Analysis of Gaseous Fuel and Air Mixing in Flames and Flame Quenching

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A model for fuel-air mixing in diffusion flames is presented and applied to study the mixing and quenching of methane-air flames. The model is based on the ideal gas law, the energy equation, the equation of continuity, and Arrhenius form of rate equation and is, therefore, strictly valid for mixtures having low density, that is, for lowpressure combustors. In the absence of preferential diffusion, chemical reactions cause an unbalanced consumption of fuel and oxygen in nonstoichiometric flames. Until the desired equivalence ratio is achieved, enhanced preferential diffusion of oxygen or fuel is required in fuel-rich or fuel-lean flames, respectively. After a desired equivalence ratio is achieved, preferential diffusion of oxygen or fuel should be reduced to the exact level required to compensate for the unbalanced consumption of fuel and air. In the absence of these conditions, flame chemistry cannot be strictly controlled. In addition, unless the desired equivalence ratio is at a position of stable equilibrium over an extended range of operational conditions, the flame may be quenched. Net transport of fuel or oxygen due to diffusion is correlated with distributions of pressure, temperature, velocity, mass fraction of species, and heat transfer through radiation and conduction. Results show that negative rates of pressure (or positive rates of temperature) and positive rates of pressure (or negative rates of temperature) can enhance preferential diffusion of oxygen and fuel, respectively. Negative velocity divergence also enhances the diffusion of oxygen, whereas positive velocity divergence enhances the diffusion of fuel. Recirculation of burnt gases improves the stability of all flames. For rates of pressure of less than 1 atm/s, heat addition through conduction or radiation can provide a position of stable equilibrium for fuel-rich flames. A position of stable equilibrium can be provided for both fuel-rich and fuel-lean flames by combining a positive rate of temperature with positive velocity divergence, for rates of pressure of up to 25 atm/s. At higher rates of pressure or temperature, increased initial pressure or temperature, respectively, also assists in flame stabilization.

Nomenclature

= rate of mass fraction of fuel burnt, 1/s = specific heat at constant pressure, J/kg K

= heat of formation of methane at a pressure of 1 atm

and temperature of 298.15 K, J/kg fuel h = specific fluid enthalpy of fluid, J/kg

= total enthalpy of fluid $\equiv h + U^2/2$, J/kg

M = molecular weight, kg/mol

m = mass, kg P = pressure, atm

Q= ratio of heat transfer and heat released by combustion rates, dimensionless

R = universal gas constant/atmospheric pressure, m³/K mol

= rate of equivalence ratio, s

S = source term T= temperature, K

U= magnitude of overall velocity, m/s

 \boldsymbol{U} = overall velocity, m/s

= mass fraction, dimensionless y

Δ = main determinant Δ_f = fuel determinant = oxygen determinant

= determinant of combustion products

= density, kg/m³ ρ $\rho(0)$ = initial density, kg/m³ = integration variable

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= equivalence ratio, dimensionless $\phi(0)$ = initial equivalence ratio

Subscripts

c= combustion = equilibrium eq = fuel f

= mixture m

n = molecular nitrogen = molecular oxygen 0

= products (water and carbon dioxide) p

= quenching

st = stoichiometric conditions

I. Introduction

P REVIOUS studies have shown that controlled mixing is very important for increasing important for increasing combustor efficiency and reducing the emission of pollutants.^{1,2} Nonflammable and flammable mixtures behave in different manners but have certain similarities.³ A model for mixing in an infinitesimal element of nonflammable mixture was presented and applied to study methane-air mixing in a previous paper by the authors.⁴ This model was based on the ideal gas law and the equation of continuity. The infinitesimal elements of mixture are analyzed in an Eulerian frame of reference. Augmenting the model for nonflammable mixtures with the energy and Arrhenius form of rate equation³ provides a model for flammable mixture elements within diffusion flames. A very large number of species and chemical reactions are involved even for a simple hydrocarbon flame of methane burning in air.⁵ Some simplifying assumptions, therefore, must be made to model mixing in flames. In this study, it was assumed that the mass fraction of molecular nitrogen is not affected by combustion because only a trace amount of nitrogen present in air participates in the chemical reactions.² Therefore, in flames, the gradients of nitrogen concentration are smaller than those for reactants or combustion products. As a consequence, the flux of nitrogen due

to random molecular motion (i.e., ordinary diffusion³) is smaller than that for reactants and combustion products because the flux of a chemical species is proportional to the divergence of that species's concentration.³ Furthermore, most fuel-air mixtures contain more than 75% nitrogen on a mass basis, which means that nitrogen and mixture density are almost equal. As a consequence, pressure diffusion of nitrogen can be neglected.³ Compared to reactants and combustion products, the rate of mass fraction of nitrogen due to both diffusion and chemical reaction is negligible. Therefore, for nitrogen the species conservation law can be written as^{3,4}

$$\frac{\mathrm{d}\rho_n}{\mathrm{d}t} \approx -\rho_n \,\mathrm{div}\,\boldsymbol{U}$$

The preceding equation and the equation of continuity yield

$$\frac{\mathrm{d}\rho_n}{\mathrm{d}t} \approx \frac{\rho_n}{\rho} \frac{\mathrm{d}\rho}{\mathrm{d}t}$$

As a consequence, the rate of nitrogen density is given by

$$\frac{\mathrm{d}\rho_n}{\mathrm{d}t} \approx y_n \frac{\mathrm{d}\rho}{\mathrm{d}t} \tag{1}$$

By definition, the density and mass fraction of any chemical species i are correlated

$$\rho_i \equiv \rho \cdot y_i \tag{2}$$

Therefore,

$$\frac{\mathrm{d}\rho_i}{\mathrm{d}t} = \frac{\mathrm{d}(y_i \rho)}{\mathrm{d}t} \equiv y_i \frac{\mathrm{d}\rho}{\mathrm{d}t} + \rho \frac{\mathrm{d}y_i}{\mathrm{d}t} \tag{3}$$

Based on Eq. (2), nitrogen density can be written as

$$\rho_n \equiv \rho \cdot y_n$$

Therefore,

$$\frac{\mathrm{d}\rho_n}{\mathrm{d}t} = \frac{\mathrm{d}(y_n \rho)}{\mathrm{d}t} \equiv y_n \frac{\mathrm{d}\rho}{\mathrm{d}t} + \rho \frac{\mathrm{d}y_n}{\mathrm{d}t} \tag{4}$$

Equations (1) and (4) show that

$$\frac{\mathrm{d}y_n}{\mathrm{d}t} \approx 0$$

Therefore, it can be assumed that the overall rate of mass fraction of nitrogen is negligible compared to the overall rates of reactants and combustion products; that is, the mass fraction of nitrogen remains almost constant.

