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STABILITY OF PREMIXED
 $H_2/O_2/N_2$ COMBUSTING TURBULENT JETS

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Abstract

Experimental measurements and theoretical predictions of the stability of hydrogen-air-nitrogen premixed turbulent flames have been carried out. The turbulent flame is theoretically modeled using finite rate chemical kinetics and a well-stirred reactor. The model contains a free parameter, which is evaluated by comparing the theoretical predictions to experimental results. A hydrogen flame stability experiment was carried out and the results were compared to the theory. The theory and experiment were found to agree, if the free parameter in the theory varied as the reciprocal of the turbulent jet radius squared.

1. INTRODUCTION

Current investigations for improvement of the efficiency of combustion have involved not only the investigation of alternate fuels, but also new and different combustion techniques. Hydrogen has received much interest as an alternate fuel. It is considered to be the most environmentally compatible energy source for usage in the residential-commercial, industrial, utility and transportation sectors. In addition, the development of advanced combustion techniques such as: (1) stratified charge burning, (2) preheating the fuel-air mixture, and (3) combustion product gas recirculation has created a need for data on the stability of hydrogen combustion. Although the hydrogen-oxygen reaction scheme is one of the best understood combustion mechanisms, very little flame stability data for hydrogen-air combustion is currently available. The object of this study is to obtain experimental hydrogen combustion sta-

bility data and to compare it with a simplified flame stability theory.

2. EXPERIMENT

The fuel used in the experiment consisted of research grade hydrogen gas with 8.8 parts per million (ppm) of ethane, while the "air" consisted of a mixture oxygen (8.73% by volume) and nitrogen (91.27% by volume) which roughly represented a mixture of 1 part oxygen to 11 parts nitrogen. The gas was purchased from Matheson Gas Products, Joliet, Illinois.

The experimental setup is shown schematically in Figure 1. The gas flow passed through pressure regulators to reduce the pressure to acceptable line values, needle valves to roughly adjust the volume flow rates and rotameters (which also had needle valves associated with them to finely adjust the flow rates) to measure the volume flow rate. The rotameters were Matheson Gas Products models 604 for the O_2-N_2 mixture and 603 for the

H₂. Both rotameters had two floats (pyrex and stainless steel) to increase the flow rate measurement range and Matheson Gas Products calibration charts were used to interpret the readings. The flow then passed through pressure gages and over a thermocouple so that the pressure and temperature were known. These values were needed to correct the rotameter readings to the actual pressure and temperature of the flow. Check valves were inserted in the gas lines to prevent flow reversal in order to keep oxygen out of the hydrogen line and hydrogen out of the oxygen-nitrogen system. After passing the check valves, the gases were brought together in the opposite branches of a tee (opposed flow). This was done to encourage mixing. The mixing continued as the gases passed through the mixing tube, which consisted of a crimped 0.170 inch I.D. dia. copper tube. At the end of the mixing tube as the flow entered the burner, the pressure and temperature were again measured. The burner had an inside diameter of 0.1854 cm (0.0730 inch) and was 4.45 cm (1.75 inches) long. All the tubing in the system was 0.25 inch I.D. diameter plastic tubing. The experiment also required the measurement of the atmospheric temperature and pressure, to provide basic data on the entrained gas.

The experiment was carried out in a simple straight-forward manner. The "air" was turned on and adjusted to its desired flow rate. Then the hydrogen was turned on. As the hydrogen was turned on, a propane torch was inserted in the jet to ignite it. With the flame ignited, the hydrogen flow rate was adjusted to its desired value and the air flow rate was increased to cause the flame to blowout.

Blowout was determined by two methods: (1) visually and (2) acoustically. The ethane in the hydrogen gas increased the flame radiation in the visible region of the spectrum, so that the flame could be observed visually. The acoustic noise emitted from the turbulent jet changed pitch during the approach to blowout. At blowout, the pitch had a dramatic step-function change, so blowout could easily be detected acoustically.

