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ON NUMERICAL RESOLUTION REQUIREMENTS IN COMBUSTION MODELING

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ABSTRACT

We discuss one-dimensional steady laminar premixed flames in a mixture of calorically imperfect ideal gases described by detailed kinetics and multi-component transport. The required spatial discretization to capture all detailed continuum physics in the reaction zone is determined through use of a robust method developed to rigorously calculate the finest length scale a posteriori. This is accomplished by reformulating the governing equations as a nonlinear system of differential algebraic equations. Then, the solution of the steady reaction zone structure is obtained, and the generalized eigenvalues of the locally linearized system are calculated at each point in the reaction zone. Their reciprocals provide all local length scales. Application of the method to laminar flames reveals that the finest length scale is on the order of 10^{-4} *cm*. Independent estimates from grid convergence studies on the continuum equations as well as from the underlying molecular collision theory verify the result. This finest length scale is orders of magnitude smaller than common engineering geometric scales, the discretization scales employed in nearly all multi-dimensional and/or unsteady laminar premixed flame simulations in the literature, and the flame thickness.

1 Introduction

In recent years, there have been orders of magnitude enhancements in computational capabilities, enabled by improvement in both hardware and software, which have spurred the scientific and engineering community to employ mathematical models to solve challenging physical problems. Some of the most difficult of these are of multi-scale nature; such problems are characterized by physics which evolve over a wide range of spatial and temporal scales. To have confidence in the results, and to guarantee that they can be repeated by other researchers with their own particular algorithms, predictions should be accompanied by evidence that all physical scales inherent in the mathematical model have been captured.

It is well recognized that most detailed kinetic systems contain a broad range of scales, and the ratio between the largest and smallest scale is a measure of the system's stiffness. In such problems, proper numerical resolution of all scales can be critical to drawing the correct conclusions. The most effective means to determine the necessary spatial resolution for an unsteady problem is to focus attention on a highly resolved baseline steady problem from which future unsteadiness develops.

There is some ambiguity in the combustion literature about what constitutes a resolved solution. Most consider a calculation to be resolved if certain global or derived quantities, such as steady flame speed, are insensitive to grid size. Indeed, these are necessary conditions. However, as discussed by Roache [1], convergence of global quantities only is not a sufficient indicator of a fully resolved solution, and taken alone can lead to incorrect conclusions. While a derived quantity may be a function of the dependent variables, it may be insensitive to errors in some of them. Which variables they are insensitive to is problem-dependent, and impossible to determine *a priori*.

Here then, we follow Roache [1] and adopt the more rigorous characterization of a resolved solution as one in which *all dependent variables* throughout the spatio-temporal domain are insensitive to changes in spatio-temporal discretization size. This

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characterization is fully consistent with standard notions found in the broader mathematical and scientific computing literature, cf. Stoer and Bulirsch [2].

It is well understood that in any mathematically based scientific theory, associated computations should have fidelity with the underlying mathematics, and the underlying mathematical model has to represent the observed physics. The first issue is demonstrated by comparing computational results with another known solution and/or performing a formal grid convergence study, while the second issue is demonstrated by comparing the computational predictions with experimental data. Addressing these two issues, in this order, is a necessity in any computational study to build confidence in both the simulation strategy and the underlying mathematical model.

The exercise of demonstrating the harmony of the discrete solution with the foundational mathematics is known as verification [3]. Neglecting this issue can give rise to solutions whose behavior depends on the size of the grid and the algorithm that has been used to solve the mathematical model. For multi-scale problems, verification is difficult due to the range of the spatio-temporal scales present, which may span many orders of magnitude. In this type of problem, usually modeled by highly non-linear equations, significant coupling across scales can occur, so that errors at small scales can rapidly cascade to the larger scales. Moreover, the strength of the coupling across the scales is not known *a priori*. So, all physical scales of the mathematical model, temporal and spatial, have to be captured in order to have full confidence that predictions are repeatable, grid-independent, and thus verifiable. Subsequently, in the validation step one can choose what physical phenomena and to what accuracy one wants to reproduce experiments.

