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Ignition and extinction in perfectly stirred reactors with detailed chemistry

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ABSTRACT

Ignition and extinction of steady state combustion are known to be associated with the lower and upper turning points on the "S"-curves. In the present study, this concept is further investigated with eigen-analysis on the Jacobian matrix for oxidation of methane and dimethyl ether (DME), respectively, in perfectly stirred reactors (PSRs). It was found that there can be multiple ignition and extinction turning points on the "S"-curves for DME–air due to negative temperature coefficient (NTC) behaviors. Furthermore, the physical extinction points for DME–air obtained from flame stability analysis can be different from the turning points on the "S"-curves although there is no differential diffusion or heat loss in PSR. Physically unstable segments were observed on the branches of an "S"-curve for DME–air corresponding to both strong and cool flames. A rigorous definition of ignition and extinction of steady state combustion based on eigen-analysis of the Jacobian matrix is proposed for practical fuels in the present study.

1. Introduction

Ignition and extinction are important limit phenomena in combustion research. For steady state systems, the ignition and extinction states were conventionally defined as the turning points on the "S"-curve as shown by the schematics in Fig. 1 [1]. The upper and the lower branches of the "S"-curve are known to be stable. The upper branch of the "S"-curve indicates strongly burning flames, the lower branch indicates weak reactions without identifiable flames, and the middle branch is known to be unstable and cannot be obtained experimentally. The upper turning point is commonly regarded as the extinction state of strong flames because a strong flame cannot be sustained with further reduced Damköhler number (*Da*) or residence time. Similarly, the lower turning point is commonly known as the ignition state since the steady state solution will jump to the upper branch with further increased *Da* or residence time. The limit phenomena associated with the turning points on the "S"-curves are important in combustion research. Experimentally measured extinction and ignition states were routinely compared with numerically obtained turning points on the "S"-curves, e.g. using the arc-length continuation method [2,3], for validation and improvement of detailed chemical kinetics [4–12].

However, while the "S"-curve in Fig. 1 correctly describes the responses of many combustion systems to the variation in flow straining or mixing, it was found that multiple criticalities may exist for practical fuels such as dimethyl ether (DME) and *n*-heptane [8,13,14]. As such, the conventional definition of ignition and extinction states based on the turning points may not be applicable for such practical fuels. Thus a new definition based on flame stability is proposed in the present study.

Linear flame stability is determined by the eigenvalues of the Jacobian matrix based on the steady state solutions. The Jacobian is important for combustion analyses involving detailed chemistry, e.g. in computational singular perturbation (CSP) [15,16], intrinsic low dimensional manifold (ILDM) [17], and chemical explosive mode analysis (CEMA) [18,19]. On the "S"-curves of a steady state system, a turning point is known to be associated with a singular Jacobian matrix [20]. As such, a turning point can separate a stable branch without positive eigenvalues, e.g. the upper and the lower branches in Fig. 1, from an unstable branch with a positive eigenvalue, e.g. the middle branch in Fig. 1. Nevertheless, in cases where the eigenvalues are complex numbers, flame stability is determined only by the sign of the real part of the eigenvalues, while the imaginary parts of the eigenvalues determine the oscillation frequencies [21,22]. In such cases, transition between a stable and an unstable flame may no longer occur at a turning point. Furthermore, oscillations, or flame instabilities, may occur near the transition. For example, flame instabilities were observed in perfectly stirred reactors (PSRs) with near-limit flames for hydrogen and small hydrocarbons with heat losses either promoting or suppressing the oscillations [23,24]. Flame oscillations were also observed in PSR for diluted methane–air at low temperatures [25]. However, flame instabilities involving detailed chemistry of practical fuels, particular those involving negative temperature coefficient (NTC) behaviors, are relatively less understood.