3. THEORY

The turbulent flame is theoretically represented by a model involving finite rate chemical kinetics, in which the fluid mixing is replaced by a well-stirred reactor (WSR) as described by Nelson and Kushida⁽¹⁾. The flame is allowed to be located either at the exit of the burner, or in a stable lifted position above the burner. Its location is a function of the ambient atmospheric environment and of the mass flow rate, the preheat, and the equivalence ratio of the turbulent flow at the burner exit. If the flame is lifted, the composition and temperature of the ambient atmosphere also influence the flame stability through entrainment of atmospheric mass and energy.

The lifted flame is assumed to exit at the position where the reaction kinetics in the flame exactly balance the rate at which combustibles are supplied to the flame. If the mass flow rate of the jet is changed, the flame adjusts to a new position, such that the chemical rate and fuel/air supply rate exactly balance. The WSR is assumed to exist at this balance position. As the mass flow rate of the fuel/air jet increases, the chemical kinetic rate increases until it reaches its maximum value. Any further increase in the mass flow rate of the fuel/air jet will cause the flame to extinguish. The extinguishment occurs because the chemical kinetics cannot supply heat at a rate sufficient to ignite the incoming fuel/air mixture. This suggests that the maximum chemical kinetic rate and the rate at which fuel/air is supplied to the flame are balanced at blowout and that the overall fluid mechanics of the entire flow field are of secondary importance. Thus, the stability of turbulent flames can be modeled using a simplified flow field.

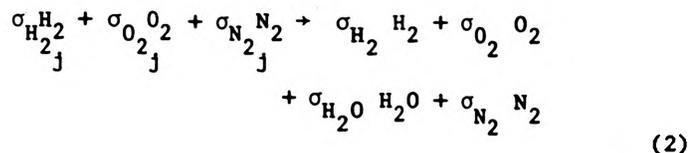
Turbulent mixing of the ambient atmosphere and the fuel/air jet is accounted for by the use of an entrainment parameter $\mu = \frac{m_s}{(m_s + m_o)}$, where m_s is the entrained mass flow rate and m_o the initial mass flow rate of the fuel/air jet. The profiles of the variables across the jet are assumed to be similar, so that the entrainment parameter becomes

the similarity parameter

$$\mu = \frac{u_o - u_j}{u_o} = \frac{H_j - H_o}{H_s - H_o} = \frac{\sigma_{x,j} - \sigma_{x,o}}{\sigma_{x,s} - \sigma_{x,o}} \quad (1)$$

Here u is the axial velocity, H is the total enthalpy per unit mass, and σ_x is the unreacted equivalent moles of species x per unit mass of mixture. The subscript j indicates a local jet value, the subscript o an initial jet value, and the subscript s an ambient atmospheric value. One essential property of μ is that it increases along the jet. In addition, the jet pressure and the ambient atmospheric pressure must balance, so that the jet and ambient densities are related by $\rho_j \bar{\sigma}_j T_j = \rho_s \bar{\sigma}_s T_s$, in which $\bar{\sigma}$ is the reciprocal of the average molecular weight and T the temperature.

The flame is modeled as a WSR of volume, V , into which a mixture of fuel (H_2), air (O_2, N_2) and entrained ambient atmosphere (O_2, N_2) flow at a mass flow rate of \dot{m}_j . The overall reaction is written as



The jet equivalence ratio ER_j , is equal to $\sigma_{H_2,j} / (2\sigma_{O_2,j})$, since two moles of oxygen are necessary to completely combust one mole of hydrogen. The concentrations with subscript j are those in the turbulent jet whose entrainment is proportional to μ according to Eq. (1). The concentrations σ_x are those that exist in the WSR, and they are defined as follows:

$$\sigma_{H_2} = \sigma_{H_2,j} - 2\epsilon y \quad (3a)$$

$$\sigma_{O_2} = \sigma_{O_2,j} - \epsilon y \quad (3b)$$

$$\sigma_{H_2O} = 2\epsilon y \quad (3c)$$

$$\sigma_{N_2} = \sigma_{N_2,j} \quad (3d)$$

where ϵ is the oxygen efficiency factor. For the fuel-lean case all the fuel is burned at $\epsilon = 1$;

therefore $y = (1/2)\sigma_{H_2,j}$ for $\sigma_{H_2,j} / \sigma_{O_2,j} \leq 2$. For the fuel-rich case all the oxygen is burned at $\epsilon = 1$; therefore $y = \sigma_{O_2,j}$ for $\sigma_{H_2,j} / \sigma_{O_2,j} > 2$.