In this paper, we summarize results presented in detail by the authors [4]. The main aim of this paper is to discuss the required spatial resolution to capture all physical scales in a standard multi-scale problem: the steady one-dimensional laminar premixed flame propagating freely at atmospheric pressure in a stoichiometric mixture of hydrogen–air described by detailed kinetics and multi-component transport.

In a complementary study, we have developed a robust method to provide an accurate determination of the finest length scale in the reaction zone of a Chapman–Jouguet detonation [5]. Here, the method of Ref. [5] is implemented with modification for deflagration. The method is robust in that it has little dependence on the details of the underlying numerical method used to calculate the laminar flame. It simply requires a local determination of the state of the system, which is followed by a Jacobian formulation, and a generalized eigenvalue analysis. As such, it is able to estimate with great accuracy the length scales on a fundamental mathematical, *non-numerical*, basis.

2 Mathematical model

We describe here, in truncated form, the mathematical model. Full details are given in Ref. [4].

2.1 Governing equations

We study a standard model for stationary laminar premixed flames evolving in $i = 1, \ldots, N$ molecular species, cf. Smooke, *et al* [6], Kee *et al.* [7], Miller *et al.* [8]:

$$\frac{d}{dx}(\rho u) = 0, \tag{1}$$

$$\rho u c_p \frac{dT}{dx} + \frac{dq}{dx} + \sum_{i=1}^{N} \left(J_i^m \frac{dh_i}{dx} + \dot{\omega}_i M_i h_i \right) = 0, \tag{2}$$

$$\rho u \frac{dY_i}{dx} + \frac{dJ_i^m}{dx} = \dot{\omega}_i M_i. \tag{3}$$

The independent spatial variable is x. The dependent variables are density ρ, velocity u, mixture specific heat at constant pressure, c_p, temperature T, diffusive heat flux q, and for species i, diffusive mass flux J_i^m, specific enthalpy h_i, molar species production rate $\dot{\omega}_i$, and mass fraction Y_i. Here, M_i is the molecular mass of species i. An appropriate set of boundary conditions is

$$x = 0: \qquad T = T_o, \quad Y_i + \frac{J_i^m}{\rho_o S} = Y_{io}, \tag{4}$$

$$x \ \to \infty: \qquad \frac{dT}{dx} \to 0, \quad \frac{dY_i}{dx} \to 0, \tag{5}$$

$$x = x_f: \qquad T = T_f, \tag{6}$$

where x_f is a specified spatial point and T_f is the specified temperature at that location [7]. These are commonly used to study deflagration, though other formulations are possible. The boundary conditions are sufficient for freely propagating flames, where the mass flow rate is unknown [8], so that the temperature at an interior spatial point has to be specified. The point x_f and temperature T_f have to be selected such that all the gradients approach zero at the cold boundary. A solution for this boundary value problem, equations (1)–(3), with boundary conditions, equations (4)–(6), can be obtained by discretizing the spatial domain using finite differences, and the resulting algebraic system of equations are solved iteratively using a damped modified Newton's method, where the solution iterate is brought into the convergence domain by using pseudo time-step integration [9].

2.2 A posteriori length scale analysis

The governing equations can be conveniently posed as a set of $2N + 2$ Differential-Algebraic Equations (DAEs) in terms of

$2N+2$ state variables [4] of the form:

$$\mathbf{A}\cdot\frac{d\mathbf{z}}{dx}=\mathbf{f}. \tag{7}$$

To accurately determine the length scales over which the system evolves, an eigenvalue analysis can be applied to equation (7). Since **A** is singular, the standard eigenvalue analysis is not applicable. Instead, the generalized eigenvalues of this dynamical system are calculated [10].

