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RESEARCH MEMORANDUM

FLAME VELOCITIES OF PROPANE- AND
ETHYLENE-OXYGEN-NITROGEN
MIXTURES

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NATIONAL ADVISORY COMMITTEE
FOR AERONAUTICS

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The laminar flame velocities of propane and ethylene with various oxygen-nitrogen mixtures were determined from schlieren photographs of Bunsen-type flames. Flame velocity was determined as a function of equivalence ratio for each oxygen-nitrogen mixture at two initial temperatures, 311° and 422° K. The mole fraction of oxygen in the oxygen-nitrogen mixture was varied from 0.166 to 0.496 for propane and from 0.166 to 0.349 for ethylene. The maximum flame velocity with respect to equivalence ratio was found to vary linearly with the mole fraction of oxygen \( \alpha \) for each initial temperature \( T_0 \) (°K). With the assumption of a common \( \alpha \)-intercept at zero flame velocity for both initial temperatures, empirical equations including the effect of \( \alpha \) and of \( T_0 \) were obtained.

The experimental results for the effect of \( \alpha \) are predicted on a relative basis, within approximately 5 to 15 percent, by approximate theoretical equations based on either a thermal mechanism, as presented by Semenov, or an active-particle diffusion mechanism, as presented by Tanford and Pease. Both theories predict a decreasing rate of change in maximum flame velocity with increasing oxygen concentration rather than the linear relation between \( U_{\text{max}} \) and \( \alpha \) observed experimentally. Neither the sum of the effective active-particle concentrations nor the square root of this sum is related linearly to maximum flame velocity for the entire range of \( \alpha \) investigated.

INTRODUCTION

A study of the effect of oxygen concentration on the flame velocity of isooctane-oxygen-nitrogen mixtures showed that flame velocity increased linearly with the mole fraction of oxygen in the oxidant mixture \( O_2/(O_2 + N_2) \) over the range from 0.210 to 0.496 (reference 1). This linear relation was shown to be at variance with the predictions of the approximate solutions of two theoretical equations, one based on a thermal and the other based on a diffusion mechanism of propagation, either
of which predicted a decreasing rate of change of flame velocity with increasing oxygen concentration. It was desirable to know whether the observed linear augmentation of flame velocity would also occur with other fuels, and whether the trends predicted by the theoretical equations would again differ from those established experimentally. Further, data on the effects of the variables equivalence ratio and initial temperature have been obtained for propane- and ethylene-air mixtures (reference 2), and it was thought desirable to obtain data on the effect of oxygen concentration for these same fuels.

For these reasons, flame-velocity data were obtained for propane- and ethylene-oxygen-nitrogen mixtures for which \( \frac{O_2}{(O_2 + N_2)} \) varied from 0.166 to 0.496 and from 0.166 to 0.349, respectively. Flame velocity was determined as a function of equivalence ratio at each \( \frac{O_2}{(O_2 + N_2)} \) at both 311° and 422° K. Measurements were taken from schlieren photographs of the Bunsen cone, and calculations were made by the total-area method. The maximum flame velocities (with respect to equivalence ratio) are compared with the values predicted by the Semenov bimolecular equation (thermal mechanism, reference 3) and the Tanford-Pease equation (diffusion mechanism, reference 4) on a relative basis.

SYMBOLS

The following symbols are used in this report:

- \( A, B, C \) empirical constants
- \( A_F \) axial cross-sectional area of flame cone, \( \text{cm}^2 \)
- \( a \) fuel concentration, molecules/cm³
- \( B_i \) term near unity arising from recombination of \( i \)th radical
- \( b \) oxygen concentration, molecules/cm³
- \( C_p \) molar heat capacity at constant pressure, \( \text{cal/(mole)(°K)} \)
- \( c_p \) specific heat, \( \text{cal/(g)(°K)} \)
- \( \bar{c}_p \) mean specific heat, \( T_0 \) to \( T_f \), \( \text{cal/(g)(°K)} \)
- \( D \) diffusion coefficient, \( \text{cm}^2/\text{sec} \)
- \( D_m \) diffusion coefficient at mean combustion zone temperature, \( \text{cm}^2/\text{sec} \)
activation energy, kcal/g-mole

base of Napierian logarithmic system raised to power in parenthesis following exp

height of cone, cm

weighted mean $k_4$ for active particles (H, OH, and O), each reacting with fuel molecules, cm$^3$/molecule/sec

specific rate constant for reaction between $i^{th}$ radical and combustible material, cm$^3$/molecule/sec

