

On the Coupling Between Length and Time Scales in Reactive Flows

J. M. Powers, S. Paolucci, A. N. Al-Khateeb

University of Notre Dame, Department of Aerospace and Mechanical Engineering, Notre Dame, Indiana, USA

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 *a posteriori* and estimated *a priori*. Application of the method to laminar premixed flames in several mixtures over a wide range of pressures and fuel–air ratios reveals that the finest length scale is on the order of 10^{-4} cm.

I. 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 temporal scales, and the ratio between the largest and smallest time scale is a measure of the system's stiffness. As the temporal scales' range widens, more stringent demands arise to assure the numerical stability of the solution algorithm. Equivalently, as the spatial scales' range widens, the required grid resolution to capture all the length scales increases. Of course, temporally implicit methods with large time steps can often be used for problems in which the dynamics are not critical, and overly large spatial discretizations often have the benign effect of smearing fine scale results over a few computational cells. However, for the challenging class of combustion problems which are inherently unsteady, the dynamics are often crucial. In such problems, proper numerical resolution of all time and space scales can be critical to drawing the correct conclusions. And 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, 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 more demanding characterization is fully consistent with standard notions found in the broader mathematical and scientific computing literature.

The main aim of this work is to rigorously determine 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. Here, the robust spatial eigenvalue-based method to calculate the length scales employed in Powers and Paolucci [2] for gas phase detonation and Al-Khateeb, *et al.* [3] for laminar flames is implemented. Verification and validation of the method is discussed. A detailed examination of the length scales of a hydrogen–air laminar flame is given. These predictions compare favorably to those of a simple *a priori* estimate from collision theory. The collision theory also estimates well the finest length scales for a wide variety of hydrocarbon laminar flames and detonations.

II. ANALYSIS

We consider equations describing a one-dimensional adiabatic laminar premixed mixture of N molecular species composed of L atomic elements which undergo J reversible reactions with no body force present. The complete system is first reduced into a system of ordinary differential equations by relaxing the time-dependent behavior to a steadily propagating flame front with constant, albeit unknown, flame speed. A low Mach number assumption is adopted. Selected conservation equations are integrated analytically, giving rise to a coupled system of differential-algebraic equations. A standard computational scheme is used to obtain the steady flame structure. Linearization of the equations about this solution gives rise to a generalized eigenvalue problem which in turn

provides full information on all scales contained in the problem.

III. RESULTS

A stoichiometric hydrogen–air mixture at $p_o = 1$ atm has been considered, where the initial molar ratio is given by $2H_2 + O_2 + 3.76N_2$. In this mechanism, the reactant species are $H_2, O_2, H, O, OH, HO_2, H_2O_2$, and H_2O , and the inert diluent for the mixture is N_2 .

A. Verification and Validation

Two types of verification are performed: 1) achieving the same results that have been obtained in a previous study, and 2) a formal grid convergence study.

For the first verification, a calculation is performed to reproduce the temperature and species profiles of a stoichiometric, atmospheric pressure hydrogen–air flame found in Smooke *et al.* [4]. Visual inspection shows that the stationary flame structure is identical to that of Smooke.

For the second more rigorous verification, the stationary structures of one-dimensional, stoichiometric, adiabatic, $2H_2 + O_2 + 3.76N_2$ laminar premixed flames at $p_o = 1$ atm and $T_o = 800$ K are obtained over a wide range of uniform grid sizes: $3 \times 10^{-2} \leq \Delta x \leq 6.25 \times 10^{-5}$ cm. The finest discretization level is close to the smallest grid size used by an adaptive method, 6×10^{-5} cm. Given that we have a steadily propagating flame, these fine length scales have clear implications on the required time scales of reaction for a given fluid particle. In this formal grid convergence study, for all dependent variables z_i , the relative errors, $E_{\infty i}$ throughout the entire domain are calculated by using the following formula:

$$E_{\infty i} = \max_{x_o \leq x \leq x_{eq}} \left| \frac{z_i^{exact}(x) - z_i(x)}{z_i^{exact}(x)} \right|, \quad (1)$$

The result for one dependent variable, OH mole fraction, is presented in figure 1. Results for all other variables bear remarkable similarity, and so are not shown here. Solutions are obtained on eight different uniform grids. The “exact” solution is estimated using Richardson’s extrapolation from the three finest grids [1]. In computing the error via equation (1), points with species mole fraction below 10^{-10} were excluded because of potential roundoff corruption in double precision calculations. Figure 1 shows that to obtain a desirable $E_{\infty} < 0.1$ in this problem, a spatial resolution of $\Delta x \leq 2 \times 10^{-4}$ cm has to be utilized. Larger discretization sizes can induce unacceptably large relative errors; e.g. for $\Delta x = 10^{-2}$ cm the relative error in OH mole fraction is 40.

A comparison with experimental results addresses the question as to whether the model represents well the observable physics. For validation purposes, a series of calculations was performed on an atmospheric pressure hydrogen–air laminar premixed flame initially at $T_o = 298$ K. For different equivalence ratios, the flame speed is determined. A comparison between the calculated flame speeds and data reveals that the computational predictions lie within the scatter of the experimental data.