A single-step chemical reaction model is considered, which for methane is

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$$

It is also assumed that diffusive transport of combustion products is negligible compared to their mass sources due to chemical reaction (i.e., the second Damkohler number³ is large); that is, $Da_{II} \equiv \text{mass}$ source from chemical reactions/diffusive transport > 10. Therefore, for water and carbon dioxide, the effect of diffusive transport is secondary compared to the effect of chemical reactions. The law of species conservation applied to water and carbon dioxide with diffusive transport being negligible yields

$$\frac{\mathrm{d}\rho_{\mathrm{H_2O}}}{\mathrm{d}t} = -\rho_{\mathrm{H_2O}} \operatorname{div} U + S_{\mathrm{H_2O}}$$

$$\frac{\mathrm{d}\rho_{\mathrm{CO}_2}}{\mathrm{d}t} = -\rho_{\mathrm{CO}_2} \operatorname{div} \boldsymbol{U} + S_{\mathrm{CO}_2}$$

By the use of Eq. (3), the given rates of density of water and carbon dioxide can be written as

$$\frac{d\rho_{\rm H_2O}}{dt} = \frac{dy_{\rm H_2O}}{dt}\rho + y_{\rm H_2O}\frac{d\rho}{dt} = -\rho_{\rm H_2O}\,{\rm div}\,\bm{U} + S_{\rm H_2O}$$

$$\frac{\mathrm{d}\rho_{\mathrm{CO}_2}}{\mathrm{d}t} = \frac{\mathrm{d}y_{\mathrm{CO}_2}}{\mathrm{d}t}\rho + y_{\mathrm{CO}_2}\frac{\mathrm{d}\rho}{\mathrm{d}t} = -\rho_{\mathrm{CO}_2}\operatorname{div}\boldsymbol{U} + S_{\mathrm{CO}_2}$$

Therefore, the rates of mass fraction of water and carbon dioxide are given, respectively, by

$$\frac{dy_{H_2O}}{dt} = \frac{1}{\rho} \left(\rho_{H_2O} \operatorname{div} U - \rho_{H_2O} \operatorname{div} U + S_{H_2O} \right) = \frac{S_{H_2O}}{\rho}$$

$$\frac{dy_{CO_2}}{dt} = \frac{1}{\rho} \left(\rho_{CO_2} \operatorname{div} U - \rho_{CO_2} \operatorname{div} U + S_{H_2O} \right) = \frac{S_{CO_2}}{\rho}$$

As a consequence

$$\frac{\mathrm{d}y_{\mathrm{H}_2\mathrm{O}}/\mathrm{d}t}{\mathrm{d}y_{\mathrm{CO}_2}/\mathrm{d}t} = \frac{S_{\mathrm{H}_2\mathrm{O}}}{S_{\mathrm{CO}_2}} = \mathrm{const}$$

Assuming that the mass fractions of water and carbon dioxide are negligible before the mixture ignities at time t_0 , the ratio between the mass fractions of water and carbon dioxide at a later time t is given by

$$\frac{y_{\text{H}_2\text{O}}(t)}{y_{\text{CO}_2}(t)} = \int_{t_0}^{t} \frac{dy_{\text{H}_2\text{O}}}{dt} dt / \int_{t_0}^{t} \frac{dy_{\text{CO}_2}}{dt} dt$$

$$= \frac{S_{\text{H}_2\text{O}}}{S_{\text{CO}_2}} \int_{t_0}^t \frac{dy_{\text{CO}_2}}{dt} dt / \int_{t_0}^t \frac{dy_{\text{CO}_2}}{dt} dt = \frac{S_{\text{H}_2\text{O}}}{S_{\text{CO}_2}} = \text{const}$$

This suggests that the ratio between water and carbon dioxide is governed by reaction stoichiometry, and for methane-air flame this ratio is about 2–1, on a volume basis.⁶ The mass fractions of intermediate species (such as CO, CH₃, HCO, OH, H, H₂, HO₂, etc.) have been assumed to be negligible.

The assumptions that fuel-air mixture contains mostly nitrogen, Damkohler number Da_{II} is large for water and carbon dioxide, the ideal gas law, the equation of continuity, and the energy equation yield

$$y_f + y_o + y_n + y_p = 1$$

$$(y_f/M_f) + (y_o/M_o) + (y_n/M_n) + (y_p/M_p) = P/RT\rho$$

$$h = h_f y_f + h_o y_o + h_n y_n + h_p y_p$$
(5)

The molecular weight of combustion products (which are assumed to be water and carbon dioxide only) is given by⁶

$$M_p = \frac{1}{3} (2M_{\rm H_2O} + M_{\rm CO_2})$$

Therefore, the specific enthalpy of combustion products can be calculated from

$$h_p = (1/3M_p) (2M_{\text{H}_2\text{O}} h_{\text{H}_2\text{O}} + M_{\text{CO}_2} h_{\text{CO}_2})$$

The mass fractions of water and carbon dioxide are, respectively, given by

$$y_{\text{H}_2\text{O}} \approx 2 \left(M_{\text{H}_2\text{O}} / M_p \right) y_p$$
 $y_{\text{CO}_2} \approx \left(M_{\text{CO}_2} / M_p \right) y_p$

The total enthalpy of fluid is defined as⁷

$$h^* = h + (U^2/2)$$

where

$$U = |U|$$

Heat transfer through conduction and radiation, the rate of total enthalpy of mixture, and the rate of energy released by combustion are correlated, as shown by the energy conservation law³:

$$\frac{\mathrm{d}h^*}{\mathrm{d}t} = (Q+1)|H_f S_f| + \frac{1}{\rho} \frac{\mathrm{d}P}{\mathrm{d}t}$$

Based on the assumption that fuel and air are thoroughly mixed within the infinitesimal element of flammable mixture, the source

term for fuel, (kilograms of fuel/kilograms of mixture-second) S_f is given by $^{8.9}$

$$S_f = \left(\frac{\mathrm{d}y_f}{\mathrm{d}t}\right)_c = -0.5(101, 315P)^2 y_o y_f \exp\left(\frac{-20000}{T}\right)$$

The heat of combustion of methane H_f at 298.15 K and 1 atm, is 50,047 kJ/kg fuel. ¹⁰

