

Spatial and Temporal Scales Coupling in Reactive Flows

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Abstract— We investigate the eigenvalue spectrum of the one-dimensional laminar premixed flame in a reactive mixture described by detailed mass-action kinetics. All the physical scales, spatial and temporal, inherent in a reacting system are accurately identified via eigenvalue analysis. The interplay between chemistry and transport is addressed via conducting a spectral analysis of the reactive flow structure. It revealed that reacting systems' physical scales are coupled, the systems short wavelength modes are dominated by diffusion-based scales, coarse wavelength modes are dominated by reaction-based scales, and modes near a cross-over wavelength have scales dictated by a combination of reaction and diffusion effects.

I. INTRODUCTION

Simulating reactive flow involves solving a large set of partial differential equations (PDEs). For combustion problems which are inherently unsteady and spatially inhomogeneous, the dynamics are crucial. A common notion in combustion theory is that chemical dynamics are somehow segregated from the dynamics of advection and diffusion; this notion is especially prevalent in discussion of so-called operator splitting strategies for numerical simulation of combustion events. In reality, unsteady, spatially inhomogeneous combustion is better viewed as an event in which reaction, advection, and diffusion time scales are often fully coupled.

For accurate modeling, the interplay between chemistry and transport needs to be captured. One way to gain a better understanding of the coupling between transport and chemistry can be achieved via conducting a spectral analysis of a plane laminar flame structure. It is important in a spectral analysis to guarantee that all length scales in the underlying steady structure problem have been brought into simultaneous focus. In recent studies [1], it has been shown that the finest length scale for an atmospheric-pressure laminar premixed flame is typically on the order of 10^{-4} cm.

This work will consider a premixed mixture of calorically imperfect ideal gases that react and diffuse at widely disparate rates. For this problem, we specifically consider a model of hydrogen-air combustion.

II. ANALYSIS

We consider equations modeling a one-dimensional unsteady laminar premixed flame propagating freely in a mixture of N molecular species composed of L atomic elements which undergoes J reversible reactions with no body force present. A detailed kinetics mechanism and

multi-component transport model are used to describe the reactive mixture. The system is simplified by neglecting the thermal diffusion effects, Soret's effect and Dufour's effect, and adopting the low-Mach number assumption [2]. This system, in a compact form, is

$$\mathbf{A}(\mathbf{z}) \cdot \frac{\partial \mathbf{z}}{\partial t} + \mathbf{B}(\mathbf{z}) \cdot \frac{\partial \mathbf{z}}{\partial x} = \mathbf{f}(\mathbf{z}), \quad \mathbf{z} \in \mathbb{R}^{2N+2}, \quad (1)$$

where \mathbf{A} and \mathbf{B} are matrices of size $(2N + 2) \times (2N + 2)$ and the $2N + 2$ state variables contained in \mathbf{z} are composed of species mass fraction, species mass flux, mixture specific enthalpy, and Fourier heat flux. To identify all the time and length scales inherent in this reactive flow's model, the problem is split into two separate problems that are treated independently: 1) an unsteady spatially homogeneous reactive system, and 2) a steady spatially inhomogeneous reactive system. Then, the coupling between the scales from the two parts is explored via conducting spectral analysis for the complete reactive flow.

For unsteady spatially homogeneous mixtures of calorically imperfect ideal gases described by detailed kinetics, the governing equations are reduced into a set of $N + 1$ non-linear autonomous ordinary differential equations (ODEs) which exhibit a temporal stiffness, \mathcal{S}_t . A standard computational scheme is used to obtain the time evolution of the system [3]. Linearization of the equations about the local solution at each time step gives rise to a standard eigenvalue problem which in turn provides full information on all the $N - L$ time scales, τ 's, contained in the problem.

For the steady spatially inhomogeneous reactive system, the complete system is reduced into a coupled system of $2N + 2$ differential-algebraic equations (DAEs) which exhibits a spatial stiffness, \mathcal{S}_x . The method introduced in [1] to obtain a resolved steady flame structure and to calculate the $2N - L$ physical length scales, ℓ 's, inherent in the problem is employed.

