

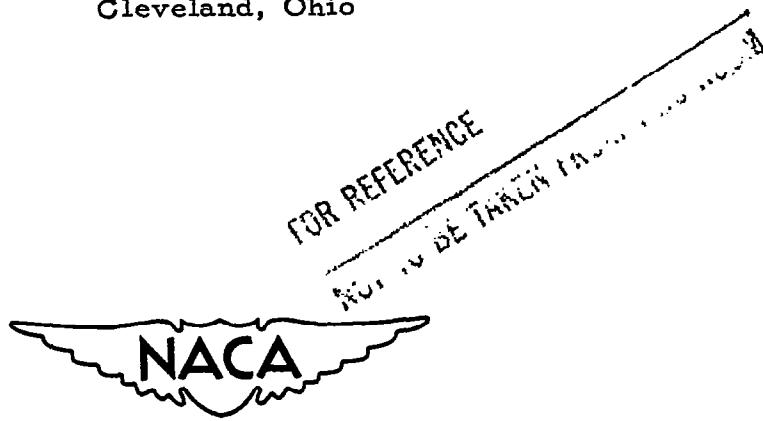
NATIONAL ADVISORY COMMITTEE FOR AERONAUTICS

TECHNICAL NOTE 3882

A RELATION BETWEEN BURNING VELOCITY AND QUENCHING DISTANCE

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SUMMARY

A thermal quenching equation and the Semenov burning velocity equation were combined to give expressions for the product and quotient of burning velocity and quenching distance.

The equation for the product was tested using published data. Experimental results for hydrocarbon-oxygen-inert gas, hydrocarbon-nitrous oxide, hydrogen-air, hydrogen-bromine, and the ethylene oxide decomposition flames were fairly well correlated by the product equation. Hydrocarbon-nitric oxide and ethylene oxide decomposition data were not correlated. It was concluded that the product is proportional to a term which has the character of a thermal diffusivity and a term which increases with the activation energy of the flame reaction. The quotient of burning velocity and quenching distance is proportional to the average flame reaction rate and to terms mildly dependent on the flame reaction activation energy and the transport properties. Average and maximum reaction rates and maximum space heating rates were calculated for several flames using the quotient equation and published data. The calculated maximum space heating rate for isoctane was about one and a half times the experimental value.

INTRODUCTION

Burning velocity and quenching distance are closely related. Several hypotheses of flame quenching relate the two (refs. 1 to 3). For example, reference 1 gives the following equation derived from the thermal considerations:

$$Ud = \alpha \left[\frac{1}{f} \frac{(T_f - T_i)}{(T_i - T_o)} \right]^{1/2} \quad (1)$$

The product of burning velocity U and quenching distance d for a given tube geometry is directly proportional to the thermal diffusivity α and a term related to the chemical properties of the flame (the ignition temperature T_i is considered a chemical property). For flames with similar

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thermal diffusivities and ignition temperatures (hydrocarbon-air flames, e.g.) the product Ud should be a constant. Experiments show this to be true (ref. 4). However, if one wishes to use equation (1) for comparison of flames having different chemical properties, the problem of definition of the ignition temperature arises. Equation (1) was derived from a simple concept of flame quenching and the Mallard-LeChatelier burning-velocity equation. A better relation of U to d can be derived using more sophisticated quenching-distance and burning-velocity equations. Thus, an equation for Ud was derived in reference 3 by combining a diffusional quenching-distance equation with a diffusional burning-velocity equation. Only a limited test of the equation was made because of the lack of suitable data. In a similar way, the thermal quenching equation (ref. 5) can be combined with the Semenov burning-velocity equation. The equations obtained in this way do not include an ignition temperature. However, a temperature T_q which is the minimum temperature to which a flame can be cooled before it is quenched does appear. This temperature is not equivalent to the usual experimental ignition temperature, which is the temperature at which unburned gas spontaneously ignites.

Two combinations are interesting, the product Ud and the quotient U/d . In this paper, expressions for these combinations are discussed and tested using published experimental data.