Following previous practice for the analysis of hydrocarbon combustion in WSR's (Refs. 2-7), it is assumed that a one-step overall reaction expression adequately represents the mean combustion rate of hydrogen. The reaction rate equation is assumed to be

$$\frac{\dot{m}}{V} (\sigma_{H_2,j} - \sigma_{H_2}) = k (\sigma_{H_2})^{2-\alpha} (\sigma_{O_2})^\alpha \rho^2 \exp[-E/(RT)] \quad (4)$$

where the density in the WSR is defined by

$$\rho = P/(\bar{\sigma}RT) \quad (5)$$

and where R is the universal gas constant. The constants α , k , and E are determined by fitting Eq. (4) to experimental blowout data. In addition, the reciprocal of the average molecular weight is written as

$$\bar{\sigma} = \sigma_{H_2,j} + \sigma_{O_2,j} + \sigma_{N_2,j} - \epsilon y = \bar{\sigma}_j - \epsilon y \quad (6)$$

The temperature in the WSR is determined from the conservation of energy by a numerical iteration process. Assuming steady adiabatic flow the enthalpy entering the WSR at the jet temperature must equal the enthalpy of the gas in the WSR at the WSR temperature

$$H_j = \sum_{x=1}^N h_{x,j} \sigma_{x,j} = \sum_{x=1}^N h_x \sigma_x \quad (7)$$

in which the sum is over all species present. The h_x (calorie/gm-mole) are evaluated at the jet or WSR temperature T_j or T , respectively.

Equations (1) through (7) are sufficient to calculate the values of σ_x for all the x species under consideration, the temperature, T , and the density, ρ , in the WSR for any given value of the mixing parameter μ , and the combustion efficiency parameter ϵ .

In order to relate the combustion model to flame stability, it is necessary to consider the reaction rate (Eq. (4)) near blowout. The fluid mechanical

convection rate can be written as

$$\Lambda = \dot{m}/V. \quad (8)$$

The mass and momentum flow rates of the jet are defined as

$$\dot{m} = \int_{A_j} \rho u dA = \bar{\rho}_j \bar{u}_j \pi b^2 \quad (9)$$

and

$$J = \int_{A_j} \rho u^2 dA = \bar{\rho}_j \bar{u}_j^2 \pi b^2 = \bar{\rho}_o \bar{u}_o^2 \pi r_o^2 \quad (10)$$

in which the total momentum flux, J, is constant for a constant pressure jet. If it is assumed that l is a characteristic reactor length, the volume of the WSR becomes $V = \pi b^2 l$.

One can write the jet mass flow rate as

$$\dot{m} = \dot{m}_j = \dot{m}_o + \dot{m}_s. \quad (11)$$

The definition of the entrainment parameter allows one to write

$$\dot{m} = \dot{m}_o / (1-\mu). \quad (12)$$

Using the above definitions, Λ can be written as

$$\Lambda = \frac{\dot{m}_o}{(1-\mu) \pi b^2 l}. \quad (13)$$

Combining the mass and momentum equations (Eqns. (9) and (10)) to eliminate b one has

$$\bar{u}_j = (1-\mu) \bar{u}_o \quad (14a)$$

and

$$b^2 = \frac{r_o^2}{(1-\mu)^2} \frac{\bar{\rho}_o}{\bar{\rho}_j} \quad (14b)$$

If one assumes that the turbulence scale length l can be written as

$$l = \kappa r_o^3 / b^2 \quad (15)$$

where κ is an arbitrary constant, r_o is the radius of the jet at the burner exit and b is the equivalent uniform jet radius, Λ can be written as

$$\Lambda = \frac{\rho_o u_o}{(1-\mu) \kappa r_o} \quad (16)$$

This relationship for l implies that the WSR volume decreases with increasing distance along the jet, since b increases along the jet.