Based on the assumption that the mass fraction of nitrogen is constant and based on the system of Eqs. (5), the rates of mass fraction of nitrogen, fuel, oxygen, and combustion products are given by

$$\frac{dy_n}{dt} = 0 \qquad \frac{dy_f}{dt} + \frac{dy_o}{dt} + \frac{dy_p}{dt} = 0$$

$$\frac{1}{M_f} \frac{dy_f}{dt} + \frac{1}{M_o} \frac{dy_o}{dt} + \frac{1}{M_p} \frac{dy_p}{dt} = \frac{d}{dt} \left(\frac{P}{RT\rho}\right)$$

$$h_f \frac{dy_f}{dt} + h_o \frac{dy_o}{dt} + h_p \frac{dy_p}{dt} = \alpha \tag{6}$$

where

$$\alpha \equiv -C_{pm} \frac{\mathrm{d}T}{\mathrm{d}t} - \frac{\mathrm{d}}{\mathrm{d}t} \left(\frac{U^2}{2}\right) + |H_f S_f| (1+Q)$$

The mixture specific heat at constant pressure is given by

$$c_{pm} = c_{pf} y_f + c_{po} y_o + c_{pn} y_n + c_{pH_2O} y_{H_2O} + c_{pCO_2} y_{CO_2}$$

The preceding system of equations (6) can be solved using Cramer's (see Ref. 11) rule; that is, the determinants of fuel Δ_f , oxygen Δ_o , and combustion products Δ_p , and the main determinant Δ can be written as

$$\Delta_f = \begin{vmatrix} 0 & 1 & 1 \\ \frac{d}{dt} \left(\frac{P}{RT\rho} \right) & \frac{1}{M_o} & \frac{1}{M_p} \\ \alpha & h_o & h_p \end{vmatrix}$$
 (7)

$$\Delta_o = \begin{vmatrix} 1 & 0 & 1\\ \frac{1}{M_f} & \frac{d}{dt} \left(\frac{P}{RT\rho} \right) & \frac{1}{M_p} \\ h_f & \alpha & h_p \end{vmatrix}$$
 (8)

$$\Delta_{p} = \begin{vmatrix} 1 & 1 & 0 \\ \frac{1}{M_{f}} & \frac{1}{M_{o}} & \frac{d}{dt} \left(\frac{P}{RT\rho} \right) \\ h_{f} & h_{o} & \alpha \end{vmatrix} \qquad \Delta = \begin{vmatrix} 1 & 1 & 1 \\ \frac{1}{M_{f}} & \frac{1}{M_{o}} & \frac{1}{M_{p}} \\ h_{f} & h_{o} & h_{p} \end{vmatrix}$$
(9)

and the rates of mass fraction of fuel $\mathrm{d}y_f/\mathrm{d}t$, oxygen, $\mathrm{d}y_o/\mathrm{d}t$, and combustion products $\mathrm{d}y_p/\mathrm{d}t$ are given by

$$\frac{\mathrm{d}y_f}{\mathrm{d}t} = \frac{\Delta_f}{\Delta} \tag{10}$$

$$\frac{\mathrm{d}y_o}{\mathrm{d}t} = \frac{\Delta_o}{\Delta} \tag{11}$$

$$\frac{\mathrm{d}y_p}{\mathrm{d}t} = \frac{\Delta_p}{\Delta} \tag{12}$$

The equivalence ratio Φ is defined as^{3,4}

$$\phi = (y_a/y_f)_{st}(y_f/y_a)$$

For methane-air flames the stoichiometric ratio between air and fuel, on a mass basis, is 17.2; therefore, for such flames, the equivalence ratio is given by

$$\phi = 17.2(y_f/y_a)$$

Air contains 21% oxygen on a volume basis or 23% on a mass basis. Because of the participation of oxygen present in the air in the combustion process, the volume or mass of oxygen changes. In flames, therefore, the ratio between oxygen and nitrogen is changed. The equivalence ratio has to be redefined based on the amount of air that would correspond to the remaining oxygen; that is,

$$\phi = 17.2(0.23y_f/y_o) = 3.956(y_f/y_o)$$
 (13)

A parameter that can be used to study fuel-air mixing is the rate of equivalence ratio r(t) defined as⁴

$$r(t) = \frac{1}{\phi} \frac{\mathrm{d}\phi}{\mathrm{d}t} \tag{14}$$

Positive or negative rates of equivalence ratio indicate that the mixture becomes more fuel-rich or more fuel-lean, respectively. Two types of rate of equivalence ratio that can be distinguished are 1) rates that enhance mixing, that is, produce or maintain a flammable mixture having a target equivalence ratio and 2) rates that hamper mixing, that is, prevent the formation of a desirable type of flammable mixture or cause the mixture to become nonflammable. The first type of equivalence ratio can be called rate of mixing.⁴ In flames, rates of equivalenceratio of the second type usually measure the rate of quenching and, therefore, will be called here rates of quenching. For example, if the desired equivalence ratio is unity, a positive or negative rate of equivalence ratio in a fuel-lean or fuelrich mixture, respectively, causes the equivalence ratio to approach the desired value and is called the rate of mixing. A negative or positive rate of equivalence ratio in a fuel-lean or in a fuel-rich mixture, respectively, yields a nonflammable mixture and is called the rate of quenching.

To maximize combustion efficiency and to reduce pollutant emissions, flame chemistry must be carefully tailored. Therefore, after achieving the desired equivalence ratio, the rate of equivalence ratio r(t) should be minimized. On the other hand, enhanced mixing requires high rates of equivalence ratio (positive in fuel-lean flames and negative in fuel-rich flames). If the desired equivalence ratio is at a position of stable equilibrium, 12 rates of equivalence ratio would be automatically minimized after reaching the position of equilibrium. Unless operational conditions are rapidly changed after the desired equivalence ratio is attained, enhanced mixing assures just as fast flame quenching.