SYMBOLS

- A Arrhenius pre-exponential term, $(\text{molecules/cc})^{1-m}/\text{sec}$
- a fuel concentration, molecules/cc
- b oxidant concentration, molecules/cc
- c_p specific heat at constant pressure, cal/(g)(deg)
- \bar{c}_p average specific heat at constant pressure, cal/(g)(deg)
- d quenching distance, cm
- E activation energy, cal/mole
- F fraction of heat produced by flame which must be retained in order for flame to exist
- f dimensionless geometrical constant
- G dimensionless geometrical constant

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k thermal conductivity, cal/(cm)(deg)
 m apparent order of flame reaction
 n order of flame reaction with respect to oxidant
 R gas constant, cal/(mole)(deg)
 T temperature, $^{\circ}\text{K}$
 U burning velocity, cm/sec
 w flame reaction rate, fuel molecules/(cc)(sec)

$$\bar{w} \text{ average flame reaction rate, } \frac{\int_{T_0}^{T_f} w \, dT}{\frac{T_f - T_0}{T_f}}, \text{ fuel molecules/(cc)(sec)}$$

α thermal diffusivity, $k/\rho c_p$, cm^2/sec
 α^* apparent thermal diffusivity, $\left(\frac{k_f k_q}{\rho_o^2 c_{p,q} \bar{c}_{p,f}} \right)^{1/2}$, cm^2/sec

ρ density, g/cc

Subscripts:

f flame
 i ignition
 l lean limit
 o initial
 q quenched

THEORY

Product of Quenching Distance and Burning Velocity

The thermal quenching equation of reference 5 can be written as

$$d^2 = \frac{FGk_q a_o}{\rho_o c_{p,q} \bar{w}_q} \quad (2)$$

The subscript r , referring to the reaction zone, was previously used. Since the concept of reaction zone is not strictly necessary for derivation of equation (2), the more general subscript q , referring to the "about-to-be quenched" flame, is used here.

The Semenov burning-velocity equation (ref. 6) can be written as

$$U^2 = \frac{2k_f \bar{w}_f}{s_0 p_0 c_p} \quad (3)$$

Two terms which normally appear in this equation are not included here. These are the ratio of heat and mass diffusivities and the ratio of moles of product to moles of reactant. These factors have been omitted from equation (3) because it is not known how they would appear in the quenching equation. This omission is quite justifiable for flames in which these factors are close to unity (this includes most flames), but must be kept in mind when discussing flames in which they differ greatly from unity.

Equations (2) and (3) can be multiplied to give

$$U_d = \alpha^* \left(2GF \frac{\bar{w}_f}{\bar{w}_q} \right)^{1/2} \quad (4)$$

The term α^* is called the apparent thermal diffusivity and is defined in the symbol list. The term G is the geometrical constant. The factor F (fraction of the total heat of combustion that must be retained by the flame for it to exist) determines the temperature of the "about-to-be quenched" flame, which in turn determines \bar{w}_q , the reaction rate in the "about-to-be quenched" flame. The value of \bar{w}_q relative to \bar{w}_f is fixed not only by F , but also by the activation energy of the reaction. For a given F value, a reaction having a small activation energy gives a smaller ratio \bar{w}_f/\bar{w}_q than one having a large activation energy. In general, the product U_d (for a given tube geometry) is directly proportional to a thermal diffusivity and to a term involving the chemical nature of the flame. The close similarity of this equation to equation (1) is thus apparent.

Calculation of apparent thermal diffusivity. - All the quantities which appear in the apparent thermal diffusivity α^* are known or can be easily calculated except the thermal conductivities. The assumptions and methods used for their calculation are given in the appendix. For consistency with the original derivation of the thermal quenching equation (ref. 5), the heat capacity and thermal conductivity in the quenching equation both refer to unburned gas at 0.7 flame temperature. Because of the vague picture used for that derivation, it is by no means certain that these are referred to the correct conditions. Only a better understanding of the flame quenching can decide this question.

Calculation of $\bar{F}\bar{w}_f/\bar{w}_q$. - In order to calculate $\bar{F}\bar{w}_f/\bar{w}_q$, it is assumed that Semenov's formulation of the average reaction rate is adequate for computing \bar{w} , and that it applies to both the free and quenched flame. Semenov's formulation of the average reaction rate for stoichiometric flames for zero, first, and second order reactions can be obtained from reference 6. Generalizing these to the case of a fuel-oxygen reaction of m^{th} order gives

$$\bar{w}_f = A a_o^{m-n} b_o^n \left[\frac{RT_o T_f}{E(T_f - T_o)} \right]^m \frac{RT_f^2}{E(T_f - T_o)} e^{-\frac{E}{RT_f}} \quad (5)$$

This generalization was made in order to permit the use of a fractional reaction order if necessary. Equation (5) is correct for zero, first, and second orders, and consequently can be expected to be approximately correct for fractional orders close to zero, one, and two.