Equation (16) can be written in terms of u_o/d_o ,

where d_o is the initial jet diameter, as

$$\frac{u_o}{d_o} = \frac{(1-\mu) \Lambda \kappa}{2 \rho_o} \quad (17)$$

Equation (17) relates the flow issuing from the nozzle to chemical kinetics of the flame when Eqn. (4) is used for Λ .

$$\frac{u_o}{d_o} = \frac{\kappa(1-\mu) k(\sigma_{H_2})^{2-\alpha} (\sigma_{O_2})^\alpha \rho^2}{2 \rho_o (\sigma_{H_2} - \sigma_{H_2j})} \exp[-E/(RT)] \quad (18)$$

One parameter, the constant, κ , remains to adjust the theory and the experiments. The form of Eqn. (18) contains two implicit parameters, the entrainment parameter μ , and the oxygen efficiency factor ϵ , which must be optimized to produce a solution⁽¹⁾. The WSR representing the flame is assumed to be operating at the maximum reaction rate condition. The influence of preheat, entrainment, composition, etc. are all contained in the expression and they influence the value of u_o/d_o for blowout through μ and ϵ . It should be noted that no specific entrainment model has been used in the development of Eqn. (18).

The relation for u_o/d_o as given by Eqn. (18) contains three arbitrary constants E , the activation energy and k , the pre-exponential rate constant and the α the exponent of the oxygen concentration. None of the constants is well known for hydrogen combustion. The values of E , k and α were obtained by fitting the one-step overall rate equation to WSR blowout data available in the literature.

The one-step overall rate equation as given by Eqn. (4) can be written in terms of the kinetic loading parameter ψ , where

$$\psi = \sigma_{air} \dot{m} / (VP^2) = (1+m) \sigma_{O_2} \dot{m} / (VP^2) \quad (19)$$

and where m is the number of moles of nitrogen in the air per mole of oxygen and \dot{m}/V is given in Eqn. (4).

Clark et al⁽⁶⁾ have used the kinetic loading parameter to present experimental WSR blowout data for $m=11$. The general expression for ψ can be written

as

$$\psi = \frac{(1+m) \sigma_{O_2} k \left(\frac{\sigma_{H_2}}{\sigma_{H_2j} - \sigma_{H_2}} \right)^{2-\alpha} \left(\frac{\sigma_{O_2}}{\sigma_{O_2j} - \sigma_{O_2}} \right)^\alpha \rho^2 \exp[-E/(RT)]}{p^2} \frac{\text{gm-moles}}{\text{sec-liter-atm}^2} \quad (20)$$

Values of α and E were tried in Eqn. (20) by trial and error until a good fit of the experimental data curve shape was obtained. The value of k was determined by adjusting the magnitude of ψ agree with the experimental data. The results are shown in Figure 2. The best fit values are as follows $\alpha=1.2$, $E=25,000$ cal/(gm-mole), and $k=3.125 \times 10^{15}$ cm³/(gm-mole, sec).

4. RESULTS AND DISCUSSION

The experimental blowout results are tabulated in Table 1. It gives the equivalence ratio, fuel mass fraction, and fuel mole fraction at the burner exit and the value of U_0/d_0 at blowout. The rotameters had two balls so that the data labeled (p) represents the pyrex ball reading while the data labeled (ss) represents the stainless steel ball reading. For low flow rates, the pyrex ball is more accurate and for high flow rates, the stainless steel ball is the most accurate. For some of the data, both balls were on scale and could be used to determine the flow rates.

The theoretical blowout calculations for $\kappa=1$ are tabulated in Table 2. It gives the same parameters as presented in Table 1. The theoretical calculations are given for $\kappa=1$; however, they are linear in κ (see Eqn. 18) so the results given in Table 2 can be adjusted to any κ value by multiplying by the desired κ value.

The experimental and theoretical results are shown graphically in Figure 3. The experimental data obtained using the stainless steel ball is represented by the circles, while that representing the pyrex ball is represented by the squares. The theoretical curve is shown for $\kappa=6$. The agreement between theory and experiment appears to be reasonable. The theory contains a strong dependence on the amount of nitrogen at the flame base. For the

results reported herein, the amount of nitrogen in the jet is continually changing due to entrainment. This causes the theory to predict blowout at different values of the local jet equivalence ratio. The maximum value of the kinetic loading, ψ , occurs near $ER=1.5$ for a ratio of nitrogen to oxygen of 11, while it occurs near 1.0 for a ratio of nitrogen to oxygen of 3.77 (normal air). Thus, as the jet entrains air, the local value of the ratio of nitrogen to oxygen moves the blowout point to leaner mixtures. In addition, the maximum value of ψ increases rapidly as the ratio of nitrogen to oxygen decreases.