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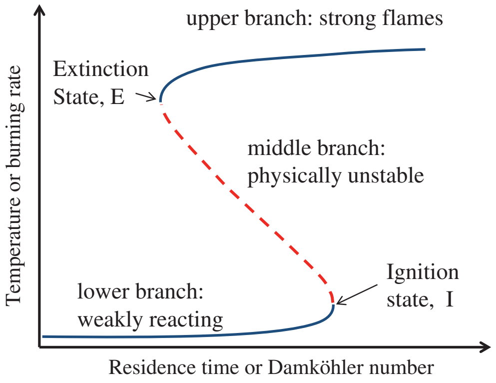

Fig. 1. Representative folded S-curve with multiple solutions and distinct ignition and extinction states [1].

Nonlinear flame stability analyses involve high order expansion of the governing equations, thus can typically only be performed with simplified chemistry with one or a few steps and transport characterized by a Lewis number. Flame extinctions due to nonlinear instabilities prior to the turning points have been observed in previous works [26–28]. Numerical studies with detailed chemistry and transport on pulsating instability induced by thermal-diffusive couplings have been conducted for premixed flames with large Lewis numbers [29–31]. It was found that the flame instability may promote extinction and the experimentally determined extinction states may not necessarily agree with the numerically obtained turning points on “S”-curves [31]. The numerical prediction of flame instabilities in Refs. [29–31] however involved time integration of unsteady flames that is time consuming, thus is difficult to apply for parametric studies. In the present study, linear flame stability analyses will be employed to study the extinction and ignition of steady state combustion involving detailed chemistry of practical fuels with NTC behaviors, and PSR is selected as a representative steady state combustion system that can be described by an “S”-curve similar to that in Fig. 1. Note that a PSR features a unity Lewis number since it only involves a single mixing timescale, that is, the residence time. Nevertheless, the results of flame stability for PSR may be extended in the future to the study of diffusive systems, such as the counterflow flames.

2. Methodologies

The equations of time evolving PSR can be written as

$$\frac{d\mathbf{y}}{dt} = \mathbf{g}(\mathbf{y}) = \omega(\mathbf{y}) + \mathbf{s}(\mathbf{y}), \quad \mathbf{y} = [Y_1,\ Y_2, \ldots Y_K,\ T]^T, \tag{1a}$$

$$\omega_i(\mathbf{y}) = \frac{\dot{m}_i}{\rho}, \quad i = 1, 2, \ldots, K, \quad \omega_{K+1}(\mathbf{y}) = -\frac{\sum_{i=1,K} \dot{m}_i h_i}{\rho c_p}, \tag{1b}$$

$$s_i(\mathbf{y}) = \frac{Y_i^0 - Y_i}{\tau}, \quad i = 1, 2, \ldots, K, \quad s_{K+1}(\mathbf{y}) = \frac{\sum_{i=1,K} Y_i^0 (h_i^0 - h_i)}{c_p \tau}, \quad \tau = \frac{\rho V}{\dot{M}_{in}}, \tag{1c}$$

where **ω** is the chemical source term and **s** is the mixing term. **y** is the vector of dependent variables including species mass fractions, **Y**, and temperature *T*. ρ is density, $\dot{m}_i$ is the volumetric mass production rate of the *i*th species, *h* is the specific enthalpy, *V* is the volume of the reactor, $\dot{M}_{in}$ is the inlet mass flow rate, *K* is the total number of species, subscript *i* indicates the *i*th species, and the superscript “0” indicates the inlet condition. The solution of a steady state PSR is characterized by an “S”-curve similar to that in Fig. 1. To show the stability of a specific state on the “S”-curve of a steady state PSR, the unsteady equations can be integrated to show the response of the system to small perturbations made to the steady state solution.

Based on linear stability analysis of ODE, the growth of a small perturbation, $\delta\mathbf{y}$, initially superposed to the steady state solution of Eq. (1) can be approximated with

$$\frac{d\delta\mathbf{y}}{dt} \approx \mathbf{J_g}(\mathbf{y}_0) \cdot \delta\mathbf{y}, \quad \delta\mathbf{y} = \delta\mathbf{y}_0 \text{ at } t = t_0, \tag{2}$$

where $\mathbf{y}_0$ is the steady state solution, and $\mathbf{J_g}$ is the Jacobian matrix defined as

$$\mathbf{J_g} = \mathbf{J}_{\boldsymbol{\omega}} + \mathbf{J_s} = \frac{\partial \mathbf{g}(\mathbf{y})}{\partial \mathbf{y}}, \quad \mathbf{J}_{\boldsymbol{\omega}} = \frac{\partial \boldsymbol{\omega}(\mathbf{y})}{\partial \mathbf{y}}, \quad \mathbf{J_s} = \frac{\partial \mathbf{s}(\mathbf{y})}{\partial \mathbf{y}}, \tag{3}$$