total concentration of gas at mean combustion zone temperature, molecules/cm$^3$

height of generating curve, cm

molecular weight

empirical constant

mole fraction

total number of molecules of H$_2$O and CO$_2$ in products per molecule of fuel by stoichiometric relation

moles of reactants per moles of products from stoichiometric equation

steric factor used herein as arbitrary constant, from $k = PZ \exp(-E/RT)$

total pressure, dynes/cm$^2$

partial pressure or mole fraction of $i^{th}$ active particle in burned gas

mole fraction of potential combustion product in unburned gas

mole fraction of combustible in unburned gas

gas constant, kcal/(g-mole)(°K) or ergs/(g-mole)(°K)
lateral surface area, cm$^2$

absolute temperature, °K
**U** flame velocity, cm/sec

**U_{\text{max}}** maximum flame velocity, varying \( \varphi \) at constant \( \alpha \) and \( T_0 \), cm/sec

**Z** collision number, molecular collisions/(sec)(one molecule of each type/cm\(^3\))

**\( \alpha \)** mole fraction of oxygen in oxygen-nitrogen mixture

**\( \alpha_x \)** empirical constant

**\( \eta \)** viscosity of mixture, poise

**\( \beta_m \)** ratio of mean combustion zone temperature to initial temperature

**\( \lambda \)** thermal conductivity, cal/(cm\(^2\))(sec)(°K/cm)

**\( \rho \)** density of mixture, g/cm\(^3\)

**\( \sigma \)** collision diameter, cm

**\( \varphi \)** equivalence ratio, fraction of stoichiometric fuel-oxygen ratio

**Subscripts:**

- **0** initial condition
- **1,2** reactants in bimolecular reaction
- **eff** effective mean reactant concentration
- **f** condition at flame temperature
- **i** \( i^{\text{th}} \) active particle
- **j** \( j^{\text{th}} \) component in mixture

**EXPERIMENTAL PROCEDURE**

**Apparatus.** - The apparatus used in this investigation is diagrammatically illustrated in figure 1. The experimental procedure was essentially the same as that reported in reference 1. The fuel and the oxygen-nitrogen mixture were metered through critical flow orifices I (fig. 1) and mixed at point K. The mixture passed through a small heated plenum chamber M and into a copper tube that passed through the oil jacket of the burner P and was connected to the base of the burner.
The mixture acquired the temperature of the circulating oil and was burned as a Bunsen-type flame at atmospheric pressure. The circulating oil, and hence the gas mixture, was maintained at either \(311^\circ\) or \(422^\circ\) K ±10 K by electric heaters.

The burner tube shown in figure 1 was a 120-centimeter length of brass tubing having an inside diameter of 1.256 centimeters. In order to avoid flashback of the higher percentage oxygen mixtures, an insert tube with a lip at the top was soft-soldered to the stationary tube. The insert tubes were 60 centimeters long and had inside diameters of 0.297, 0.467, 0.617, and 0.838 centimeter.

Optical system. - A two-mirror (z-tye) schlieren system (reference 5) with a horizontal knife edge was used. A 25-watt concentrated arc lamp served as the light source. The camera lens gave a 1:1 image size, and the exposure time was 1/50 second.

Flame-velocity measurement. - Flame velocities were determined from the schlieren photographs by the total-area method, wherein the average normal flame velocity is equal to the volume rate of flow of the unburned mixture divided by the surface area of the cone formed by the combustion zone. This surface area \(S\) was determined by the approximate relation for the cone-like surfaces of revolution:

\[
S = \pi A_P l/11
\]  

Measurements were based on the outside edge of the schlieren image. In order to aid in determining the maximum flame velocities, data scatter was reduced by the method described in reference 2 (an averaging method which diminishes errors in measurements for equation (1)); however, essentially the same results would have been obtained without this technique.

Fuels and oxygen-nitrogen mixtures. - The minimum purities claimed by the supplier of the propane and ethylene were 96 and 99.5 mole percent, respectively. Oxygen and nitrogen were obtained premixed and were analyzed by the supplier to ±0.1 mole percent. For the 21-percent mixture, compressed air containing approximately 0.4 mole percent water was used.