Linearization yields the generalized eigenvalue problem

$$\lambda\mathbf{A}^*\cdot\mathbf{v}=\mathbf{B}^*\cdot\mathbf{v}, \tag{8}$$

where λ is in general a complex number denoting the generalized eigenvalue, and **v** is the corresponding generalized eigenvector [4]. Solving for λ_i, $i=1,\ldots,2N-L$, it is easily seen that the length scales over which the dependent variables evolve are given by the reciprocal of the real part of the eigenvalues,

$$\ell_i=\frac{1}{|Re(\lambda_i)|},\quad i=1,\ldots,2N-L. \tag{9}$$

By evaluating the eigenvalues at each spatial point, the length scales over which the system evolves through the reaction zone are determined. As a result, the minimum size of discretization to capture the finest scale of the system can be determined. In general, the eigenvalues are complex. In this work, the eigenvalues are purely real, except for limited regions.

3 Computational method

The resolved structure is obtained by solving equations (1)–(3), and the eigenvalues are calculated by using the dynamical system equation (7). A double precision code has been developed and linked with the `CHEMKIN` package to obtain kinetic rates, thermodynamic properties, and multi-component transport coefficients [11–13]. The `PREMIX` algorithm has been used to obtain the steady structure of adiabatic laminar premixed flames [9]. Except for systematic grid convergence studies discussed below, all results are obtained on a grid that is adaptively refined to capture regions of steep gradient. A second order central difference scheme is employed to discretize all spatial derivatives. The mass and heat fluxes are estimated at intermediate grid points to maintain second order accuracy.

4 Results

A stoichiometric hydrogen–air mixture at $p_o=1$ atm has been considered, where the initial molar ratio is given by $2H_2+O_2+3.76N_2$. Kinetics identical to those of Smooke *et al.* [6], with $L=3$ elements, $N=9$ species, and $J=19$ reactions is used. The reactant species are $H_2, O_2, H, O, OH, HO_2, H_2O_2$, and H_2O. The inert diluent is N_2.

4.1 Mathematical verification and experimental validation

We first verify our algorithm by reproducing the temperature and species profiles of the stoichiometric, atmospheric pressure hydrogen–air flame found in Smooke *et al.* [6], under the conditions they specify. Visual inspection of the results (not show here, but see [4]) shows that the stationary flame structure is identical to that of Ref. [6]. For a more rigorous verification, the stationary structures of the laminar premixed flame at $p_o=1$ atm and $T_o=800$ K is obtained using a wide range of uniform grid sizes: $3\times10^{-2}\le\Delta x\le6.25\times10^{-5}$ cm. The finest discretization level is close to the smallest grid size used by the adaptive method, 6×10^{-5} cm. The somewhat high value of $T_o=800$ K is employed to avoid corruption of numerical roundoff errors in early portions of the flame structure.

The spatial distribution in the flame zone reveals the nature of the computational error shown in Fig. 1. Here, χ_{OH} is plotted

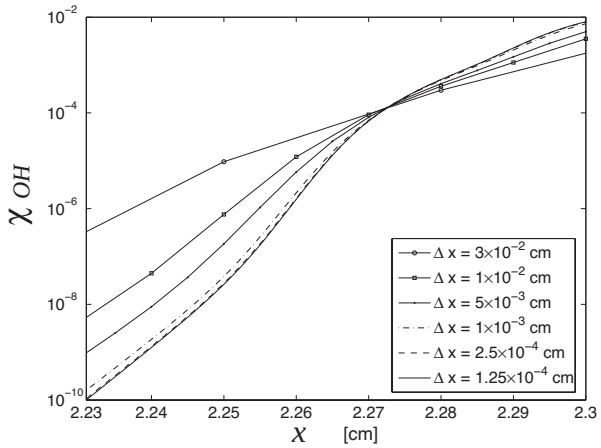

Figure 1. χ_{OH} vs. x for the hydrogen–air flame simulation with $T_o=800$ K and $p_o=1$ atm for various grid resolutions.

on a semi-log scale versus x for various Δx. Certainly, the solution is converging as $\Delta x\to0$. In the early stages of the flame, the relative errors in χ_{OH} can span many orders of magnitude unless Δx is brought down to the micron-scale. This is not obvious when viewed on a linear scale.