From reference 5, (assuming that T_q is the average flame temperature in the "about-to-be quenched" flame, and that heat capacity is constant)

$$F = (T_q - T_o)/(T_f - T_o) \quad (6)$$

Combining equations (5) and (6) and an expression for \bar{w}_q analogous to equation (5), and assuming A to be independent of temperature,

$$\frac{\bar{w}_f}{\bar{w}_q} = \left(\frac{T_q - T_o}{T_f - T_o} \right)^{2+m} \left(\frac{T_f}{T_q} \right)^{2+m} e^{-\frac{E}{R} \left(\frac{1}{T_f} - \frac{1}{T_q} \right)} \quad (7)$$

From equation (7), it can be seen that values of the flame activation energy are required to calculate $\bar{F}\bar{w}_f/\bar{w}_q$. Now the activation energy obtained from burning-velocity data depends somewhat on the assumptions made concerning the reaction order and the temperature dependence of the Arrhenius constant. The best procedure appears to be to use a consistent set of activation energies. Assuming the Arrhenius constant to be temperature independent (as done in the derivation of eq. (7)) and a bi-molecular reaction, the author of reference 7 found that the low-temperature activation energy for propane oxidation of 38 kilocalories per mole was a satisfactory value for propane-air flames. The authors of reference 8 found that the activation energy was nearly the same for most hydrocarbon-air flames. On the basis of these results, an activation energy of 38 kilocalories per mole was used for all hydrocarbon-oxygen-inert gas flames. For some of the other flames, no activation energies

are available. However, the activation energy for hydrocarbon-air flames is directly proportional to the lean-limit flame temperature (ref. 8). Activation energies were calculated by this method for hydrogen-air, hydrocarbon-nitric oxide, and hydrocarbon-nitrous oxide flames, using lean-limit flame temperatures for hydrogen-air, pentane-nitric oxide, and methane-nitrous oxide flames. The lean-limit flame temperatures were multiplied by 24 to make them consistent with a value of 38 kilocalories per mole for propane-air flames. Low-temperature activation energies were used for the hydrogen-bromine and ethylene oxide decomposition flames.

Another quantity necessary to calculate \bar{Fw}_f/\bar{w}_q is the reaction order m . Values of m are not available for most flames. Fortunately, \bar{Fw}_f/\bar{w}_q is quite insensitive to the value chosen for m . Since the order is close to two for most of the flames discussed herein, the order was taken to be two for all flames except the ethylene oxide decomposition flame, for which an order of one was assumed.

The minimum temperature to which a flame can be cooled and continue to exist is T_q . In order to determine T_q from equation (6), the value of F must be known. For lack of any other information, it will be assumed that F is the same for all flames. It is found that this assumption is a good one. Using the set of activation energies just discussed, a suitable value (found by trial and error comparison of experiment and theory) is 0.78. If the activation energies are decreased by 30 percent to bring them in line with the values given in reference 8, the best value for F drops to about 0.70.

Some values of $(\bar{Fw}_f/\bar{w}_q)^{1/2}$ for several different flames along with the activation energies used are given in table I.

Once a value for F has been chosen (0.78 in this case), the size of $(\bar{Fw}_f/\bar{w}_q)^{1/2}$ is determined solely by the size of the activation energy relative to the flame temperature. This is clearly shown in table I. For flames with similar activation energies and flame temperatures, very little difference is seen in the $(\bar{Fw}_f/\bar{w}_q)^{1/2}$ values. For these flames, one can write equation (4) as

$$Ud \propto \alpha^* \quad (8)$$

Sources of experimental data. - The values of quenching distance and burning velocity used for a test of equation (4) are shown in table II. Quenching distance is fairly reproducible, so that quenching data were taken directly from published results. All quenching distances are for plane parallel plates. When necessary, quenching data for cylinders were converted to plane parallel plates by multiplying them by 0.66