In a flame, the rate of equivalence ratio is given by

$$r(t) = \frac{3.956}{\phi} \left[\left(y_o \frac{\mathrm{d}y_f}{\mathrm{d}t} - y_f \frac{\mathrm{d}y_o}{\mathrm{d}t} \right) \middle/ y_0^2 \right] = \frac{1}{y_f} \frac{\mathrm{d}y_f}{\mathrm{d}t} - \frac{1}{y_o} \frac{\mathrm{d}y_o}{\mathrm{d}t}$$
(15)

Equation (15), used in conjunction with Eqs. (10) and (11), yields

$$r(t) = 1/\Delta[(\Delta_f/y_f) - (\Delta_o/y_o)]$$
 (16)

Therefore, the rate of equivalence ratio in a flame depends on the distributions of pressure, temperature, velocity, and mass fraction of oxygen, fuel, and combustion products [see Eqs. (7), (8), and (16)]. The rate of equivalence ratio also depends on heat transfer through radiation and conduction. High-frequency sounds that provide rates of pressure that are alternately positive and negative have been shown to affect mixing in nonflammable mixtures.⁴ As seen from Eqs. (7), (8), and (16), high rates of pressure have a significant effect on mixing. Therefore, high-frequency sound waves also affect flammable mixtures.

High rates of quenching may cause the equivalence ratio to either drop below the fuel-lean ($\Phi_l = 0.5$) or exceed the fuel-rich ($\Phi_r = 1.68$) flammability limit, ³ thereby quenching the flame. If the distributions of pressure, temperature, velocity, chemical species,

and intensity of heat transfer through conduction and radiation are known at any instant of time, then the rate of equivalence ratio can be expressed as a function of time using Eq. (16). The mixing time t_m or the quenching time t_q with known value of r(t), can both be calculated from

$$\phi(t) = \phi(0) \exp \int_0^t r(\tau) d\tau$$
 (17)

For example, if the initial equivalence ratio $\Phi(0)$ is 0.5, the target value is 1.0, and r(t) has a value of 4000 s⁻¹, the mixing time (in seconds, s) is given by

$$t_m = \frac{\ln[(\phi(t_m)/\phi(0))]}{4000} = 0.00017 \text{ s}$$

This mixing time would be similar for an initial equivalence ratio of 1.68.

The residence time in a characteristic gas turbine combustor is about 1 or 2 ms. Mixing times that are more than 10 times shorter than the residence time are desirable. Therefore, it is tempting to pursue rates of equivalence ratio that are greater than 4000 s^{-1} . However, quenching times, unless the stoichiometric value is at a position of stable equilibrium, would also be shorter than 0.1 s, as shown by Eq. (15). By making the assumption that there is a constant rate of quenching r the quenching time is 1/r. For example if the rate of quenching has a constant value of 1000 s^{-1} and the initial equivalence ratio $\Phi(0)$ is 1.0, Eq. (15) shows that the quenching time is given by

$$t_q = \frac{\ln[(\phi(t_q)/\phi(0)]}{r} = \frac{\ln(1.68)}{1000} = 5.188 \cdot 10^{-4} \text{ s}$$

An initial equivalence ratio of 1.0 and a rate of quenching of -1000 s⁻¹ provide a quenching time of

$$t_q = \frac{\ln(0.5/1)}{-1000} = 6.931 \cdot 10^{-4} \text{ s}$$

In typical gas turbine combustors, the fuel is required to burn in about 1 ms. To assure complete combustion of fuel, the quenching time should exceed the time required to burn the fuel, which means that the rate of quenching should be kept below $1000 \, {\rm s}^{-1}$.

To summarize, if the desired equivalence ratio is at a position of unstable equilibrium, the magnitude of the rate of equivalence ratio should exceed $4000 \, \text{s}^{-1}$ for enhanced mixing, and immediately after achieving the target equivalence ratio, the rate should be reduced to less than $1000 \, \text{s}^{-1}$. It is a challenge to achieve such an abrupt change. These results show that enhanced mixing and flame stability can be mutually exclusive when the target equivalence ratio is at a position of unstable equilibrium.

In practical combustor designs, r is not constant, and Eq. (17) has to be solved numerically. A constant rate of mixing or quenching, having an absolute value given by

$$|r| = \max(|r(t)|)$$

can provide a lower limit for the mixing or quenching time.

It is known that flame stretch¹³ can be used to quantify various phenomena involved in flame stabilization.^{14,15} The capacity of stretch to stabilize, as well as to quench the flame has been discussed previously.¹⁶ The results show that the behavior of flame also depends on diffusion, heat transfer, initial concentrations of products and reactants, type of fuel, distributions of pressure, and temperature.¹⁶

II. Diffusion in Flame

If pressure, temperature, and velocity are kept constant and heat transfer and chemical reactions are negligible, the determinants Δ_f , Δ_o , and Δ_p are zero, as seen from Eqs. (7–9). As a consequence, the rates of mass fraction of fuel $(\mathrm{d}y_f/\mathrm{d}t)$, oxygen $(\mathrm{d}y_o/\mathrm{d}t)$ and combustion products $(\mathrm{d}y_p/\mathrm{d}t)$ are zero, as shown by Eqs. (10–12). The

mass fraction of fuel, oxygen, and combustion products are constant only in the absence of convective and preferential diffusion transport, that is, the net transport of fuel, air, and combustion products is linked to velocity gradients, rates of pressure, temperature, chemical reactions or heat transfer through conduction, and radiation.

The rate of the equivalence ratio quantifies the combined effect of net transport and combustion on the equivalence ratio, as shown by Eqs. (7), (8), and (16). As a consequence, the rate of equivalence ratio can be positive or negative even under net transport of oxygen or fuel, respectively. Nevertheless, preferential diffusion of fuel or oxygen causes an increase or decrease, respectively, in the rate of equivalence ratio r(t).

III. Flame Stability

A flame is stable over a range of operational conditions only if the equivalence ratio is at a position of stable equilibrium 12 (i.e., Φ_{eq}), that lies between the flammability limits. For an equilibrium position, the rate of equivalence ratio is zero. Equation (15) shows that the rate of equivalence ratio is zero if

$$\left(\frac{\mathrm{d}y_f}{\mathrm{d}t}y_o - y_f \frac{\mathrm{d}y_o}{\mathrm{d}t}\right) / y_o^2 = 0$$

The consumption of fuel and oxygen is balanced (i.e., maintains a constant equivalence ratio in the absence of preferential diffusion) if

$$\left(\frac{\mathrm{d}y_f/\mathrm{d}t}{\mathrm{d}y_o/\mathrm{d}t}\right)_o = \frac{y_f}{y_o}$$

because

$$\left(\frac{\mathrm{d}y_f/\mathrm{d}t}{\mathrm{d}y_o/\mathrm{d}t}\right)_c \equiv \left(\frac{y_f}{y_o}\right)_{\mathrm{st}}$$

In the absence of preferential diffusion, Eq. (15) can be written as

$$r(t) = \frac{(\mathrm{d}y_o/\mathrm{d}t)_c}{y_f} \left[\left(\frac{y_f}{y_o} \right)_{\mathrm{st}} - \frac{y_f}{y_o} \right]$$

By definition

$$\left(\frac{\mathrm{d}y_o}{\mathrm{d}t}\right)_{a} < 0$$

Also from definition, in a fuel-rich mixture

$$(y_f/y_o)_{st} - (y_f/y_o) < 0$$

As a consequence, in a fuel-rich mixture, the rate of equivalence ratio is positive, that is, combustion causes the equivalence ratio to increase. A similar reasoning shows that combustion reduces the equivalence ratio in a fuel-lean flame. Therefore, the consumption of fuel and oxygen is unbalanced in a nonstoichiometric flame. As a consequence, in the absence of net transport, rates of quenching are produced in both fuel-rich and fuel-lean flames.