EXPERIMENTAL RESULTS

In figure 2(a), flame velocity is plotted against the equivalence ratio (fraction of stoichiometric fuel-oxygen ratio) for propane-oxygen-nitrogen mixtures for \(\alpha\) values from 0.166 to 0.496 at an initial temperature of \(311^\circ\) K and atmospheric pressure. Similar data are presented
for propane-oxygen-nitrogen mixtures at 422º K in figure 2(b) and for ethylene-oxygen-nitrogen mixtures over a range of a from 0.166 to 0.349 at each of the two temperatures in figures 2(c) and 2(d). These curves exhibit flame velocity maximums at equivalence ratios between 1.00 and 1.06 for propane mixtures and between 1.08 and 1.16 for ethylene mixtures. These maximum flame velocities are presented in table I along with those from some additional curves.

In figure 3, maximum flame velocities from figure 2 are plotted against a for initial temperatures of 311º and 422º K. Linear relations are observed for each fuel at each temperature; that is,

$$U_{\text{max}} = Aa - B$$

The constants A and B, determined for each temperature by the method of least squares, are presented in table II. The constants for isooctane (2,2,4-trimethylpentane) from reference 1 are included for comparison.

An empirical equation for each fuel including the effects of both initial temperature $T_0$ and a was derived by the method described in reference 1, on the assumption that the variation of the a-intercept with temperature would be negligible for a small temperature range:

$$U_{\text{max}} = C T_0^m (a - a_x)$$

The constants C and m were also determined by the method of least squares by forcing the linear equations of type (2) to go through $a_x$, the average intercept for the two temperatures; these constants are presented in table II. The maximum flame velocities computed from equations (2) and (3) are included in table I.

THEORETICAL PREDICTIONS

Thermal mechanism. - An approximate theoretical equation for flame velocity based primarily on a mechanism of heat conduction from the flame zone into the unburned gases is presented by Semenov in reference 3. For a bimolecular reaction between fuel molecules and oxygen molecules, the equation may be written in the form (see reference 3, pp. 31, 45, 48, and 49):

$$U = \sqrt{2 \lambda P Z_{\text{eff}} b_{\text{eff}} \over a_0 \rho_0 c_p (T_f - T_0) \left( \frac{\lambda}{c_p D \rho_f} \right)^2 \left( \frac{n_f}{n_p} \right)^2 \frac{RT_f^2}{E} \exp \left(- \frac{E}{RT_f} \right)}$$

$$U = \sqrt{2 \lambda P Z_{\text{eff}} b_{\text{eff}} \over a_0 \rho_0 c_p (T_f - T_0) \left( \frac{\lambda}{c_p D \rho_f} \right)^2 \left( \frac{n_f}{n_p} \right)^2 \frac{RT_f^2}{E} \exp \left(- \frac{E}{RT_f} \right)}$$
where, for stoichiometric or fuel-rich mixtures \((\Phi \geq 1)\), \(a_{\text{eff}}\) and \(b_{\text{eff}}\) are computed by:

\[
a_{\text{eff}} = a_0 \frac{T_0}{T_f} \left[ 1 - \frac{1}{\Phi} \left( 1 - \frac{RT_f^2}{T_f - T_0} \right) \right]
\]

\[
b_{\text{eff}} = b_0 \frac{T_0}{T_f} \frac{RT_f^2}{T_f - T_0}
\]

The term \(T_f\) was assumed to be the equilibrium flame temperature, which was calculated by the method of reference 6. Low-temperature activation energies of 38 kilocalories per mole for propane (reference 7) and 40 kilocalories per mole for ethylene (reference 8) were used. The transport properties were estimated by extrapolation from tables presented in references 6, 9, and 10 and by the relations:

\[
\lambda = (C_f + \frac{5}{4} R) \frac{n}{M} \quad \text{(reference 10)}
\]

\[
\eta = \sum_j \eta_j \bar{N}_j
\]

For combustion products, equation (7) gives values within 1 percent of those obtained by method of reference 11.

\[
C_p = \sum_j C_j N_j \quad \text{(8)}
\]

\[
D = 1.336 \frac{\eta}{\rho} \quad \text{(reference 12)}
\]

\[
\rho = \frac{p}{\rho R T} \quad \text{(10)}
\]

\[
z = \left( \frac{C_1 + C_2}{2} \right)^2 \sqrt{8 \pi R T \frac{M_1 + M_2}{M_1 M_2}} \quad \text{(reference 13)}
\]

The remaining factor \(P\), used herein as a semiempirical factor, is found by averaging the values calculated by substituting the experimental values of maximum flame velocity for a given fuel and temperature into equation (4). Therefore, the predicted flame velocities discussed hereinafter are actually relative values for particular fuels and temperatures.