(ref. 9). Burning velocities vary according to the method of measurement, so the data were arbitrarily adjusted to a common basis whenever possible. Sets of data were chosen which contained a value for propane-air flames. These data were multiplied by a factor to make the stoichiometric propane burning velocity equal to 40 centimeters per second at 25° C. Data from references 10 and 11 were adjusted to a common basis through the burning velocities given for benzene-air flames. Stoichiometric burning velocities were used when available; otherwise, maximum values were used. Temperature exponents of burning velocity were assumed to be 1.7 for hexane and hexene-1 flames, and to be 2 for hydrogen and propane with argon and helium "air". For hydrogen-bromine flames, the burning velocity on the 1.5-centimeter burner was used rather than that for the 1.0-centimeter burner. The values used for the quenching distance and burning velocity of ethylene oxide are believed to be the best available.

Comparison of theory and experiment. - The test of equation (4) is shown in figure 1, where the observed product U_d is plotted against the calculated product for the 30 flames given in table II. Generally, hydrocarbon-oxygen-inert gas flames are well correlated, although flames at elevated initial temperatures show a trend away from a mean line. It seems likely that the assumption of a constant F value is incorrect for these flames. The significance of this is not clear at this time. Several other flame types are well correlated; these are hydrocarbon-nitrous oxide, hydrogen-air, and hydrogen-bromine flames. The hydrocarbon-nitric oxide and ethylene oxide decomposition flames are not correlated. These flames present considerable experimental difficulty, and this may explain the discrepancy.

From these results, one can conclude that the product U_d is directly proportional to a term having the nature of a thermal diffusivity and a term that increases with increasing activation energy of the flame reaction.

Empirical relation between U and d . - Occasionally, the relation between U and d for chemically similar flames, such as hydrocarbon-oxygen-nitrogen flames, is of interest. According to equation (8), for systems such as this, the product is directly proportional to the apparent thermal diffusivity. From the definition of the apparent thermal diffusivity, it can be seen that this quantity varies directly as the initial temperature, inversely as the pressure, and directly as flame temperature to a power describing the temperature dependence of the thermal conductivities and heat capacities. By comparison with experiment, it is found that a good value for the flame temperature exponent is 1. A plot of U_d for hydrocarbon-oxygen-nitrogen flames at 1 atmosphere against $T_o T_f$ is shown in figure 2. By least squares, it is found that the relation has the following form:

$$U_d = 1.204 \times 10^{-5} T_o T_f \quad (9)$$

Quotient of Burning Velocity and Quenching Distance

By dividing equation (3) by equation (4) and rearranging, one obtains

$$\frac{U}{d} = \frac{\bar{w}_f}{a_0} \sqrt{\left(\frac{\bar{w}_q}{F\bar{w}_f}\right) \left(\frac{2k_f \bar{c}_{p,q}}{Gk_q \bar{c}_{p,f}}\right)} \quad (10)$$

The terms under the square root sign do not vary much from flame to flame. The reciprocal of the first of these, $F\bar{w}_f/\bar{w}_q$, is discussed previously, and it was found to be a mild function of activation energy. The other term contains the ratios of thermal conductivity and heat capacity in the free and "about-to-be quenched" flame. This term can be expected to be nearly constant. Its value depends largely on the assumptions made about the temperature and composition of the "about-to-be quenched" flame. Thus, for similar flames (e.g., hydrocarbon-oxygen-inert gas flames),

$$\frac{U}{d} \propto \frac{\bar{w}_f}{a_0} \quad (11)$$

Using the assumptions previously made concerning $F\bar{w}_f/\bar{w}_q$, it is possible to use equation (10) to calculate average reaction rates from experimental burning velocities and quenching distances. This has been done for some representative flames, and the results are shown in table III. The rates are quite sensitive to the choice of F ; a 3-percent change in F changes the calculated rate about 10 percent. To compare the flames on a common basis, the average rate calculated from the data was corrected to an initial temperature of 25° C using equation (5). Two values of particular interest are those for propane with argon "air" and with helium "air". Since replacement of argon by helium does not affect flame temperature, it is often assumed that reaction rates in the two flames are identical. The reaction rates calculated from equation (10) differ considerably for the two flames. Since two experimental results are combined in the quotient, the difference may be experimental error. Other evidence that a real difference may exist is the fact that the pressure exponents of quenching distance (related to the order of the reaction) are different for the two flames (ref. 12). This seeming difference could mean that an important step in the complex chain reaction of the combustion process is a three-body reaction involving the inert gas.