The output of a linear system that is stable remains bounded (finite) for any bounded input.^{17–19} The system is stable if the variation of the system output about the initial steady state (i.e., stable equilibrium position) tends to zero.¹⁷ The system is unstable if the output increases indefinitely with time, that is, unstable equilibrium position. The system is marginally stable if the output undergoes continuous bounded oscillations. A system has limited stability if the output attains a constant value other than the initial one. System stability requires negative feedback. If at a time t_0 , the equivalence ratio is at an equilibrium position, $\Phi_{\rm eq}$ (which is also the desired value), then the rate of equivalence ratio $r(t_0)$ is zero. At time $t_0 + \delta t$, due to the effect of a perturbation in the flowfield, distribution of pressure or temperature, or via changes in heat transfer through radiation and conduction, the equivalence ratio is $\Phi(t_0 + \delta t)$, which is $\neq \Phi_{\rm eq}$, the rate of equivalence ratio $r(t_0 + \delta t)$ is no longer zero.

The rate of equivalence ratio, $r(t_0 + \delta t)$, can be the rate of mixing only when providing negative feedback for the equivalence ratio under perturbations, that is, when the equivalence ratio satisfies the condition

$$[\phi(t_0 + \delta t) - \phi_{eq}]r(t_0 + \delta t) < 0$$
 (18)

Inequality in Eq. (18) is necessary but not sufficient to assure flame stability. When inequality in Eq. (18) is satisfied, the rate of equivalence ratio counters the effect of the perturbation and tends to preserve the desired equivalence ratio, unless the equivalence ratio begins to oscillate with continuously increasing (unbounded) amplitude. An assurance that the equivalence ratio remains at a stable position of equilibrium over an extended range of operational conditions requires an in-depth analysis of flame in terms of system stability. 17–19

Usual rates of quenching provide positive feedback, i.e.,

$$\left[\phi(t_0 + \delta t) - \phi_{\text{eq}}\right] Rm(t_0 + \delta t) > 0 \tag{19}$$

If the preceding condition is satisfied, then Φ_{eq} is immediately recognized as a position of unstable equilibrium, that will provide flame quenching whenever the rates of quenching, which are caused by perturbations in flowfield, pressure, temperature, or heat transfer, exceed $1000~{\rm s}^{-1}$.

The model presented here quantifies the effect of heat transfer, initial concentration of reactants and products, and distributions of pressure, temperature, and velocity on flame behavior. Previous studies of flame quenching rely on the implicit assumption that only a few of these factors affect flame stability. For example, attempts to determine a critical value of flame stretch that triggers flame blowoff rely on the assumption that the velocity distribution and flame geometry are the only factors that cause flame extinction. The system of equations (6) shows that flame stretch alone cannot provide a truly physical model of flame behavior. The use of flame or fluid element stretch may be considered only if the mass fraction of nitrogen is less than 50%, if the Damkohler number Da_{II} is small (for water or carbon dioxide), if there is a nonnegligible mass fraction of intermediate combustion products, or if the ideal gas law is not accurate. Similarly, flame studies based on the Karlowitz number assume that only the rate of chemical reactions, velocity distribution, and flame geometry affect flame quenching.3

This study is focused on infinitesimal elements of a mixture. Extrapolating the results obtained on this scale to the entire flame requires further examination. Flames need to be analyzed using a mesh of infinitesimal elements. The mesh structure has to be chosen carefully to span the entire combustion region and to reveal the effect of flame geometry on quenching.

IV. Computational Approach

The rates of mixing, based on Eq. (16), have been calculated here using MATHCAD 5.0. Results have been obtained here on flame quenching under low and moderate pressures ranging from 1 to 5.0 atm because the ideal gas law is strictly valid at low pressures. $The \, operational conditions analyzed here \, are \, intended to \, emulate \, not \,$ only those normally encountered in low-pressure combustors, but also extreme conditions leading to flame extinction such as rapid cooling of the reactants during their contact with a cold surface. The rates of pressure are varied from -100 to 100 atm/s. The initial temperature of mixture is varied from 1050 to 2250 K and the rate of temperature from -2×10^7 to 2×10^7 K/s. The rate of the kinetic energy of fluid is varied from -30.0 to 30.0 MW/kg. The rate of heat transfer per unit mass of fluid through radiation and conduction is varied from -150% (as in the case of flame extinction on contact with a cold surface) to 150% of the rate of energy released by combustion (due to high temperature variations in flame, the heat produced by combustion within some mixture elements may exceed the heat received through radiation). The mass fraction of carbon dioxide and water ranged from 0.0 to 0.25 (mostly burnt gases given a stoichiometric mixture). The initial equivalence ratio ranged from fuel-lean (0.5) to fuel-rich (1.68), including all flammable mixtures. The velocity divergence was varied from -600

to 600 1/s, a range that is usual in current combustor designs.²⁰ The velocity divergence provides the time rate of the volume of fluid element. The range of initial pressures is limited by the use of the ideal gas law. The ranges of rates of temperature, pressure, and kinetic energy of fluid exceed those present in most subsonic combustors.²⁰

V. Results and Discussion

Results are presented here on the effect of equivalence ratio, distribution of pressure and temperature, kinetic energy of mixture, intensity of heat transfer through conduction and radiation, mass fraction of combustion products, and velocity divergence on mixing and quenching of methane–air flames. Baseline pressure and temperature were constant at 1 atm/s and 1050 K, respectively. Baseline velocity divergence, rate of kinetic energy of fluid, heat transfer, and mass fraction of combustion products were negligible. Results presented here for extreme rates of pressure (about 100 atm/s) or temperature (about 2×10^7 K/s) are only approximate because pressure or temperature diffusion, respectively, is no longer negligible compared to the mass source from chemical reactions, that is, the Damkohler number Da_{II} is reduced (less than 10). As a consequence, the ratio between mass fractions of water and carbon dioxide is no longer about 2.