Next, the time spectrum of the full reacting flow system is studied. To this end, the governing equations for the unsteady spatially inhomogeneous reactive system are posed as a set of $2N + 2$ partial differential algebraic equations (PDAEs). A perturbation from the steady structure is introduced to (1). Linearization of the resulting system about the steady structure is performed. Spatial discretization of the spatial derivative operators gives rise to a generalized eigenvalue problem. Each time scale,

given by the reciprocals of the generalized eigenvalues, is associated with a particular Fourier mode of wavenumber κ , which has a wavelength Λ . For numerical results, instead of Λ , the modified wavelength $\hat{\Lambda}$ is employed, which is defined based on the eigenvectors' number of zero crossings n , *i.e.* normal mode nodes,

$$\hat{\Lambda} = \frac{4L}{2n-1}, \quad n = 1, 2, 3, \dots, \quad (2)$$

where L is the spatial length.

III. RESULTS

A stoichiometric atmospheric pressure hydrogen–air mixture is considered, where the initial molar ratio is given by $2H_2 + (O_2 + 3.76N_2)$. A kinetic model identical to that of Smooke *et al.* [4], with $L = 3$ elements, $N = 9$ species, and $J = 19$ reversible reactions is used. In this mechanism, the reactive species are $H_2, O_2, H, O, OH, HO_2, H_2O_2$, and H_2O . The inert diluent for the mixture is N_2 .

A. Temporal scales

For the unsteady spatially homogeneous stoichiometric hydrogen–air system, under adiabatic and isobaric conditions, and initially at temperature $T^* = 800$ K, the evolution of species in time is determined. Simultaneously, the system's temporal scales, given by the reciprocal of the eigenvalues, over which the system evolves are calculated. There are $N - L = 6$ independent modes; six τ 's in the spectrum. Near equilibrium, the slowest time scale is 1.85×10^{-4} s, and the fastest time scale is 1.03×10^{-8} s, giving rise to $\mathcal{S}_t \sim \mathcal{O}(10^4)$. Thus, to capture all the physical dynamics in such a numerical simulation $\Delta t < \tau_{fastest} \approx 10^{-8}$ s, needs to be employed.

B. Spatial scales

The stationary structure of the one-dimensional, stoichiometric, adiabatic, hydrogen–air laminar premixed flame with unburned mixture's temperature of $T_u = 800$ K is determined. Having the fully resolved structure in hand, the local spatial eigenvalues are calculated from the cold boundary to hot region. As a result, the local length scales ℓ 's are predicted throughout the domain. The finest length scale and the largest length scale found for this system vary from 7.60×10^{-4} cm and 1.62×10^7 cm in the preheat zone to 2.41×10^{-4} cm and 2.62×10^0 cm in the reaction zone, respectively. The spatial stiffness in the hot region is $\mathcal{S}_x \sim \mathcal{O}(10^4)$. Thus, to resolve the flame structure in such a numerical simulation a spatial resolution of $\Delta x < \ell_{finest} \approx 2 \times 10^{-4}$ cm, needs to be utilized.

C. Spatio-temporal spectrum

Now, instead of the steady laminar flame structure, which presents overwhelming computational demands in solving for eigenvalues, a system initially near a spatially homogenous chemical equilibrium state is perturbed. The unperturbed state is identical to the equilibrium state of the unsteady spatially homogeneous system. This is

certainly relevant for the laminar flame structure, as it represents the hot end. So, the spatially homogeneous system at chemical equilibrium is subjected to a spatially inhomogeneous perturbation, and its spatio-temporal response is predicted.

In Fig. 1, the system's times scales associated with the fundamental modes, *i.e.* eigenfunctions with $n = 1$, are tracked as we vary the system's length. For large L , the reaction-advection-diffusion system's time scales and the reaction-only system's time scales at equilibrium are identical. However, for $\hat{\Lambda}/(2\pi) \equiv 2L/\pi \sim 10^{-1}$ cm the effect of diffusion can be noted; it increases monotonically as L decreases. Also, the balance between reaction and diffusion is clear: short wavelength modes are dominated by diffusion, and large wavelength modes are dominated by reaction. Furthermore, the effect of adopting non-uniform diffusion coefficients, the multicomponent diffusion coefficients \mathcal{D}_{ij} , is noted in the time scale's falloff region, $L \leq 10^{-4}$ cm. One would expect $\tau \sim L^2/\mathcal{D}_{ij}$, and thus on the log-log scale, $\ln \tau \sim 2 \ln L - \ln \mathcal{D}_{ij}$, so that the slope of each should be the same, but the intercept is different for each \mathcal{D}_{ij} . It is obvious that in the diffusion-dominated region, there is a two decade drop in τ for every one decade drop in L , consistent with our prediction.