The maximum reaction rate attained in the flame is frequently of interest. Provided the flame reaction can be approximated by a simple one-step reaction of the kind treated by Semenov, the maximum rate can be found from the average rate. Again generalizing from equations in reference 6, the reaction rate in the flame is

$$w = A a_o^{m-n} b_o^n \left(\frac{T_o}{T}\right)^m \left(\frac{T_f - T}{T_f - T_o}\right)^m e^{-\frac{E}{RT}} \quad (12)$$

The temperature corresponding to the maximum rate can be obtained by differentiation of equation (12) with respect to temperature and setting the result equal to zero. This optimum temperature is

$$T_{opt} = \frac{T_f}{\frac{mRT_f}{1 + \frac{E}{R}}} \quad (13)$$

To find the relation of the maximum reaction rate to the average rate, equations (5), (12), and (13) are combined. Then,

$$w_{max} = \bar{w}_f \frac{E}{R} \frac{(T_f - T_o)}{T_f^2} m^m e^{-m} \quad (14)$$

Values for w_{max} for some representative flames are given in table III, along with maximum space heating rates (derived from w_{max} by multiplying it by the heat of combustion of the fuel). These maximum space heating rates may be valuable for comparison of different fuels. They are not absolute values because of the many approximations introduced. By way of comparison, Longwell's value of the space heating rate of isoctane-air flames at an initial temperature of 300° K and 1 atmosphere is 2.1×10^8 Btu per cubic foot per hour (ref. 13), and the calculated value for the same initial conditions is 3.6×10^8 Btu per cubic foot per hour. The values of reaction order m used are given in table III. They were calculated from the pressure dependence of quenching distance using the procedures of reference 14, and are corrected for the effect of pressure on flame temperature. The references cited in table III are the sources of the pressure exponents.

CONCLUSIONS

Combination of a thermal quenching equation

$$\text{Quenching distance} \propto \left(\frac{\text{Transport property}}{\text{Reaction rate}} \right)^{1/2}$$

and a burning velocity equation

$$\text{Burning velocity} \propto [(\text{Reaction rate})(\text{Transport property})]^{1/2}$$

gives the following expressions for the product and quotient of burning velocity and quenching distance: The product is

$$(\text{Burning velocity})(\text{Quenching distance}) \propto \text{Transport property}$$

The quotient is

$$\frac{(\text{Burning velocity})}{(\text{Quenching distance})} \propto \boxed{\text{Reaction rate}} \quad f(t)$$

In practice these statements must be modified somewhat, since reaction rates and transport properties differ in the free and quenched flames. However, if similar flames (e.g., hydrocarbon-oxygen-inert gas flames) are compared, the proportionalities are correct as stated.

Lewis Flight Propulsion Laboratory
 National Advisory Committee for Aeronautics
 Cleveland, Ohio, August 15, 1956

$$QD = \frac{R_u u_B}{R_i R_e}$$

$$\frac{u_B}{R_i R_e} = \frac{\text{reaction rate}}{C^2} = C^2 \boxed{K.P.R}$$

$$\frac{u_B}{R_i R_e} = \frac{u_B}{C^2} = \frac{u_B}{d}$$

$$C^2 = \frac{u_B}{d}$$

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APPENDIX - CALCULATION OF FLAME TEMPERATURE, THERMAL
CONDUCTIVITY AND HEAT CAPACITY

Flame temperatures for the hydrocarbon-air flames at an intial temperature of 378° K were calculated from the tables given in reference 15. All other flame temperatures are values taken from the references in which burning velocity is given.

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Thermal conductivities for the Lennard-Jones (6-12) potential were calculated for individual components of the mixtures for which conductivity values were desired, using the procedures and data given in chapter 8 of reference 16. Mixture conductivities were calculated by the methods described in reference 17. The viscosity equation given by Hirschfelder (ref. 16) was used in the Lindsay-Bromley mixing rule rather than the Sutherland equation. For mixtures containing water vapor, the procedure given on page 600 of reference 16 was used to obtain force constants for use in Hirschfelder's viscosity equation.