Figure 1 shows the effect of initial equivalence ratio on mixing and quenching of flames. Two constant temperatures of 1050 and 2250 K of mixture were considered. A tight control of local equivalence ratio is needed to increase combustion efficiency and limit the pollutants emission level. To maintain the desired ratio of fuel and air, the rate of local equivalence ratio has to be minimized. Positive rates of equivalence ratio show mixtures become more fuel-rich (for r > 0, see the upper half of Fig. 1). Negative rates indicate mixtures become more fuel-lean. Net transport of fuel and oxygen due to diffusion is negligible, as shown in Sec. II. As a result, rates of quenching are produced in both fuel-rich and fuel-lean mixtures, as shown in Sec. I. Figure 1 shows that the rate of quenching is proportional to the difference between the initial equivalence ratio and 1.0. In a mixture with an initial equivalence ratio of 0.5, the rates of quenching are about -125,000 and -15 s⁻¹ at temperatures of 2250 and 1050 K, respectively. At an initial equivalence ratio of 1.6, the rates of quenching are about 150,000 and 15 s⁻¹ corresponding to the temperatures just given. The line for 2250 K has a higher slope than the line for 1050 K. This shows that a higher flame temperature increases the net transport of fuel or air in non-stoichiometric flames. As a consequence mixing is enhanced, but flame stability is reduced. The rate of quenching observed at 1050 K would quench the flame in about 60 ms. This time is much larger than the time required to burn all of the fuel in a gas turbine combustor (characteristic burn times are of the order of a few milliseconds, as shown in Sec. I). Therefore, at 1050 K, the rate of quenching can be neglected. At 2250 K, however, fuel-rich and fuel-lean flames are very unstable, that is, at such conditions flames are quenched in time that can be as short as 0.01 ms. If the flame is locally quenched, pockets of

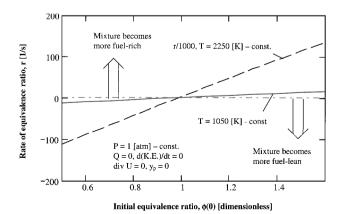


Fig. 1 Effect of equivalence ratio on mixing in flame and flame quenching.

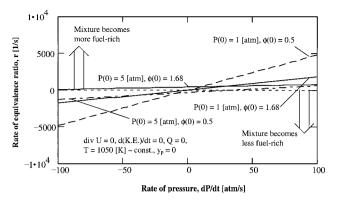


Fig. 2 Effect of pressure and rate of pressure on mixing in flame and flame quenching.

unburned hydrocarbons will form, with the result of increased pollutants emission and reduced combustion efficiency. If the region where the mixture becomes nonflammable is too large the flame may be completely extinguished. At high temperatures, preferential diffusion of oxygen or fuel is required to avoid quenching fuel-rich or fuel-lean flames, respectively. Net transport due to diffusion has been shown to correlate with rates of pressure (or temperature), velocity gradients, or heat transfer through conduction and radiation, see Sec. II. Therefore, gradients of pressure, temperature, or velocity, or heat transfer are required to balance fuel and air consumption in nonstoichiometric flames at high temperatures.

Figure 2 shows the effect of pressure on mixing and flame quenching. Calculations were made for initial pressures of 1 and 5 atm. Initial equivalence ratios of 0.5 and 1.68 are analyzed. The results show that the rate of equivalence ratio is proportional to the rate of pressure. At a pressure of 1 atm and rate of pressure of -100 atm/s, the rates of equivalence ratio are about -1600 and -5000 s⁻¹ for equivalence ratios of 1.68 and 0.5, respectively. Below an initial pressure of 1 atm, a positive rate of pressure of 100 atm/s provides rates of equivalence ratio of about 1600 and 5000 s⁻¹ for initial equivalence ratios of 1.68 and 0.5, respectively. A higher slope shows that pressure fluctuations have more impact on mixing and flame stability. Rates of equivalence ratio that exceed 5000 s⁻¹ are sufficient to provide enhanced mixing, but the required rate of pressure of about 100 atm/s is not practical. Given a higher initial pressure (5 atm), the rates of equivalence ratio produced are insufficient to provide enhanced mixing. Given a low initial pressure, rates of pressure of more than 50 atm/s can cause flame quenching. For an initial pressure of 5 atm and rates of pressure between -40 and 0 atm/s, the rates of equivalence ratio are positive in fuel-rich flames and negative in fuel-lean flames. Therefore, the rate of equivalence ratio is zero for an equivalence ratio Φ_{eq} that is at a position of equilibrium and has a value between 0.5 and 1.68. A slight perturbation in the rate of pressure yields an equivalence ratio and rate of equivalence ratio of $\Phi(t) = \Phi_{eq} + \delta$ and $r(t) \neq 0$, respectively. The equivalence ratio $\Phi(t)$ and the rate of equivalence ratio r(t) satisfy the condition given by inequality (19). Therefore, Φ_{eq} is at a position of unstable equilibrium. Figure 2 shows that the position of equilibrium can be shifted toward higher equivalence ratios by reducing the rate of pressure or toward lower equivalence ratios by increasing the rate of pressure. Fuel-lean flames that are particularly unstable would benefit from the use of a positive rate of pressure. For a initial pressure of 1 atm, rates of pressure of more than about 1 atm/s provide rates of equivalence ratio that are either positive or negative in both fuel-lean and fuel-rich mixtures. For such conditions, there is no position of equilibrium and flame quenching may occur.

Enhanced mixing in flame and flame stability are mutually exclusive for rates of pressure that act alone. For fuels with a molecular weight smaller than that of air, negative rates of pressure in fuel-rich flames and positive rates of pressure in fuel-lean flames can help enhance preferential diffusion of oxygen and fuel, respectively. This then promotes both mixing and flame stability when used in conjunction with rates of temperature, heat transfer, and/or velocity di-

vergence. Fuel-rich mixtures should not be exposed to positive rates of pressure because positive rates enhance preferential diffusion of fuel that will then add to rates of quenching, that is, positive in this case. Negative rates of pressure are not recommended in fuel-lean flames because they enhance preferential diffusion of oxygen. Rates of pressure exceeding 30 atm/s and an initial pressure of 1 atm provide rates of quenching of more than $1000 \, \text{s}^{-1}$ and, therefore, cause flame extinction. For rates of pressure exceeding 30 atm/s, rates of equivalence ratio are lower at an initial pressure of 5 atm/s. The result shows that if the rates of pressure must be increased, flame stability can be improved by choosing a higher initial pressure.