It is seen that $\mathbf{J_g}$ is comprised of two components, namely $\mathbf{J}_{\boldsymbol{\omega}}$ for the chemical source term and $\mathbf{J_s}$ for the mixing term, respectively. It is noted that the chemical Jacobian describes the chemical properties of a mixture and can be employed to distinguish between the pre- and post-ignition mixtures in CEMA [18,19]. In the present work, the full Jacobian matrix, $\mathbf{J_g}$, will be employed for flame stability analysis for PSR using two representative fuels, methane and dimethyl ether (DME), respectively. Methane is a hydrocarbon fuel without NTC behaviors and DME features NTC behaviors that involve low temperature chemistry.

Using similarity transformation $\mathbf{J_g}(\mathbf{y}_0) = \mathbf{A\Lambda B}$, Eq. (2) can be written as

$$\frac{d\delta\mathbf{f}}{\mathrm{dt}} = \boldsymbol{\Lambda} \cdot \delta\mathbf{f}, \quad \delta\mathbf{f} = \mathbf{B} \cdot \delta\mathbf{y}, \tag{4}$$

where **A** and **B** are matrices that consist of the right and left eigenvectors, respectively. $\boldsymbol{\Lambda}$ is a diagonal matrix with the diagonal elements being the eigenvalues of $\mathbf{J_g}(\mathbf{y}_0)$. Based on Eq. (4), the growth of a small perturbation in the direction of the *i*th eigenvector can be approximated as

$$\delta f_i \approx \delta f_{i,0} e^{\lambda_i t}, \quad \delta f_i = \mathbf{b}_i \cdot \delta\mathbf{y}, \tag{5}$$

where $\mathbf{b}_i$ is the *i*th row in **B**, that is, the *i*th left eigenvector of $\mathbf{J_g}(\mathbf{y}_0)$. The perturbation tends to exponentially grow if the real part of an eigenvalue is positive, or to decay if the real parts of all the eigenvalues are negative. A bifurcation point, where $\mathrm{Re}(\lambda_i) = 0$, may separate a stable and an unstable segment on the “S”-curve. Note that “Re()” indicates the real part of a complex number in the present study. As such, the flame stability may change not only at a turning point, where $\lambda_i = 0$, but also at other types of bifurcation, e.g. the Hopf bifurcation where the imaginary part of the eigenvalue is non-trivial [21]. While the turning points can be readily located on the “S”-curves, the identification of a Hopf bifurcation requires eigen-analysis that will be demonstrated with PSR for DME–air in the next section.

To further identify the controlling chemical species and reaction pathways at a bifurcation point, the contribution of a species to the mode involving eigenvalue crossovers can be quantified by the species pointer (SP), defined similarly to the explosion index in CEMA [18,19] and the radical pointer in CSP [15,16]. The contribution of a reaction to a mode can be quantified by the participation index (PI) defined in CSP [15,16]. If oscillatory modes associated with complex conjugate eigenvalues are of interest, the values of SP and PI can be expressed similarly to the radical pointer and PI in Ref. [32] for complex CSP. Specifically, if $\mathbf{a}^i$ and $\mathbf{b}^i$ are complex eigenvectors, i.e.

$$\mathbf{a}_i = \mathbf{a}_{re}^i + \mathbf{a}_{im}^i, \quad \mathbf{b}_i = \mathbf{b}_{re}^i + \mathbf{b}_{im}^i, \tag{6}$$

where the subscripts *re* and *im* indicate the real and imaginary parts of the complex eigenvectors, respectively, the SP can be defined as