For k_q values, the conductivity of the unburned mixture at $0.7 T_f$ was calculated. Since heat is lost to the wall through unburned mixture, this seems qualitatively reasonable.

For k_f values, the conductivity of the burned gases at T_f was calculated. For simplicity, dissociation was ignored, and k_f was calculated for the simple products of reaction at T_f .

Heat capacities were calculated from the tables of reference 18. For $c_{p,q}$, the heat capacity of the unburned mixture at $0.7 T_f$ was calculated. For $c_{p,f}$, the average heat capacity of the undissociated products from initial temperature to flame temperature was calculated.

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TABLE I. - VALUES OF $(F\bar{w}_f/\bar{w}_q)^{1/2}$ FOR SEVERAL FLAMES

Flame system	$(F\bar{w}_f/\bar{w}_q)^{1/2}$	Activation energy, E, kcal/mole	Flame tempera- ture, T_f , °K
Propane-air ^a	2.33	38	2289
Isooctane-air	2.32	↓	2302
Methane-oxygen	2.06	↓	3020
Hydrogen-air	1.69	23	2380
Ethane-nitrous oxide	1.80	29	2740
Ethane-nitric oxide	2.69	49	2840
Hydrogen-bromine ^b	4.32	40.2	1490
Ethylene oxide decomposition	8.56	52.7	1217

^aInitial temperature T_o , 378° K.^bEquivalence ratio, 2/3.

TABLE II. - BURNING VELOCITY, QUENCHING DISTANCE, AND ACTIVATION ENERGY
FOR VARIOUS STOICHIOMETRIC FLAMES

Flame system	Initial temperature, T_0 , °K	Burning velocity, U , cm/sec	Reference for burning velocity	Quenching distance, d , cm	Reference for quenching distance	Activation energy, E , kcal	Reference for activation energy	Flame temperature, T_f , °K
Propane-air	378	a58.3	7	0.184	14	38	7	2289
n-Hexane-air		a57.3	10	.182		b38		2300
n-Decane-air		a55.9	11	.183				2303
Isooctane-air		a44.5	19	.233				2302
Hexene-1-air		a65.1	10	.186				2324
Decene-1-air		a60.6	11	.175				2322
Benzene-air		a63.6		.171				2365
n-Butyl benzene-air		a53.5		.203				2339
Propane-17% oxygen-nitrogen	313	a20.3	19	.377	20			2051
Propane-21% oxygen-nitrogen		a42.3		.199				2253
Propane-50% oxygen-nitrogen		a91.8		.087				2558
Propane-50% oxygen-nitrogen		a202		.051				2844
Propane-70% oxygen-nitrogen		a312		.032				2970
Propane-argon "air"		a90.1	21	.104	12			2557
Propane-helium "air"		a158		.253	12			2557
Propane-air	400	a64.1	7	.183	22			2313
Propane-air	483	a89.0		.159				2351
Propane-air	558	a115.5		.144				2384
Ethylene-air	298	a69.9	10	.125	3			2362
Methane-air		a32.8	23	.25	24			2214
Methane-oxygen		a344		.03				3020
Hydrogen-air	313	a216		.0495	14	c24	8	2380
Acetylene-nitrous oxide	298	160	4	.056	4	c29	25	2940
Ethylene-nitrous oxide		120		.048				2820
Ethane-nitrous oxide		90		.059				2740
Acetylene-nitric oxide		85		.24				3080
Ethylene-nitric oxide		40		.44		c49	26	2920
Ethane-nitric oxide		23		.86				2840
Hydrogen(60%)-bromine(40%)	323	28	27	.59	27	40.2	28	1490
Ethylene oxide decomposition	298	2.7	29	4.6	30	52.7	29	1217

^aValues adjusted as described in the text.

^bEstimated.

^cCalculated from lean-limit flame temperature T_l ; $E = 24T_l$.