Im et al.21 studied the correlation between sound characteristics (amplitude and frequency) and flame quenching. Nevertheless, the effect of sounds on mixing and flame stability was not completely understood. Seagrave et al.²² and Mcmanus et al.²³ reported a reduction of NO_r emission level, an increase of unburned hydrocarbons (UHC) emissions, and increased volumetric energy release in flame. They also provide an indication on flame quenching with sound effects at frequencies up to 500 Hz. The new flame model presented here can easily explain such observations. Sound waves create rates of pressure that are alternately positive and negative.⁴ Standing waves may enhance mixing in the flame and, thereby, combustor performance. In contrast, depending on the distribution of pressure caused by sound waves, high-amplitude and/or highfrequency sound waves may not allow the equivalence ratio to be maintained near the desired value. This may then cause local flame quenching.

Figure 3 shows the effect of temperature on the flame. The rate of equivalence ratio is shown to be proportional to the rate of temperature. For an initial temperature of 1050 K, the rate of equivalence ratio is less than 15 s⁻¹ (a value that is negligible), when the rate of temperature is zero (see Fig. 1), a value that is negligible. For an initial temperature of 1050 K and equivalence ratios of 0.50 and 1.68, a rate of temperature of 2.0×10^7 K/s provides rates of equivalence ratio of about -9×10^5 and -3.2×10^5 , respectively. A rate of temperature of -2×10^7 K/s provides rates of equivalence ratio of about 9×10^5 and 3.2×10^5 , for extreme range equivalence ratios of 0.5 and 1.68. Sustained rates of equivalence ratio having a magnitude of more than $5 \times 10^5 \text{ s}^{-1}$ provide mixing and flame quenching times of less than 2 μ s. For a significant rate of temperature (greater than about 50,000 K/s) and an initial temperature of 1050 K, positive rates of temperature provide negative rates of mixing in both fuellean and fuel-rich mixtures. Negative rates of temperature provide positive rates of equivalence ratio in both fuel-lean and fuel-rich

If the initial temperature is 2250 K, a rate of temperature of 2×10^7 K/s provides rates of equivalence ratio of about -5.5×10^5 and 0 for initial equivalence ratios of 0.5 and 1.68, respectively. For any rate of temperature between about -6×10^6 and 2×10^7 K/s, the rate of equivalence ratio is positive for an equivalence ratio of 1.68 and negative for an equivalence ratio of 0.5. Therefore, there is an equilibrium equivalence ratio in between 1.68 and 0.5. This equivalence ratio and the rates of equivalence ratio that occur in its

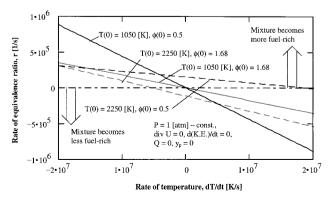


Fig. 3 Effect of temperature and rate of temperature on mixing in flame and flame quenching.

vicinity satisfy the inequality given in Eq. (19) and, therefore, the equilibrium position is unstable. The position of equilibrium can be shifted toward more fuel-rich or more-fuel mixtures by increasing or decreasing the rate of temperature, respectively.

The unstable position of equilibrium suggests that enhanced mixing causes flame quenching. Higher initial temperatures may be used to reduce quenching rates under higher rates of temperature.

Rates of temperature are expected to be positive, at least in the early stages of combustion, due to exothermic chemical reaction. As a consequence, the rate of equivalence ratio is negative causing the equivalence ratio to drop below the fuel-lean flammability limit. Therefore, positive rates of temperature cause fuel-lean flames to be more susceptible to flame quenching. However, when used together with other factors, positive rates of temperature may help increase the time in which a fuel-rich mixture becomes nonflammable by enhancing the preferential diffusion of oxygen.

Rapid cooling of flame, obtained, for example, when the mixture comes in contact with a cold metallic surface, can produce a negative rate of temperature having high magnitude. If the magnitude exceeds $6 \times 10^6\,$ K/s, rates of equivalence ratio are positive in both fuellean and fuel-rich flames. In the absence of other factors that might reduce the rate of equivalence ratio; there is no position of equilibrium for any equivalence ratio. Flame quenching should then be expected. Again, combined with other factors, negative rates of temperature may be used to enhance the net transport of fuel in fuel-lean flames.

The effect of heat transfer through radiation and conduction on flame is shown in Fig. 4. The intensity of heat transfer through conduction and radiation is given as a percent value of the rate of energy released by combustion. The rates of heat transfer and energy released by combustion are given per unit mass of fluid. The rate of equivalence ratio is proportional to intensity of heat transfer. In general, heat transfer is associated with a rate of temperature.³ When mixing is due to both heat transfer and rate of temperature, the overall rate of equivalence ratio is the sum of rates of mixing provided by heat transfer and rate of temperature. For a rate of heat loss of 150%, the rates of equivalence ratio for initial equivalence ratios of 1.68, 1.0, and 0.5 are about 1400, -0.2, and 5 s⁻¹, respectively. For heat addition through conduction or radiation having a rate of 150%, the rates of equivalence ratio produced by heat transfer are about -1400, 0.8, and -37 s^{-1} for the same equivalence ratios. These rates of equivalence ratio become much larger for higher initial temperature, but general features of the results shown in Fig. 4 remain the same. Fuel-lean flames are less sensitive to heat addition. Rates of equivalence ratio due to heat addition are positive in a stoichiometric mixture and negative for $\Phi = 1.68$, respectively. Therefore, the rate of equivalence ratio must be zero for values of equivalence ratio between 1.0 and 1.68. This equivalence ratio may be at a stable position of equilibrium if the rate of temperature associated with heat addition is low and has little effect on the rate of equivalence ratio (see Fig. 3). Heat transfer values of 100-150% of the rate of energy released by combustion provide a position of unstable equilibrium for fuel-lean flames. Figure 4 shows that a rate of heat loss exceeding the rate of energy released by combustion

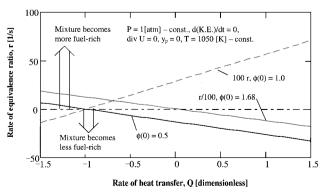


Fig. 4 Effect of heat transfer on mixing in flame and flame quenching.

can also provide a stable position of equilibrium for an equivalence ratio value between 0.5 and 1.0. However, such a high rate of heat loss has to be associated with rapid cooling of fluid. Therefore, the overall rate of equivalence ratio is much higher than that shown in Fig. 4, and there is no position of equilibrium. In addition, cooling alone may quench the flame, because the rates of chemical reactions are negligible at low temperatures.