5. CONCLUSIONS

The turbulent $H_2-O_2-N_2$ flame is theoretically modeled using finite rate chemical kinetics. The fluid dynamics are represented by a well-stirred-reactor. The model allows the flame to be located either at the exit of the burner, or in a stable lifted position above the burner exit. Its position is a function of the ambient atmospheric environment, the flow rate and the equivalence ratio at the burner exit. The model contains a free parameter, which is evaluated by comparing the theoretical predictions to the experimental results.

A simple hydrogen flame stability experiment was run and the results were compared to the theory. The theory and experiment were found to agree if the free parameter in the theory varied as the reciprocal of the turbulent jet radius squared.

6. REFERENCES

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APPENDIX A: DATA REDUCTION

The volume flow rates, Q_r , read from the rotameters in liters/minute were corrected to standard conditions by

$$Q = \left[\frac{T_r}{T} \frac{P}{P_r} \right]^{\frac{1}{2}} Q_r \quad (A1)$$

where the subscripted variables are those recorded during the experiment and the non-subscripted variables represent values at standard conditions. The standard conditions are taken as $T=274.0^\circ\text{K}$ and $P=760.0\text{ mm Hg}(=1\text{ atmosphere})$. The rotameter calibration curves, which give volume flow rates Q_r in terms of the height of the rotameter float, were supplied by the Matheson Gas Products Company. The nitrogen calibration data was used for the Q_r values for the oxygen-nitrogen mixture since the mixture was 11/12 nitrogen by volume. The values of P_r and T_r were determined from the thermocouple and pressure gauge located at the exit of the mixing tube.

The mole fractions of H_2 and $\text{O}_2\text{-N}_2$ are

$$X_{\text{H}_2} = \frac{Q_{\text{H}_2}}{Q_{\text{H}_2} + Q_A} \quad (A2)$$

$$X_A = \frac{Q_A}{Q_{\text{H}_2} + Q_A} \quad (A3)$$

where the subscript A represents the oxygen-nitrogen mixture.

The mass fractions of H_2 and the $\text{O}_2\text{-N}_2$ mixture are

$$Y_i = M_i X_i / \bar{M} \quad i = \text{H}_2, A \quad (A4)$$

where M_i is the molecular weight of the gas i and \bar{M} is the average molecular weight of the mixture.

$$M_{\text{H}_2} = 2.016 \text{ gm/gm-mole} \quad (A5)$$

$$M_A = \frac{1}{12} (32.0) + \frac{11}{12} (28.016) = 28.35 \text{ gm/gm-mole} \quad (A6)$$

$$\bar{M} = \sum_i M_i X_i = M_{\text{H}_2} X_{\text{H}_2} + M_A X_A \quad (A7)$$

Thus, one can write the mass fractions in terms of the measured quantities as

$$Y_{\text{H}_2} = \frac{M_{\text{H}_2} Q_{\text{H}_2}}{M_{\text{H}_2} Q_{\text{H}_2} + M_A Q_A} \quad (A8)$$

$$Y_A = \frac{M_A Q_A}{M_{\text{H}_2} Q_{\text{H}_2} + M_A Q_A} \quad (A9)$$

In order to compare the experiment and theory, one needs to compute u_o/d_o from the experiment. This can be calculated as follows:

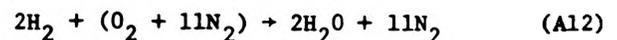
$$\frac{u_o}{d_o} = \frac{u_o A_o}{d_o A_o} = \frac{Q_o}{d_o A_o} = \frac{(Q_{\text{H}_2} + Q_A)}{d_o A_o} \quad (A10)$$

where A_o is the nozzle exit area. ($A_o = 0.0270\text{ cm}^2$; $d_o = 0.1854\text{ cm} = 0.0730\text{ inches}$)

Finally, one must calculate the fuel/oxygen equivalence ratio. It is defined as

$$\text{ER} = \frac{X_{\text{H}_2}/X_{\text{O}_2}}{\left(\frac{X_{\text{H}_2}}{X_{\text{O}_2}}\right)_{\text{Theoretical}}} \quad (A11)$$