TABLE III. - REACTION RATES AND MAXIMUM SPACE HEATING RATES

Flame system	Initial temperature, T_0 , $^{\circ}\text{C}$	Order of reaction, n	Average reaction rate, \bar{w} , moles/sec	Maximum reaction rate, w_{\max} , moles/sec	Maximum space heating rate at 1 atm, Btu/(cu ft)(hr)
Hydrogen-air	25	^a 2.17	169.	463.	108×10^8
Methane-air		^b 1.60	1.60	5.07	3.9×10^8
Ethylene-air		^c 1.60	5.00	15.1	19.3×10^8
Propane-air		^a 1.60	1.04	3.27	6.44×10^8
Propane - argon "air"		^d 1.71	3.82	11.3	22.4×10^8
Propane - helium "air"		^d 1.63	2.94	8.40	16.6×10^8
Isooctane-air		^a 1.52	.24	.72	3.6×10^8

^aRef. 14.^bEstimated.^cRef. 3.^dRef. 12.

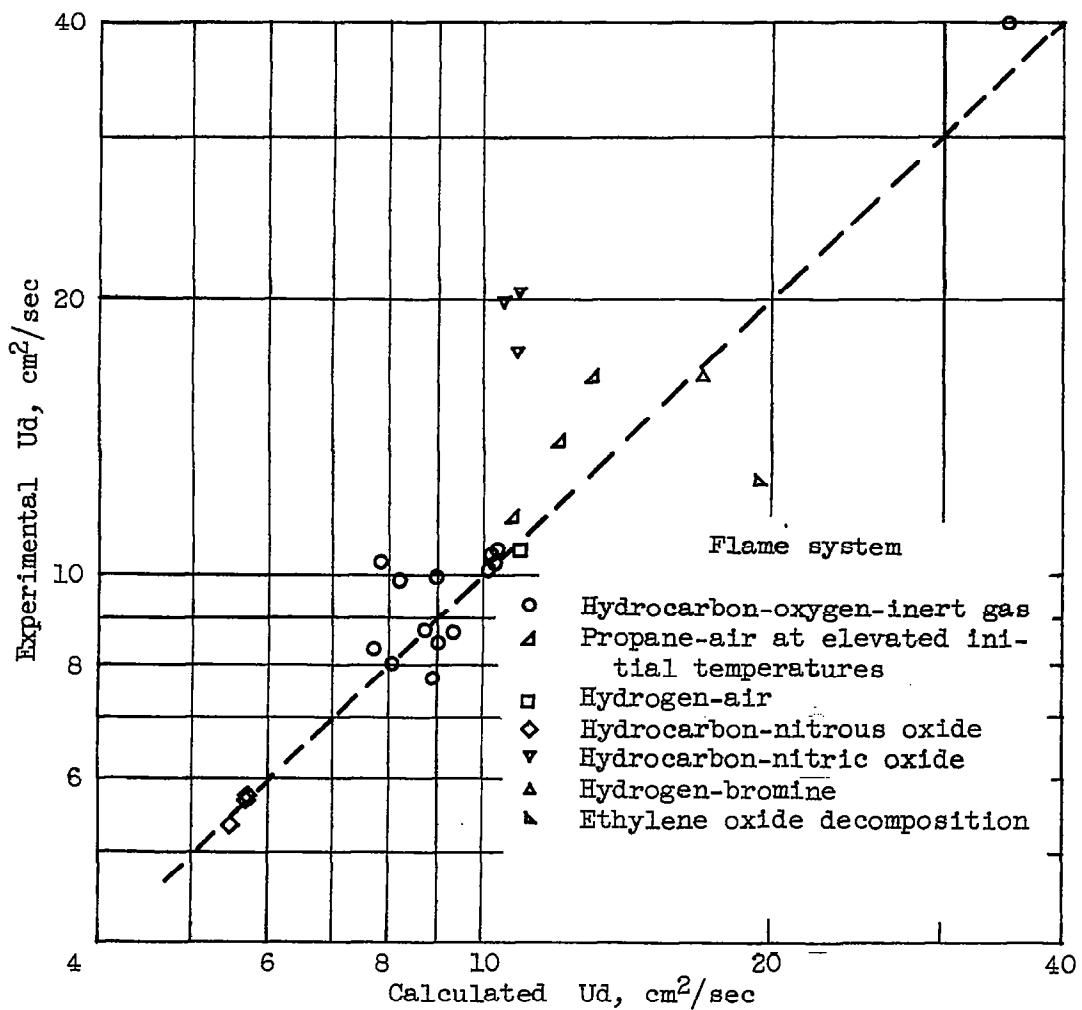


Figure 1. - Comparison of experimental and calculated values of burning-velocity - quenching-distance product U_d .