Gottenberg et al.²⁴ have shown that UHC released into the atmosphere is produced by the flame quenching process that occurs near the combustor wall. Near to the combustor wall, conduction will cause rapid heat loss. As mentioned by the authors of Ref. 24, many previous studies were made to determine the factors that affect flame quenching under idealized conditions, that is, laminar flame and low pressure. The flame model presented here allows one to study flame quenching near wall over a large range of pressures and turbulence levels. It also explains why flame quenching is reduced at higher pressures (higher pressures reduce the overall rate of equivalence ratio, see Fig. 2). In addition it explains why flame quenching is reduced in mixtures having a higher specific heat at constant pressure (due to the higher specific heat, the rate of temperature and, therefore, the rate of quenching are reduced, see Fig. 3). Predictions based on the model presented here also support the observation that higher thermal conductivity and flame temperature enhance flame quenching near walls.²⁴ As shown in Fig. 2, under reduced rates of pressure and velocity divergence, the rates of quenching caused by a moderate rate of temperature are higher at higher initial temperatures (see Fig. 3). Increased thermal conductivity causes increased heat loss to increase the overall rate of quenching (see Fig. 4).

The time for a fluid element to remain in contact with the combustor wall can be reduced by increasing the fluid velocity. Therefore, a higher velocity can reduce the amount of heat that is lost and reduce the rate of quenching. The present flame model also shows that flame quenching near walls depends on the mass fraction of combustion products that are present in the flame, the local equivalence ratio, and the molecular weight of fuel. The less stoichiometric is the mixture, the faster is flame quenching with the wall contact.

The effect of mass fraction of combustion products on the flame is shown in Fig. 5. At an initial temperature of 1050 K, the effect of mass fraction of combustion products on flame is negligible. The high rates of quenching observed at an initial temperature of 2250 K (in excess of $100,000~\rm s^{-1}$) are reduced by 25–30% in burnt gases compared to the fresh mixture. This supports the general known fact that recirculation improves flame stability.

By themselves, usual rates of kinetic energy have a negligible effect on mixing and flame quenching. The effect of velocity divergence on the flame is shown in Fig. 6. Three equivalence ratios, 1.68, 1.0, and 0.5 are analyzed. The rate of equivalence ratio is proportional to velocity divergence. Velocity divergence of $-600 \, {\rm s}^{-1}$ provides rates of equivalence ratio of about -16,000,-10,000, and $-30,000 \, {\rm s}^{-1}$ for the three equivalence ratios of 1.68, 1.0, and 0.5, respectively (see Fig. 6). Velocity divergence of $600 \, {\rm s}^{-1}$ provides rates of equivalence ratio of about 16,000,10,000, and $30,000 \, {\rm s}^{-1}$ for the aforementioned equivalence ratios (see Fig. 6). Therefore, velocity divergence can be used to compensate the undesirable effect

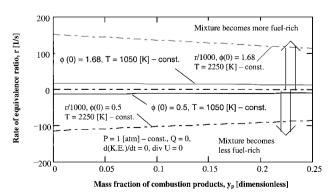


Fig. 5 Effect of mass fraction of carbon dioxide and water on mixing in flame and flame quenching.

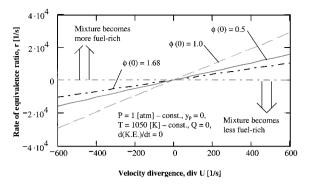


Fig. 6 Effect of velocity divergence on mixing in flame and flame quenching.

of a positive rate of pressure in a fuel-rich mixture or negative rate of pressure in a fuel-lean mixture (compare Figs. 2 and 6). To a lesser extent, velocity divergence can be used to compensate the effect of rate of temperature or heat transfer. Velocity divergence can help stabilize the flame only in conjunction with other factors. If the flame is subject only to velocity divergence, rates of quenching exceeding 1000 s⁻¹ may occur in both fuel-lean and fuel-rich mixtures (for a velocity divergence exceeding 100 s⁻¹) and flame quenching should be expected. The operational parameters should be chosen to achieve an optimum compromise between the requirements of 1) reducing the mixing time in flames and 2) maximizing the quenching time. If a position of stable equilibrium cannot be achieved, quenching time is increased by those factors that enhance preferential diffusion of fuel or oxygen in fuel-lean or fuel-rich flames, respectively. High levels of turbulence may increase the velocity divergence in an uncontrollable manner. As a consequence, the flame may wrinkle or even break up into small individual flamelets.³ The flame model presented here explains such a behavior.

VI. Conclusions

Precise control over fuel-air mixing must be assured for increased combustor efficiency and reduction of pollutants emission level. Before achieving the desired equivalence ratio, mixing has to be enhanced. In flames, enhanced mixing is provided by levels of preferential diffusion of oxygen or fuel, in fuel-rich or fuel-lean flames, respectively, that exceed by far the levels required to compensate the unbalanced consumption of fuel and oxygen. Enhanced mixing is shown by a high rate of equivalence ratio. Preferential diffusion is shown to be associated with rates of pressure or temperature, velocity gradients, or heat transfer through radiation and conduction. Positive rates of pressure, negative rates of temperature, negative velocity divergence, and heat addition through conduction and radiation enhance preferential diffusion of oxygen. In fuel-lean flames, heat loss also enhances preferential diffusion of oxygen. Negative rates of pressure, positive rates of temperature, and positive velocity divergence enhance preferential diffusion of fuel. In fuel-rich flames, preferential diffusion of fuel is also enhanced by heat loss. Therefore, control over the equivalence ratio can be achieved by a suitable combination of heat transfer, velocity gradients, and rates of pressure and temperature over an extended range of operational conditions. High rates of equivalence ratio required during mixing are not automatically reduced after achieving the desired equivalence ratio and this can lead to flame quenching. High pressure or temperature provided by high rates of pressure or temperature, respectively, reduces the rate of equivalence ratio. As combustion progresses, the mass fraction of combustion products increases, which further reduces the rates of equivalence ratio. In addition, velocity divergence, heat transfer, rates of pressure, and velocity must be reduced to levels according to the combustor design requirements.

Acknowledgments

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