



NATIONAL ADVISORY COMMITTEE FOR AERONAUTICS

TECHNICAL NOTE 3882

A RELATION BETWEEN BURNING VELOCITY AND QUENCHING DISTANCE

By A. E. Potter, Jr. and A. L. Berlad

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. Hydrocarbonnitric 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 isooctane 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:

 $Ud = \alpha \left[\frac{1}{f} \frac{(T_{f} - T_{i})}{(T_{i} - T_{o})} \right]^{1/2}$ (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_1 is considered a chemical property). For flames with similar

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 burningvelocity 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_d which is the minimum temperature to which a flame can be cooled before it is guenched 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

А	Arrhenius	pre-exponential	term, (molecules	/cc) ^{⊥-m}	/sec
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- a fuel concentration, molecules/cc
- b oxidant concentration, molecules/cc
- c_p specific heat at constant pressure, cal/(g)(deg)
- $\overline{c_p}$ 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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m apparent order of flame reaction

n order of flame reaction with respect to oxidant

R gas constant, cal/(mole)(deg)

T temperature, ^OK

- U burning velocity, cm/sec
- w flame reaction rate, fuel molecules/(cc)(sec)

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

$$\alpha \quad \text{thermal diffusivity, k/pc_p, cm^2/sec}$$

a* apparent thermal diffusivity,
$$\left(\frac{k_{f}k_{q}}{\rho_{0}^{2}c_{p},q^{\overline{c}}p,f}\right)^{1/2}$$
, cm²/sec

$$\rho$$
 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

$$d^{2} = \frac{FGk_{q}a_{o}}{\rho_{o}c_{p}, q^{\overline{w}}q}$$
(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 \vec{q} , referring to the "about-to-be quenched" flame, is used here.

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

$$U^{2} = \frac{2k_{f}\overline{w}_{f}}{a_{O}\rho_{O}\overline{c}_{p}}$$
(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

$$Ud = \alpha^* \left(2GF \frac{\overline{w}_f}{\overline{w}_q} \right)^{1/2}$$
 (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 "aboutto-be quenched" flame, which in turn determines \overline{w}_q , the reaction rate in the "about-to-be quenched" flame. The value of \overline{w}_q relative to \overline{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 $\overline{w}_f/\overline{w}_q$ than one having a large activation energy. In general, the product Ud (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 at 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. オーーキ

Calculation of Fw_f/w_q . - In order to calculate Fw_f/w_q , it is assumed that Semenov's formulation of the average reaction rate is adequate for computing \overline{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 mth order gives

$$\overline{w}_{f} = Aa_{o}^{m-n}b_{o}^{n} \left[\frac{RT_{o}T_{f}}{E(T_{f} - T_{o})} \right]^{m} \frac{RT_{f}^{2}}{E(T_{f} - T_{o})} e^{-\frac{E}{RT_{f}}}$$
(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)

$$F = (T_{q} - T_{o}) / (T_{f} - T_{o})$$
(6)

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

$$F \frac{\overline{w}_{f}}{\overline{w}_{q}} = \left(\frac{T_{q} - T_{o}}{T_{f} - T_{o}}\right)^{2+m} \left(\frac{T_{f}}{T_{q}}\right)^{2+m} e$$
(7)

From equation (7), it can be seen that values of the flame activation energy are required to calculate $F\overline{w}_f/\overline{w}_q$. 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 bimolecular reaction, the author of reference 7 found that the lowtemperature 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-oxygeninert gas flames. For some of the other flames, no activation energies

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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 $F\overline{w_f}/\overline{w_q}$ is the reaction order m. Values of m are not available for most flames. Fortunately, $F\overline{w_f}/\overline{w_q}$ 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 $(F\overline{w}_f/\overline{w}_q)^{1/2}$ 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 $(F\overline{w}_{f}/\overline{w}_{q})^{1/2}$ 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 $(F\overline{w}_{f}/\overline{w}_{q})^{1/2}$ values. For these flames, one can write equation (4) as

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

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(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 1.0centimeter burner. The values used for the quenching distance and burning velocity of ethylene oxide are believed to be the best available.

<u>Comparison of theory and experiment.</u> - The test of equation (4) is shown in figure 1, where the observed product Ud 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 hydrocarbonnitrous oxide, hydrogen-air, and hydrogen-bromine flames. The hydrocarbonnitric 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 Ud 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 hydrocarbonoxygen-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 Ud for hydrocarbon-oxygen-nitrogen flames at 1 atmosphere against T_0T_f is shown in figure 2. By least squares, it is found that the relation has the following form:

$$Ud = 1.204 \times 10^{-5} T_0 T_f$$
 (9)

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Quotient of Burning Velocity and Quenching Distance

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

$$\frac{U}{d} = \frac{\overline{w}_{f}}{a_{O}} \sqrt{\left(\frac{\overline{w}_{q}}{F\overline{w}_{f}}\right) \left(\frac{2k_{f}\overline{c}_{p,q}}{Gk_{q}\overline{c}_{p,f}}\right)}$$
(10)

The terms under the square root sign do not vary much from flame to flame. The reciprocal of the first of these, $F\overline{w}_f/\overline{w}_q$, 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),

$$\frac{\mathbf{U}}{\mathbf{d}} \propto \frac{\mathbf{\overline{w}}_{\mathbf{f}}}{\mathbf{a}_{O}}$$
 (11)