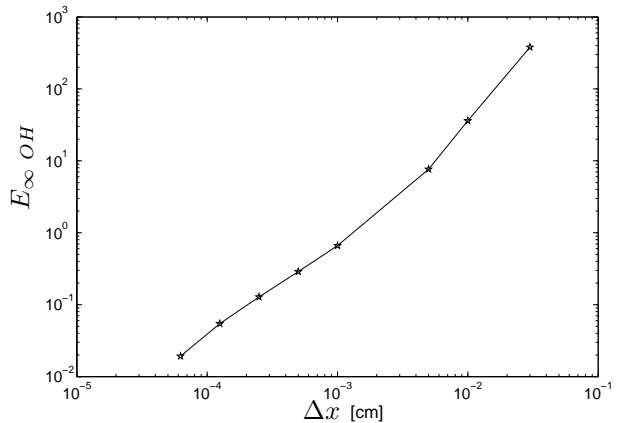


Fig. 1. Relative error of OH mole fraction vs. the discretization size for the hydrogen–air flame simulation with $T_o = 800$ K and $p_o = 1$ atm.

B. Premixed Laminar Flame

The stationary structure of a one-dimensional, stoichiometric, adiabatic, $2H_2 + O_2 + 3.76N_2$ premixed laminar flame at atmospheric pressure is determined. Having the fully resolved structure in hand, the local Jacobian and the spatial eigenvalues are calculated from the cold boundary to near equilibrium. As a result, the local length scales ℓ_i 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. Most unsteady, multi-dimensional calculations in the literature employ discretization sizes which are two to three orders of magnitude too coarse to capture these scales. However, recently Chen, *et al.* [5] have carefully analyzed the necessary grid resolution to obtain grid-independent species concentration profiles, and find results entirely consistent with those given here.

IV. A PRIORI ESTIMATES FROM COLLISION THEORY

In a totally independent calculation, the mean free path ℓ_{mfp} for the mixture studied in the previous section is estimated. The estimate reveals that $\ell_{mfp} = 5.87 \times 10^{-5}$ cm, which is roughly one order of magnitude smaller than the continuum-based ℓ_{finest} .

A comparison between the predicted finest length scale ℓ_{finest} , the flame thickness $\ell_{reaction}$, and the mean free path estimate ℓ_{mfp} , over a wide range of pressures is presented in figure 2. It reveals that the finest length scale is well correlated with the mean free path and that both of them decrease as pressure is increased. On the other hand, ℓ_{finest} is orders of magnitude smaller than $\ell_{reaction}$, which indicates the presence of scales smaller than the flame thickness. This approach is extended to non-stoichiometric hydrogen–air mixtures and several other stoichiometric mixtures: 1) methane–air, 2) ethane–air, 3) propane–air, 4) ethylene–air, and 5) acetylene–air. For each mixture, two cases have been studied: 1) the freely propagating laminar flame, and 2) the Chapman–Jouguet detonation. In each case, the simple collision

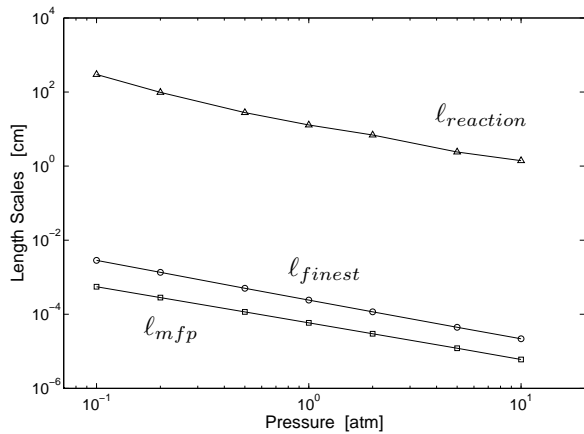


Fig. 2. The reaction zone length, the finest length scale predicted by eigenvalue analysis, and the mean free path vs. pressure for stoichiometric hydrogen-air flame, $T_o = 800$ K.

theory estimate accurately predicts the finest length scale revealed by the eigenvalue analysis.

In addition, we explore further the possibility of the existence of a direct connection between the finest length scale predicted by the eigenvalue analysis and that predicted by the simple collision theory estimate. Such an analysis is analytically tractable for the simple kinetic system of isothermal oxygen dissociation with no diffusion. Analysis given in detail in Ref. [3] reveals a precise analytic correlation between the finest length scale predicted by the eigenvalue analysis of the continuum system and the mean free path length of collision theory for this simple example: $l_{finest} \approx 5l_{mfp}$. *The analysis does not depend on any discrete numerical method.* Based on the earlier numerical results, we believe it is reasonable to speculate that this result extends to systems with more complex kinetics.

V. CONCLUSION

The present one-dimensional steady calculations reveal that the required 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 is 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 is demonstrated by showing that the finest length scale is well-correlated with the mean free path cutoff length scale estimated from kinetic theory. 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 analyses as well as further DNS results.

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