The theoretical reaction is



and therefore,

$$\left(\frac{X_{\text{H}_2}}{X_{\text{O}_2}}\right)_{\text{Theoretical}} = 2 \quad (A13)$$

and

$$X_{\text{H}_2}/X_{\text{O}_2} = Q_{\text{H}_2}/Q_{\text{O}_2} \quad (A14)$$

so that one can write the equivalence ratio at the burner exit as

$$ER_o = (Q_H/Q_{O_2})_o / 2 = 6 (Q_{H_2}/Q_A)_o \quad (A15)$$

BIOGRAPHY

H. Frederick Nelson is an Associate Professor of Aerospace Engineering at the University of Missouri-Rolla. He holds a B.S. Degree from Iowa State University and the M.S. and Ph.D. Degrees from Purdue University.

TABLE 2: Theoretical Results: $\kappa = 1$

$(ER)_o$	$(Y_f)_o$	$(X_f)_o$	u_o/d_o sec^{-1}
1.0	.0115	.140	563
2.0	.0227	.245	2,985
4.0	.0443	.392	7,452
6.0	.0651	.490	10,211
8.0	.0849	.560	11,889
10.0	.104	.613	13,112

TABLE 1: Experimental Blowout Conditions

Equiv. Ratio ER_o	Mass Fractions		Mole Fractions		u_o/d_o (p) (sec^{-1})	u_o/d_o (ss) (sec^{-1})
	Y_{H_2} (p)	Y_{H_2} (ss)	X_{H_2} (p)	X_{H_2} (ss)		
11.087	*	0.120	*	0.65	*	78391.06
10.271	*	0.110	*	0.63	*	76913.12
9.607	*	0.104	*	0.62	*	74854.53
9.348	*	0.100	*	0.61	*	72414.85
8.484	*	0.093	*	0.59	*	70225.16
7.752	*	0.083	*	0.56	*	67701.26
7.142	*	0.077	*	0.54	*	64666.72
6.316	*	0.069	*	0.51	*	61936.10
5.677	*	0.064	*	0.49	*	57489.48
5.006	0.057	0.055	0.46	0.45	55067.78	53054.18
4.897	*	0.055	*	0.45	*	54207.28
4.133	0.049	0.047	0.42	0.41	51243.31	49849.21
3.536	0.042	0.040	0.38	0.37	46392.24	44524.45
2.640	0.035	0.031	0.34	0.31	39197.69	36652.49
2.100	0.030	0.024	0.30	0.26	34446.49	32070.67
1.756	0.027	0.021	0.28	0.23	29457.61	26979.97
1.248	0.022	0.014	0.24	0.17	22975.42	20701.85
0.833	0.019	0.0096	0.21	0.12	14864.78	13914.41

* - indicates instrument was off - scale or "pegged".

(p) = pyrex ball

(ss) = stainless steel ball

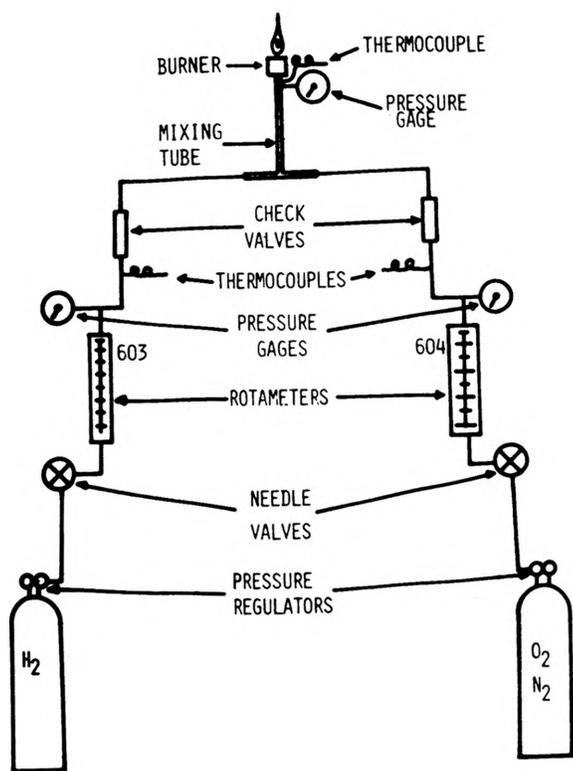


FIGURE 1. SCHEMATIC OF EXPERIMENTAL SET-UP.