For all dependent variables z_i, $i=1,\ldots,N+3$, the maximum

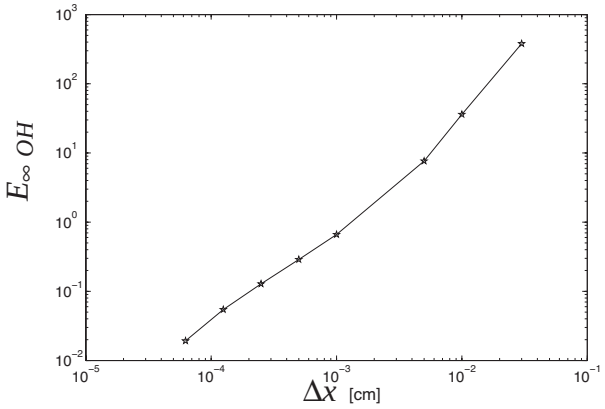

Figure 2. Relative error of χ_{OH} vs. the discretization size for the hydrogen–air flame simulation with $T_o = 800$ K and $p_o = 1$ atm.

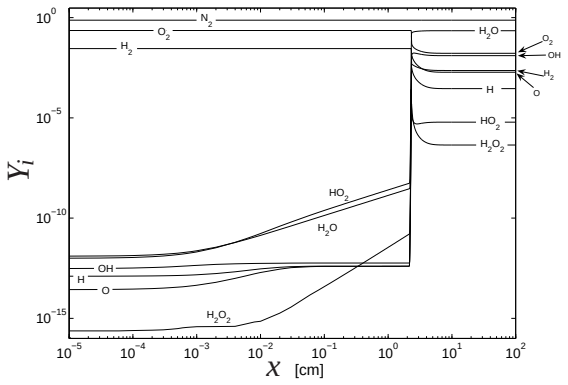

Figure 3. Species mass fraction vs. distance for a stoichiometric hydrogen–air flame, $T_o = 800$ K, $p_o = 1$ atm.

relative error, $E_{\infty i}$, is calculated by using the following formula:

$$E_{\infty i} = \max_{x_o \le x \le x^{eq}} \left| \frac{z_i^{exact}(x) - z_i(x)}{z_i^{exact}(x)} \right|. \qquad (10)$$

Fig. 2 shows the relative maximum error as a function of grid size. To obtain a desirable $E_\infty < 0.1$, a resolution of $\Delta x \le 2 \times 10^{-4}$ cm has to be utilized. Larger values can induce unacceptably large relative errors; e.g. for $\Delta x = 10^{-2}$ cm the relative error in χ_{OH} is 40.

Next, comparison with experiment addresses the question as to whether the model represents the observable physics well. For validation purposes, a series of calculations is performed on an atmospheric pressure hydrogen–air laminar premixed flame initially at $T_o = 298$ K. For different equivalence ratios Φ, the flame speed is determined. A comparison between the calculated flame speeds and experimental data [14] reveals that the computational predictions lie within the scatter of the experimental data, and they are as good as have been found by others [6, 14] (not shown here, but see [4]).

4.2 Stoichiometric hydrogen–air premixed laminar flame

Next, the PREMIX code [9] is used to determine the stationary structure of the stoichiometric premixed laminar flame at atmospheric pressure. The specified temperature is $T_f = 900$ K, the specified temperature location is assigned at $x_f = 2.30$ cm, and the mixture temperature at the cold boundary is $T_o = 800$ K.