Using the assumptions previously made concerning FW_{f}/W_{a} , 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

$$w = Aa_{o}^{m-n}b_{o}^{n} \left(\frac{T_{o}}{T}\right)^{m} \left(\frac{T_{f} - T}{T_{f} - T_{o}}\right)^{m} e^{-\frac{E}{RT}}$$
(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

$$T_{opt} = \frac{T_{f}}{\frac{mRT_{f}}{1 + \frac{-f}{E}}}$$
(13)

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

$$w_{\max} = \overline{w}_{f} \frac{E}{R} \frac{(T_{f} - T_{o})}{T_{f}^{2}} m^{m} e^{-m}$$
(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 isooctaneair flames at an initial temperature of 300° K and 1 atmosphere is 2.1×10⁸ 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. H Que

CONCLUSIONS

Combination of a thermal quenching equation

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

and a burning velocity equation

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

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

(Burning velocity) (Quenching distance) ~ Transport property

The quotient is

f(t)(Burning velocity) ∝Reaction rate. (Quenching distance)

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

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.

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

TABLE I. - VALUES OF $(F\overline{w}_f/\overline{w}_q)^{1/2}$ for several flames

^aInitial temperature T_0 , 378^o K. ^bEquivalence ratio, 2/3.

TABLE II. - BURNING VELOCITY, QUENCHING DISTANCE, AND ACTIVATION ENERGY

FOR VARIOUS STOICHIOMETRIC FLAMES

Flame system	Initial temper- ature, T _O , ^O K	Burning velocity, U, cm/sec	Refer- ence for burning velocity	Quenching distance, d, cm	Reference for quenching distance	Activa- tion energy, E, kcal	Refer- ence for activa- tion energy	Flame tempera- ture, T _f , K
Propane-air	378	⁸ 58.3	7	0,184	14	38	7	2289
n-Hexane-air		a57.3	10	.182		^b 38		2300
n-Decane-air		^a 55,9	11	.183				2303
Isooctane-air		844.5	19	.233				2302
Hexene-1-air		865,1	10	.166				2324
Decene-1-air		8-60.6	11	.175		·		2322
Benzene-air		863,6		.171				2365
n-Butyl benzene-air	1	^a 53.5		.203	[†			2339
Propane-17% oxygen-mitrogen	313	820.3	19	.377	20		1	2051
Propane-21% oxygen-nitrogen		^B 42.3	1	.199				2253
Propane-30% oxygen-nitrogen		^a 91.8		.087				2558
Propane-50% oxygen-nitrogen		a202		.051				2844
Propane-70% oxygen-nitrogen		⁸ 312		.032	1 🛉			2970
Propane-argon "air"		⁸ 90.1	21	.104	12	.		2557
Propane-helium "air"	1 1	a 158	┝╴╁	.253	12			2557
Propane-air	400	⁸ 64.1	þ 7	.183	22			2313
Propane-air	483	⁸ 89.0	1	.159				2351
Propane-air	568	^a 115.5		.144	J 🕴			2384
Ethylene-air	298	⁸ 69.9	10	,125	3			2362
Methane-air		^a 32.8	23	.25	24			2214 ₁
Methane-oxygen	1 1	8344		.03]			3020
Hydrogen-air	313	⁸ 216	1	.0495	114	°24	8	2380
Acetylene-nitrous oxide	298	160	4	.036	4	c29	25	2940
Ethylene-nitrous oxide		120		.048	• I			2820
Ethane-nitrous oxide	·	90		.059	*		ľ 🕴	2740
Acetylene-nitric oxide		85		.24		°49	26	3080
Ethylene-nitric oxide		40		.44	[f. Ï	I Ï	2920
Ethane-nitric oxide		23	1	.86	1 1			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

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^aValues adjusted as described in the text. ^bEstimated.

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^cCalculated from lean-limit flame temperature T_1 ; E = 24 T_1 .

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TABLE III. - REACTION RATES AND MAXIMUM SPACE HEATING RATES

Flame system	Initial temper- ature, T ₀ , °C	Order of reaction, m	Average reaction rate, \overline{w} , moles/sec	Maximum reaction rate, Wmax, moles/sec	Maximum space heating rate at l atm, Btu/(cu ft)(hr)
Hydrogen-air	25	^a 2.17	169.	463.	108x10 ⁸
Methane-air		b1.60	1.60	5.07	3.9x10 ⁸
Ethylene-air		c1.60	5.00	15.1	19.3x10 ⁸
Propane-air		al.60	1.04	3.27	6.44x10 ⁸
Propane - argon "air"		d1.71	3.82	11.3	22.4×10 ⁸
Propane - helium "air"		đ1.63	2,94	8.40	16.6×10 ⁸
Isooctane-air	ł	al.52	.24	.72	3.6×10 ⁸

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a_{Ref}. 14.

^bEstimated. ^cRef. 3.

dRef. 12.







Figure 2. - Relation of empirical burning-velocity - quenching-distance product Ud to empirical temperature product $T_0 T_f$ for hydrocarbon-oxygen-nitrogen flames.

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