The curves for \(U_{\text{max}}\) against \(\alpha\) predicted by this method are compared with the experimental lines in figure 4, and the individual points are given in table III. For propane, the mean deviation of the relative
predictions is approximately 17 percent at 311° K and 12 percent at 422° K; for ethylene, the mean deviation is approximately 4 percent at 311° K and 3 percent at 422° K. The predicted curves show a negative second derivative, \( \frac{d^2U_{\text{max}}}{da^2} < 0 \), just as the isoctane curves did in reference 1.

Diffusion theory. - The approximate equation given by Tanford and Pease (reference 4), which is based on the diffusion of chain carriers for the oxidation process (active particles) from the flame zone into the unburned gas, may be written

\[
U = \sqrt{\frac{L_m Q' n}{\sqrt{Q' m}}} \sum \frac{k_i P_i D_m}{B_i} \tag{12}
\]

For these calculations, the active particles are assumed to be hydrogen and oxygen atoms and hydroxyl radicals. The mean combustion-zone temperature is assumed to be 0.7 \( T_f \) (reference 4); \( T_f \) and the active-particle concentrations \( p_i \) are calculated by the method of reference 6 on the assumption of adiabatic thermal equilibrium. The diffusion coefficients \( D_m \) are calculated by use of the temperature dependence determined from equation (9) and reference values from the Stefan-Maxwell equation (reference 12). The recombination factor \( B_i \) is calculated by the method of Tanford (reference 14) for hydrogen atoms and is assumed to be unity for hydroxyl radicals and oxygen atoms. It was assumed that the specific rate constants for all three types of active particles could be represented by a weighted mean specific rate constant \( k_a \). By solving for \( k_a \) with the aid of experimental flame velocities in equation (12), it was found that \( k_a \) does not exhibit an Arrhenius-type temperature dependence, but is nearly constant. As a result, \( k_a \) has been used as an arbitrary constant much as \( P \) was in the treatment of the Semenov equation, and the predicted flame velocities are again relative values for a given fuel and temperature.

The flame velocity predictions obtained by this method are presented in figure 5 and table III. The mean deviations for both propane and ethylene are seen to be approximately 5 to 6 percent. All the predicted curves show a negative second derivative.

It is shown in reference 1 that for isoctane-oxygen-nitrogen mixtures no linear correlation between maximum flame velocity and either \( (6.5 \, P_H + P_{OH} + P_O) \) or the square root of this sum held for the entire range of \( \alpha \). Such correlations have been reported when flame velocity was varied by varying initial temperature (reference 2) or equivalence ratio (reference 15). The present data again show no linear correlations that hold for the entire experimental range as maximum flame velocity is varied by varying \( \alpha \); this is evidenced by the propane data presented in figure 6.
SUMMARY OF RESULTS

A study of the effect of oxygen concentration on the flame velocities of propane and ethylene at two initial temperatures and atmospheric pressure yielded the following results:

1. Maximum flame velocity $U_{\text{max}}$ varied linearly with the mole fraction of oxygen in the primary oxygen-nitrogen mixture $\alpha$, over the range 0.166 to 0.496 for propane and from 0.166 to 0.349 for ethylene, at both 311° and 422° K, as represented by

$$U_{\text{max}} = A\alpha - B$$

Empirical equations which also include the effect of initial temperature $T_0$ over the range from 311° to 422° K take the form:

$$U_{\text{max}} = C T_0^m (\alpha - \alpha_x)$$

The least-squares values of the constants $A$, $B$, $C$, $m$, and $\alpha_x$ for this equation are presented.

2. Subject to the approximations inherent in the theoretical equations and on the assumption that adiabatic thermal and chemical equilibria are attained at the end of the flame zone, it was found that:

a. Relative flame velocities predicted by the Semenov bimolecular equation with activation energies for low-temperature oxidation showed mean deviations from experimental flame velocities of the order of 5 percent for ethylene and 15 percent for propane.

b. With an average specific rate constant, which was assumed to be valid for hydrogen atoms, oxygen atoms, and hydroxyl radicals and to be independent of temperature, the Tanford-Pease equation predicted relative flame velocities with mean deviations from experimental flame velocities of the order of 5 percent for either fuel.

c. Both these equations predicted a decreasing rate of change of maximum flame velocity with increasing oxygen concentrations; this same behavior was previously reported for isooctane.
Neither the sum of the effective active particle concentrations nor the square root of this sum was found to be related linearly to maximum flame velocity for the entire range of \( \alpha \) investigated.