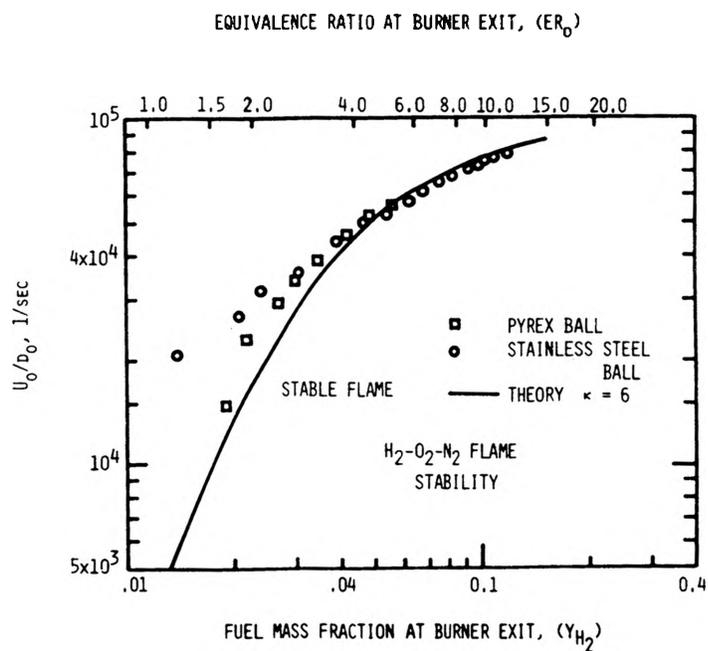


FIGURE 3. COMPARISON OF THE THEORETICAL AND EXPERIMENTAL FLAME STABILITY RESULTS FOR TURBULENT $H_2/O_2/N_2$ FLAMES.

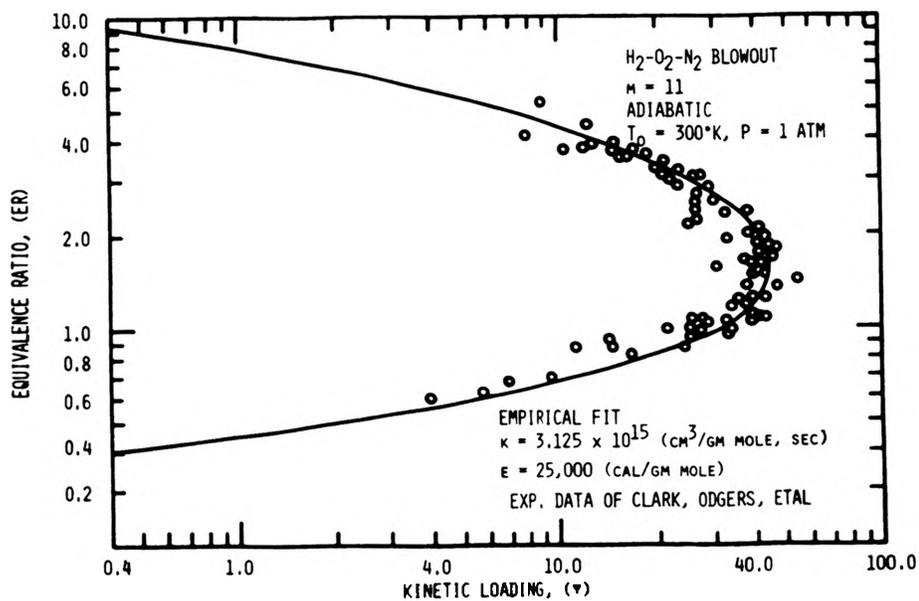


FIGURE 2. EMPIRICAL AND EXPERIMENTAL $H_2/O_2/N_2$ WELL STIRRED REACTOR BLOWOUT AS A FUNCTION OF EQUIVALENCE RATIO. THE EMPIRICAL CURVE IS GIVEN BY EQUATION (20) WITH $m=11$.