It is clear from Fig.1 that the branch associated with the slowest chemical time scales starts to become influenced by diffusion before branches associated with the faster chemical time scales; the turning point for the fastest chemical time scale branch is $2L/\pi \sim 10^{-3}$ cm and for the slowest chemical time scale branch is $2L/\pi \sim 10^{-1}$ cm. These turning points represent the length scale where diffusion starts to balance reaction.

Now we can also try to independently predict the turning points by employing an *ad hoc* formula to estimate the length scales,

$$\ell_1 = \sqrt{D_{mix}\tau_{slowest}}, \quad (3)$$

$$\ell_2 = \sqrt{D_{mix}\tau_{fastest}}, \quad (4)$$

where $\tau_{slowest}$ and $\tau_{fastest}$ are, respectively, the slowest and fastest time scales of the unsteady spatially homogeneous version of the problem, and D_{mix} is the mixture average diffusion coefficient. This is subject to greater error because we actually have a multicomponent diffusion process, coupled with diffusion of energy as well. The mixture average diffusion coefficient D_{mix} is estimated and it taken to be the average of the species mass diffusion coefficients,

$$D_{mix} = \frac{1}{N^2} \sum_{i=1}^N \sum_{j=1}^N \mathcal{D}_{ij}. \quad (5)$$

As a result, we estimate the turning points for fast and slow reactions to be

$$\ell_1 = 1.06 \times 10^{-1} \text{ cm}, \quad (6)$$

$$\ell_2 = 7.94 \times 10^{-4} \text{ cm}, \quad (7)$$

where $\tau_{fastest} = 1.03 \times 10^{-8}$ s and $\tau_{slowest} = 1.85 \times 10^{-4}$ s. Both of these estimates, illustrated as dashed lines in Fig. 1, predict well the turning points.

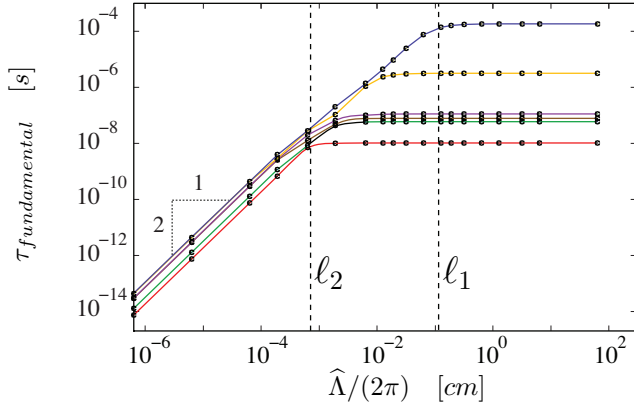


Fig. 1. Time scales associated with the fundamental modes for the hydrogen-air reactive system versus length scales.

For the more rigorous calculation of the system's finest length scale, Sec. III.B, it has been found that the finest length scale admitted by the steady spatially homogeneous version is $\ell_{\text{finest}} = 2.41 \times 10^{-4} \text{ cm}$. Interestingly, the simple estimate, Eq. (7), is close in magnitude to ℓ_{finest} obtained by spatial eigenvalue analysis. Subsequently, it is clear that the reactive systems' temporal and spatial scales are coupled, and for a resolved structure, Fourier modes of varying wavelength are associated with time scales which are dictated by a balance between transport and chemistry.

IV. CONCLUSION

The time scale spectrum of a one-dimensional atmospheric-pressure hydrogen-air system was calculated via conducting a generalized eigenvalue analysis. It was shown that when the reaction zone structure is resolved, the small wavelength modes critical in the thin reaction zone structures induced by fast reaction have associated with them time scales which are dictated by a balance between chemistry and transport. Moreover, it was revealed that short wavelength modes have very fast time scales which are dominated by diffusion, modes which have wavelengths ranging from the finest combustion length scale to the coarsest combustion length scale have time scales which are dictated by a combination of reaction and diffusion effects, and modes which have coarse wavelengths have time scales which are reaction dominated. These results have been achieved by conducting a spectral analysis of onedimensional premixed reactive mixtures of calorically imperfect ideal gases.

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