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PHYSICAL AND CHEMICAL PROCESSES IN FLAMES

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SUMMARY/OVERVIEW

The objectives of the present program are to develop detailed and simplified chemical kinetics model for hydrocarbon combustion, and to understand and quantify the dynamics of flames. During the reporting period progress were made in the following projects: (1) Laminar flame speeds of mixtures of ethylene and *n*-butane were studied. Results suggest the dominant influence of the adiabatic flame temperature in assessing the mixture flame speed. (2) A theory of complex CSP (computational singular perturbation) for chemistry reduction and analysis was developed. An algorithm was derived through which detailed mechanisms can be systematically reduced to simpler ones without compromising the comprehensiveness of the original mechanism. (3) Stretch effects on premixed flame pulsation were computationally and analytically studied. Results show that positive (negative) stretch promotes (retards) the development of pulsating instability, which is completely opposite to the influence of stretch on cellular instability. (4) Pulsating instability of near-limit diffusion flames were computationally studied. Results show that the instability and the subsequent extinction of methane and hydrogen flames assume the characteristics of diffusion and premixed flames respectively.

TECHNICAL DISCUSSION

1. Laminar Flame Speeds of Ethylene/*n*-Butane/Air Mixtures

The laminar flame speed is a useful parameter towards development and validation of detailed kinetic mechanisms of hydrocarbon fuels. The objectives of the present study are: (a) to extend our previous study involving single fuels to fuel blends, and (b) to experimentally determine the flame speeds by using Digital Particle Image Velocimetry (DPIV).

Regarding the first objective, we note that since nearly all practical fuels are multi-component, it is important to develop theoretical and semi-empirical capabilities to predict the flame speeds of fuel mixtures. Furthermore, existing reaction models of hydrocarbon combustion were mostly developed based on results from single fuels and as such have not been adequately verified for fuel mixtures. Finally, it is also of interest to develop semi-empirical mixing rules.

Mixtures of ethylene and *n*-butane were selected for study because ethylene is an important intermediate in hydrocarbon oxidation while *n*-butane is a representative *n*-alkane. Figure 1.1 shows experimental data of the laminar flame speeds of ethylene/air and *n*-butane/air mixtures. These experimental data compare well with the calculated values obtained by using the kinetics model developed under the present program.

Using this kinetics model, further calculations were performed for mixtures of ethylene and *n*-butane in the ratios of 2:1, 1:1, and 1:2, as shown in Fig. 1.2. As is reasonable to expect, the flame speeds of the fuel mixtures are bounded by those of ethylene/air and *n*-butane/air. However, results for the mixtures seem to be weighted towards the n-butane values. The same

biasing is also exhibited for the calculated adiabatic flame temperatures (Fig. 1.2), indicating that the flame temperature instead of kinetic coupling could have the dominant influence on the flame speed.

The biasing could simply be a consequence of how the composition of mixture is defined. The present mixing ratio is the conventional one, based on the molar ratios. There are, however, other definitions that could be more relevant physically. This aspect is being studied.

Preliminary experimental data were also taken for the laminar flame speeds of the fuel blends, showing the same trend of biasing. In order to prepare ourselves for massive amount of data taking, we have been developing a DPIV system which, compared to LDV, is capable of significantly reducing the test time and positioning error.

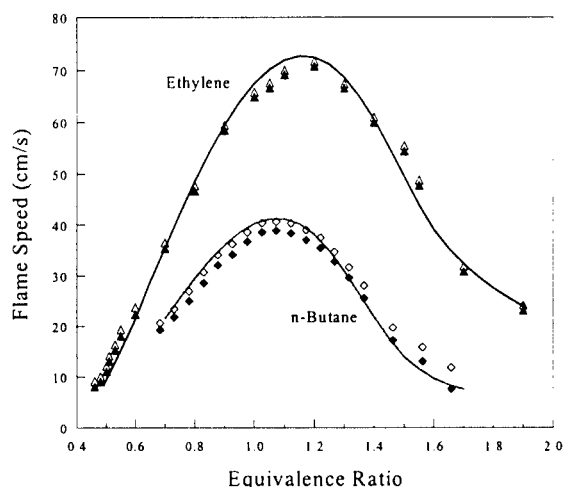Results of the above activities are reported in Publication No. 1.