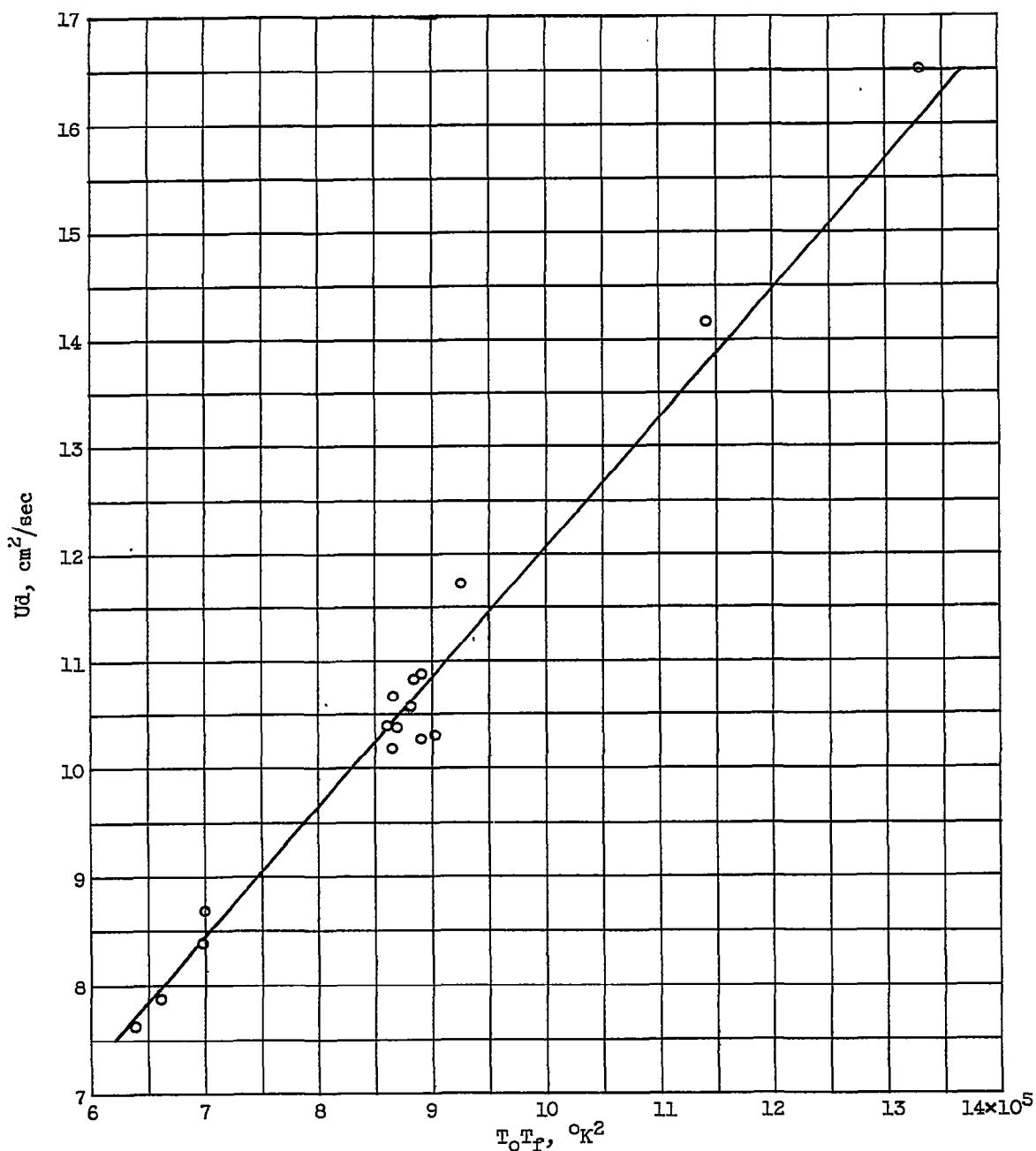


Figure 2. - Relation of empirical burning-velocity - quenching-distance product U_d to empirical temperature product $T_o T_f$ for hydrocarbon-oxygen-nitrogen flames.