Fully resolved steady species mass fraction profiles are shown in Fig. 3. Although linear scales are usually used in the literature, a log–log scale has been employed to better illustrate the disparate scales. The figure shows the spatial distribution of species mass fractions throughout the entire flame zone. We

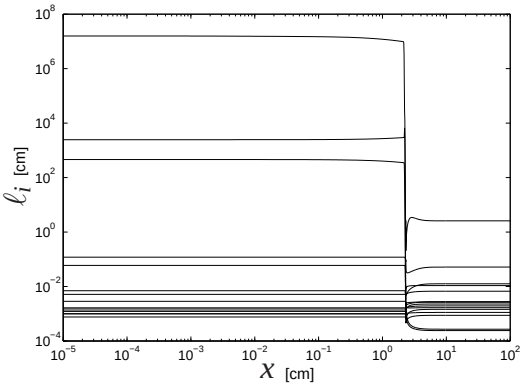

Figure 4. Predicted length scales over which stoichiometric hydrogen–air flame evolve vs. distance, $T_o = 800$ K, $p_o = 1$ atm.

adopt the simple estimate for the reaction length scale (i.e. flame thickness) given by Williams [15], pp. 130–136,

$$\ell_{reaction} = \frac{k}{\rho_o c_p S}, \qquad (11)$$

where for this case $\ell_{reaction} = 1.60 \times 10^{-2}$ cm. In the above expression, k is the mixture thermal conductivity and S is the flame speed.

Having the resolved structure, the local Jacobian and the eigenvalues, and thus the local length scales, ℓ_i, are obtained from the cold boundary to near equilibrium, as shown in Fig. 4. The multi-scale nature of the problem and the length scales over which the species evolve are clearly shown. The length scales 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 evolution of a particular species is not associated with a particular length scale, since the species mass fractions depend on local linear combinations of all eigenmodes. Thus, the species mass fractions vary on these scales through the entire domain. The important finest scale is $\ell_{finest} = 2.41 \times 10^{-4}$ cm, which occurs at equilibrium. The predicted finest length scale and the smallest scale over which the species vary, $x \approx 10^{-4}$ cm, are nearly identical. Moreover, the finest length scale effect in the preheat zone can be observed in the variation of the minor species mass fractions, which ensures the consistency between the eigenvalue-determined finest length scale and the smallest scale over which the species vary. As the system approaches equilibrium, all of the eigenvalues are real. Additionally, the analogous finest length scale in an inviscid Chapman–Jouguet detonation in a comparable hydrogen–air mixture has been found using the algorithm of Ref. [5] to be close in magnitude, 5.92×10^{-5} cm.

Following the same procedure, a comparison between the predicted finest length scale ℓ_{finest} and the flame thickness $\ell_{reaction}$ over a range of pressures is presented in Fig. 5. It reveals that ℓ_{finest} is well correlated with $\ell_{reaction}$ and that both decrease as pressure is increased. On the other hand, ℓ_{finest} is at least one order of magnitude smaller than $\ell_{reaction}$, which indicates the presence of scales smaller than the flame thickness.

5 A Priori Estimates from Collision Theory

Next, we explore the possibility that there may be a fundamental explanation for the finest length scales revealed by the eigenvalue analysis. We thus report an *ad hoc*, but plausible, analysis based on straightforward estimates from standard molecular collision theory. We find that a remarkably simple formula is able to accurately predict the length scales obtained by the preceding eigenvalue analysis. In a totally independent calculation, the mean free path ℓ_{mfp} for the mixture studied above is estimated based on the simple relation given by Vincenti and Kruger [16]:

$$\ell_{mfp} = \frac{\mathsf{m}}{\sqrt{2}\pi d^2 \rho}. \qquad (12)$$

Here, the parameters are d the molecular collision cross-section diameter, $\mathsf{m} = M/\mathcal{N}$ the mass of a molecule, and $\mathcal{N} = 6.0225 \times 10^{23}$ mole^{-1} Avogadro's number. For the calculation of ℓ_{mfp}, the estimate of $d = 3.70 \times 10^{-8}$ cm for air is adopted from Ref. [16]; $M = 28.00$ g mole^{-1} and $\rho = 1.11557 \times 10^{-4}$ g cm^{-3} were obtained for the mixture from `PREMIX` calculations. The estimate (12) reveals that $\ell_{mfp} = 5.87 \times 10^{-5}$ cm, which is roughly one order of magnitude smaller than the continuum-based ℓ_{finest}. The variation of ℓ_{mfp} with initial pressure is also shown in Fig. 5. Finally, it is noted that analagous results have been obtained for a wide variety of hydrocarbon laminar flames as well as detonations [4].