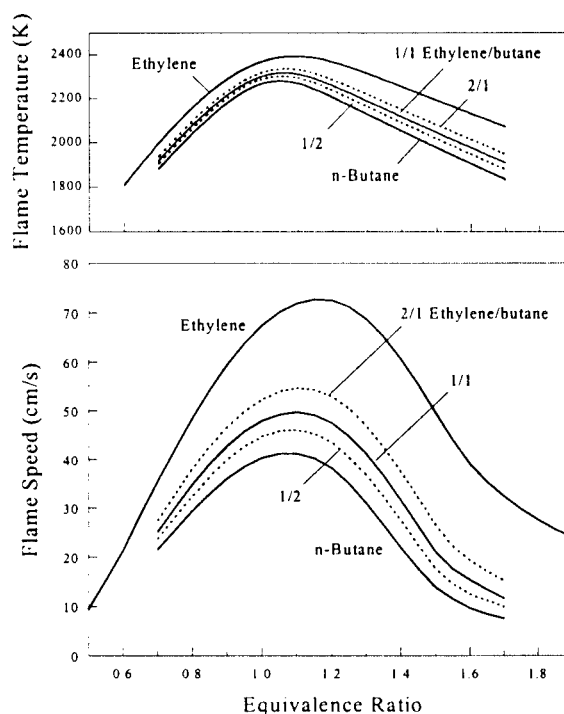Figure 1.1.   Experimental and computed laminar flame speeds of ethylene/air and n-butane/air mixtures.

Figure 1.2   Computed adiabatic flame temperatures and laminar flame speeds of mixtures of ethylene and n-butane fuels.

2. Theory of Complex Computational Singular Perturbation

The method of Computational Singular Perturbation (CSP) for the analysis and reduction of detailed chemical mechanisms has been extended [3] to the complex eigensystem. Results show that the time scales of chemical species change dramatically and non-monotonically, and that oscillatory modes appear frequently in large chemical reaction mechanisms. The present method is then employed to generate reduced mechanisms for hydrogen/air and methane/air oxidation. Using the hydrogen/air system as an example, Fig. 2.1 shows that the number of complex modes increases with decreasing residence time and hence increasing chemical sensitivity. Figure 2.2 shows the relation between the size of the reduced mechanism with a measure of the residence time – the longer the residence time (smaller α) the more the number of steady-state species and hence the smaller the mechanism. The validity of these reduced mechanisms is evaluated based on the responses of the perfectly stirred reactors and the one-dimensional planar propagating premixed flames. Comparison between the reduced and detailed chemistries over a wide range of pressures and equivalence ratios show good agreement on the flame speed, temperature and structure.
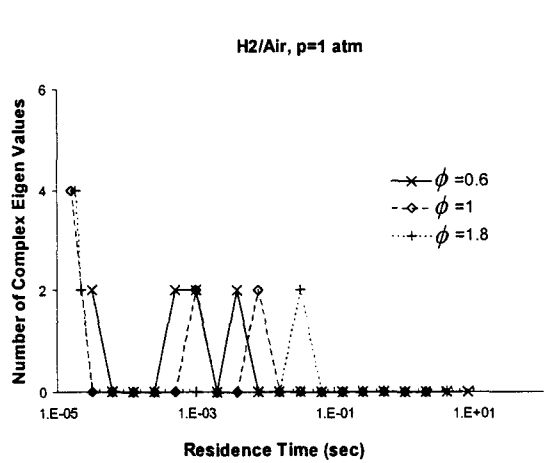
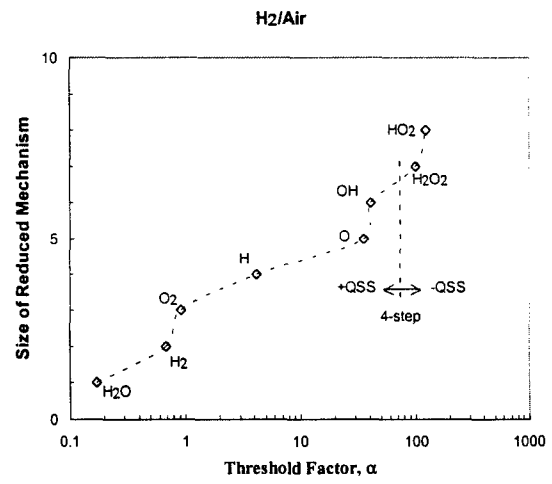Figure 2.1 Number of complex modes for hydrogen/air mixtures for different residence times in the PSR.

Figure 2.2 Diagram indicating the variation of the size of the reduced mechanism with different residence times; smaller α implies longer residence time, more steady-state species, and smaller reduced mechanism.

3. Effects of Stretch on Flame Pulsation

Effects of stretch on the pulsating instability of premixed flames have been investigated [10] via the negatively-stretched inwardly propagating spherical flame (IPF) and the positively-stretched counterflow flame (CFF). Computational and asymptotic analysis results show that pulsating instability is suppressed by the former and promoted by the latter.