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National Advisory Committee for Aeronautics
Cleveland, Ohio

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<tr>
<th>Fuel</th>
<th>( \phi_2 )</th>
<th>( T_0 )</th>
<th>Initial mixture temperature, ( \text{K} )</th>
<th>Stream flow, ( \text{cm/hr} )</th>
<th>Tube Reynolds number</th>
<th>Maximum flame velocity, ( \text{cm/sec} )</th>
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TABLE II - EMPIRICAL EQUATIONS AND CONSTANTS FOR ISOOCTANE-, PROPANE-, AND ETHYLENE-OXYGEN-NITROGEN MIXTURES

<table>
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<tr>
<th>Fuel</th>
<th>Equation (2): $U_{\text{max}} = A\alpha - B$</th>
<th>Equation (3): $U_{\text{max}} = C T^m (\alpha - \alpha_x)$</th>
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<td>Constants</td>
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<td>$A$ at $311^\circ K$</td>
<td>$B$ at $311^\circ K$</td>
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<td>Isooctane</td>
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<td>Ethylene</td>
<td>882</td>
<td>103.8</td>
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*aReference 1.*
### Table III - Theoretically Predicted Values of Relative Maximum Flame Velocity of Propane- and Ethylene-Oxygen-Nitrogen Mixtures

<table>
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<tr>
<th>Fuel</th>
<th>$\alpha$</th>
<th>Initial mixture temperature, $T_0$ (°K)</th>
<th>Flame temperature, $T_f$ (°K)</th>
<th>Equivalence ratio $\varphi$</th>
<th>Equilibrium radical concentrations $\left(\text{mole fraction} \times 10^2\right)$</th>
<th>Maximum flame velocity (cm/sec)</th>
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<td>$P_H$, $P_{H_2}$, $P_{O_2}$</td>
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A Arc lamp for schlieren system
B Spherical mirror
C Burner port showing flame cone
D Aspirating thermocouple for port
E Oxygen-nitrogen mixture cylinder
F Pressure regulator
G Gage, 0-100 lb/sq in.
H Thermocouple
I Critical flow orifice
J Cil bath for oxidant
K Mixing tee
L Knife edge
M Cil-jacketed plenum chamber
N Camera
O Fuel cylinder
P Cil-jacketed burner tube
Q Gear pump
R Primary oil heater (three stage)
S Thermostat controller
T Secondary oil heater (three stage)
U Insert-tube lip soft-soldered to burner port
V Insert tube
W Spacing ring
X Flashback screen

Figure 1. - Diagrammatic sketch of experimental apparatus.
Figure 2. - Flame velocity as function of equivalence ratio over range of oxygen concentrations.

(a) Propane-oxygen-nitrogen mixtures at 311° K.
(b) Propane-oxygen-nitrogen mixtures at 422° K.

Figure 2. - Continued. Flame velocity as function of equivalence ratio over range of oxygen concentrations.
Mole fraction of oxygen

\[ \alpha = \frac{O_2}{O_2 + N_2} \]

0.347

0.296

0.250

0.210

0.166

(c) Ethylene-oxygen-nitrogen mixtures at 3110 K.

Figure 2. - Continued. Flame velocity as function of equivalence ratio over range of oxygen concentrations.
Mole fraction of oxygen
\[ \alpha = \frac{O_2}{O_2 + N_2} \]

C.349

Figure 2. - Concluded. Flame velocity as function of equivalence ratio over range of oxygen concentrations.
Figure 3. - Effect of oxygen concentration on maximum flame velocities of propane- and ethylene-oxygen-nitrogen mixtures at 311° and 422° K.

(a) Propane.
(b) Ethylene.
Mole fraction of oxygen, $a = \frac{c_2}{c_2 + N_2}$

(a) Propane.

(b) Ethylene.

Figure 4. - Comparison of curves predicted by Semenov bimolecular equation with experimental results.
Figure 5. Comparison of curves predicted by Tanford-Pease theory with experimental results.
Figure 6. - Relation between maximum flame velocity of propane-oxygen-nitrogen mixtures and effective active-particle concentrations.
Laminar flame velocities of propane and ethylene with oxygen-nitrogen mixtures containing 0.166 to 0.496 mole fraction of oxygen were determined as a function of mixture composition at 311^0 and 422^0 K by a Bunsen-burner method. For each case flame velocity increased linearly with oxygen concentration. Empirical equations including the effects of initial temperature and oxygen concentration were obtained. Thermal and diffusion theories were used to predict the relative effect of oxygen concentration on maximum flame velocity within 5 to 15 percent. No linear correlations between maximum flame velocity and calculated active particle concentrations were obtained. Copies obtainable from NACA, Washington.