6 Comparison with previous results

Though this work focuses on one-dimensional steady laminar premixed flames, the estimates provide bounds for problems where multi-dimensional and unsteady effects are simulated. A comparison between the predicted finest length scale and the utilized discretization in some of the best calculations of laminar premixed flames in hydrogen–air mixtures has been done. In all cases, the predicted finest length scales are at the micron-level, and they are well correlated with the associated cutoff length scales admitted by the continuum theory. In literature examined, [17–19], the minimum grid size was 2.5×10^{-2} cm.

Thus, the main result of this exercise is that none of these studies have utilized a grid resolution Δx that is less than or equal to the finest length scale ℓ_{finest} which is required to have unambiguously resolved results for a steady one-dimensional laminar premixed flame in a comparable mixture under the same conditions. Moreover, the utilized grid resolution Δx is at least two orders of magnitude greater than ℓ_{finest}. In each study, different physical phenomena are simulated, and the mathematical models vary, but the commonality in all studies is the usage of a detailed kinetics model to simulates flame in a premixed mixture.

Lastly, our results are in agreement with independent estimates found in direct numerical simulations (DNS) of turbulent reacting flows. Recently, Chen *et al.* [20], presented a two-dimensional DNS of hydrogen–air combustion described by detailed kinetics. The domain size was 4.1×4.1 mm^2, and the calculations required a grid resolution of $\Delta x = 4.30 \times 10^{-4}$ cm to resolve the ignition fronts. We note that this grid size is of the

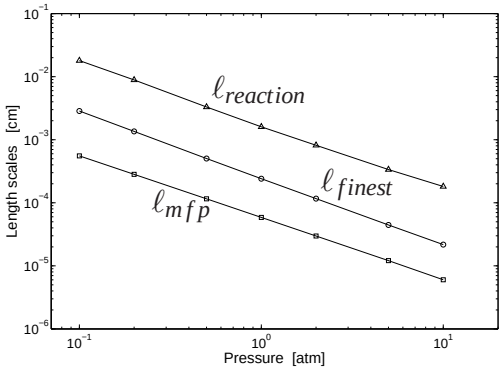

Figure 5. The flame thickness, the finest length scale predicted by eigenvalue analysis, and the mean free path estimate vs. pressure for stoichiometric hydrogen–air flame with $T_o = 800$ K.

same order as the finest length scale predicted here!

7 Conclusion

The present one-dimensional steady calculations reveal that, for an adiabatic laminar premixed flame freely propagating in stoichiometric mixtures described by detailed kinetics and multicomponent transport, the grid resolution to formally resolve the flow structures is at the micron-level. This length scale is predicted by utilizing a rigorous eigenvalue analysis. Moreover, a formal grid convergence study has been performed independently, and the same length scale requirement is obtained. The length scale predictions are fully reflective of the underlying physics and *not* the particular numerical method chosen. This has been verified by showing that the finest length scale is well-correlated with the mean free path cutoff length scale estimated from kinetic theory. Thus, it is possible to use a simple mean free path calculation as an *a priori* estimate of the lower bound for grid discretization. This finest length scale for deflagration is nearly identical to the finest length scale for detonation. Related calculations of unsteady and multi-dimensional laminar flames in the literature typically employ much larger discretizations than suggested by the present analysis. The full consequences of this under-resolution await rigorous linear and non-linear stability analysis as well as DNS in order to be determined.

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