In particular, it is shown that for a given rich hydrogen/air mixture whose one-dimensional, freely propagating flame is pulsatingly unstable, the IPF initially propagates at the laminar flame speed when the flame radius is large. Oscillation subsequently develops, and is then amplified, damped, and eventually suppressed as the flame propagates inward and the magnitude of stretch increases.
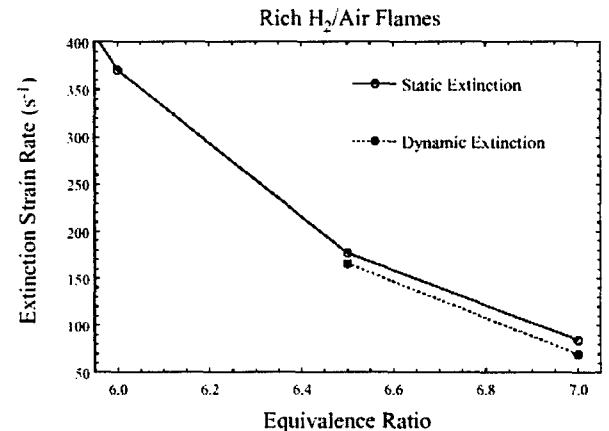

Figure 3.1 Extinction strain rates for the steady and oscillating counterflow flames

For the positively stretched rich hydrogen/air CFF, oscillation is initiated at an equivalence ratio much smaller than the one-dimensional rich threshold. Furthermore, the critical strain rate leading to pulsation is smaller than the corresponding static extinction limit, implying that the flame extinguishes in the pulsating instead of the steadily propagating mode such that the flammable range is accordingly narrowed (Fig. 3.1). In addition, it is seen that the pulsating flames are quasi-steady in nature in that the period of oscillation is larger than the characteristic

flame time. As such, the unsteady flame cannot recover once the instantaneous flame temperature is reduced below the corresponding steady-state extinction temperature.

The computationally determined pulsation limits using realistic chemistry and transport were also found to agree well with the asymptotic results based on simplified chemistry and transport, provided that the global activation energies and Lewis numbers are appropriately extracted from laminar flame responses.

4. Oscillatory Extinction of Diffusion Flames

The transient behavior of spherical diffusion flames was computationally studied [11] in both the high-velocity and radiation-induced limit of the isola response of flame extinction. Oscillatory instability was observed near both steady-state extinction limits, with the oscillation amplitude growing until it becomes large enough to extinguish the flame. For the hydrogen and methane flames studied, oscillation always leads to extinction, although the characteristics of oscillation are qualitatively different. Specifically, the methane flame exhibits large amplitude temperature fluctuations at a frequency of about 0.35 Hz, while the hydrogen flame has small amplitude fluctuation with a frequency of about 60 Hz. The oscillations in the methane flame appear to be characteristic of those studied based on the diffusion flame structure, while those of hydrogen flames are reminiscent of those observed in rich hydrogen/air premixed flames.

MAJOR/RELEVANT PUBLICATIONS (May 2000 – April 2001)

1. "Determination of laminar flame speeds of ethylene/n-butane/air flames using digital particle image velocimetry," Paper No. 138, Second Joint Meeting of the US Section of the Combustion Institute, Oakland, CA, March 25-28, 2001.
2. "Detailed kinetic modeling of 1,3-butadiene oxidation at high temperatures," by A. Laskin, H. Wang, and C. K. Law, *International Journal of Chemical Kinetics*, Vol. 32, pp. 589-614 (2000).
3. "Theory of complex CSP for chemistry reduction and analysis," by T. Lu, Y. Ju, and C. K. Law, *Combustion and Flame*, in press.
4. "Structure, aerodynamics, and geometry of premixed flamelets," by C.K. Law and C.J. Sung, *Progress in Energy and Combustion Science*, Vol. 26, pp. 459-505 (2000).
5. "Structural sensitivity, response, and extinction of diffusion and premixed flames in oscillating counterflow," by C. J. Sung and C. K. Law, *Combustion and Flame*, Vol.123, pp. 375-388 (2000).
6. "On curvature-affected pulsating instability in inwardly-propagating spherical flames," by C. J. Sung, C. J. Sun, and C. K. Law, *Combustion Science and Technology Communications*, Vol.1, pp..7-10 (2000).
7. "Radiation induced instability of stretched premixed flames," by Y. Ju, C. K. Law, K. Maruta, and T. Niioka, *Proceedings of the Combustion Institute*, Vol. 28, pp. 1891-1900 (2000).
8. "The role of pulsating instability and global Lewis number on the flammability limit of lean heptane/air flames," by E. W. Christiansen, C. K. Law, and C. J. Sung, *Proceedings of the Combustion Institute*, Vol. 28, pp. 807-814 (2000).
9. "Steady and pulsating propagation and extinction of rich hydrogen/air flames at elevated pressures," by E. W. Christiansen, C. J. Sung, and C. K. Law, *Combustion and Flame*, Vol. 124, pp. 35-49 (2001).
10. "On stretch-affected pulsating instability in rich hydrogen/air flames: asymptotic analysis and computation," by C.J. Sung, A. Makino, and C.K. Law, Paper No. 262, Second Joint Meeting of the US Section of the Combustion Institute, Oakland, CA, March 25-28, 2001.
11. "A computational study on oscillatory extinction of spherical diffusion flames," by E.W. Christiansen, S.D. Tse, and C.K. Law, AIAA-2001-1084, 39th Aerospace Sciences Meeting, January 8-11, 2001, Reno, NV.