarities between the empirical Equation 8 and one proposed by Semenov⁸ in his approximate thermal theory offer the possibility of viewing the empirical equation in terms of "effective" quantities in a manner analogous to that used by Semenov. From this view, 50.6 Kcal is the effective activation energy of the overall reaction. This can be compared to the value of 51 Kcals used in other investigations.² The terms $(O_2)_O - 0.934(CH_4)_O$ and $(CH_4)_O - 0.0634(O_2)_O$ can be viewed as the effective oxygen and fuel concentrations, respectively, in the reaction zone which permit description of the reaction as being a single step reaction between those components. As presented above, the term $\exp(\alpha_I M_I)$ represents an effective transport term.

Another interesting aspect of Equation 8 is that, on extrapolation to lower temperatures, it predicts the existence of flammability limits, i.e. $V_b = 0$, when $(O_2)_O = 0.934(CH_4)_O$ (the upper limit) and when $(CH_4)_O = 0.0634(O_2)_O$ (the lower limit). These agree in order of magnitude with the observed values.¹

A more tenuous interpretation of the effective oxygen and fuel concentration terms is that they correspond to quantities controlling or determining the slow step of the reaction path. For example, the term $(O_2)_O - 0.934(CH_4)_O$ corresponds to the remaining oxygen after the oxidation of the fuel to formaldehyde and water or their chemical equivalent. An equally simple evaluation of the effective fuel term was not obtained.

CONCLUSIONS

In the use of burning velocity studies to add to the knowledge of the combustion process, it is necessary to know the manner in which burning velocity depends upon fuel gas composition as a function of stoichiometry, transport properties and flame temperature. When a single oxidant, such as air, is employed to study the variation of burning velocity with fuel-to-air ratio, flame temperature is a variable and overshadows all other factors. To minimize the effect of temperature, the concentration of the components of CH_4-O_2 -diluent systems were adjusted in this investigation to give flames of varying equivalence ratio but constant adiabatic reaction of 2000, 2250 and 2500°K. To the extent that the actual flame temperature can be identified with these theoretical flame temperatures, the process is then being studied under the same thermal conditions.

In the $\text{CH}_4\text{-O}_2\text{-N}_2$ system at the 2000°K temperature level; the burning velocity apparently passes through a maximum with increasing feed gas oxygen/methane ratio. Since the occurrence of this maximum is not predicted by the approximate theories of combustion which assume the rate-determining step to be the reaction between O_2 and CH_4 ,⁸ or that between the free radicals, H, OH, and O, and the fuel,¹⁰ an empirical correlation was developed relating the burning velocity to the composition of the feed gas and the flame temperature. This relationship describes all three isotherms and the effect of the diluents N_2 , He and A. It contains five experimental parameters, four of which are independent of the inert gas, and indicates, 1) the existence of mixtures for which $V_b = 0$, that is, flammability limits, which are of the correct order of magnitude, and 2) an energy of activation of $50.6 \text{ Kcal/moles-}^\circ\text{K}$ for the methane flames, which is in good agreement with that found by others.² The only term showing dependence on the inert gas is assumed to represent the effect of variation in transport properties.

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NOMENCLATURE

$(N)_O$	= mole fraction of component in burner gas mixture.
M_N	= $(N)_O / (\text{CH}_4)_O$
$M_{\text{O}_2}^O$	= $(\text{O}_2)_O / (\text{CH}_4)_O$ for the lean, diluent-free mixture.
$V_{b,I}$	= Experimental burning velocity (ft/sec) of ternary system $\text{CH}_4\text{-O}_2\text{-I}$.
R	= $1.986 \text{ cal/mole-}^\circ\text{K}$
T_O	= Initial temperature (300°K)
T_f	= Calculated adiabatic reaction temperature
α_I	= Empirically evaluated constant for the ternary system $\text{CH}_4\text{-O}_2\text{-I}$

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