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Computational Flame Diagnostics for Direct Numerical Simulations with Detailed Chemistry of Transportation Fuels

PI: Tianfeng Lu

Department of Mechanical Engineering
University of Connecticut, Storrs, CT 06269-3139
tlu@engr.uconn.edu

I. Program Scope

The goal of the proposed research is to create computational flame diagnostics (CFLD) that are rigorous numerical algorithms for systematic detection of critical flame features, such as ignition, extinction, and premixed and non-premixed flamelets, and to understand the underlying physicochemical processes controlling limit flame phenomena, flame stabilization, turbulence-chemistry interactions and pollutant emissions etc. The goal has been accomplished through an integrated effort on mechanism reduction, direct numerical simulations (DNS) of flames at engine conditions and a variety of turbulent flames with transport fuels, computational diagnostics, turbulence modeling, and DNS data mining and data reduction. The computational diagnostics are primarily based on the chemical explosive mode analysis (CEMA) and a recently developed bifurcation analysis using datasets from first-principle simulations of 0-D reactors, 1-D laminar flames, and 2-D and 3-D DNS (collaboration with J.H. Chen and S. Som at Argonne, and C.S. Yoo at UNIST). Non-stiff reduced mechanisms for transportation fuels amenable for 3-D DNS are developed through graph-based methods and timescale analysis. The flame structures, stabilization mechanisms, local ignition and extinction etc., and the rate controlling chemical processes are unambiguously identified through CFLD. CEMA is further employed to segment complex turbulent flames based on the critical flame features, such as premixed reaction fronts, and to enable zone-adaptive turbulent combustion modeling.

II. Recent Progress

A. Reduced mechanisms and reduction methods

a. Development of reduced mechanisms

Chemistry of practical engine fuels involves a large number of species and reactions, as well as severe chemical stiffness. A necessary step to accommodate realistic chemistry in large-scale flame simulations is to obtain reduced mechanisms with small sizes and satisfactory accuracy. In this project, reduced mechanisms were developed for various fossil and renewable engine fuels [1,6,7,9,10]. The reduction was primarily based on the directed relation graph (DRG) methods, analytically solved linearized quasi steady state approximations, while chemical stiffness was dynamically removed during flame simulations [ii].

The reduction methods feature high computational efficiency and rigorous error control. Specifically, a 30-transported species reduced mechanism [10] was developed for dimethyl ether (DME) as a diesel surrogate and applied in DNS of a turbulent DME jet flame. A 28-species reduced mechanism [7] was developed for ethanol/air including NOx formation, and was applied for 2-D and 3-D DNS of homogeneous charge compression ignition (HCCI) and spark assisted compression ignition (SACI) (collaboration with Dr. J.H. Chen at Sandia National Laboratories). A 116-species reduced mechanism [1] was developed for primary reference fuels (PRF) as a gasoline surrogate, and a 73-species reduced mechanism [9] was developed for a biodiesel surrogate (ternary mixtures of *n*-heptane, methyl decanoate and methyl-9-decanoate) from the detailed mechanisms developed by the Lawrence Livermore National Laboratory. Both reduced mechanisms were used for 2-D DNS of HCCI combustion involving different flame propagation modes (collaboration with Prof. C.S. Yoo at UNIST) [1,9]. The comprehensiveness and accuracy of developed 28-species reduced mechanism for ethanol/air and a non-stiff 171-species skeletal and 116 species reduced mechanism for PRF/air are demonstrated in Fig. 1 for ignition and extinction for selected parameter ranges.

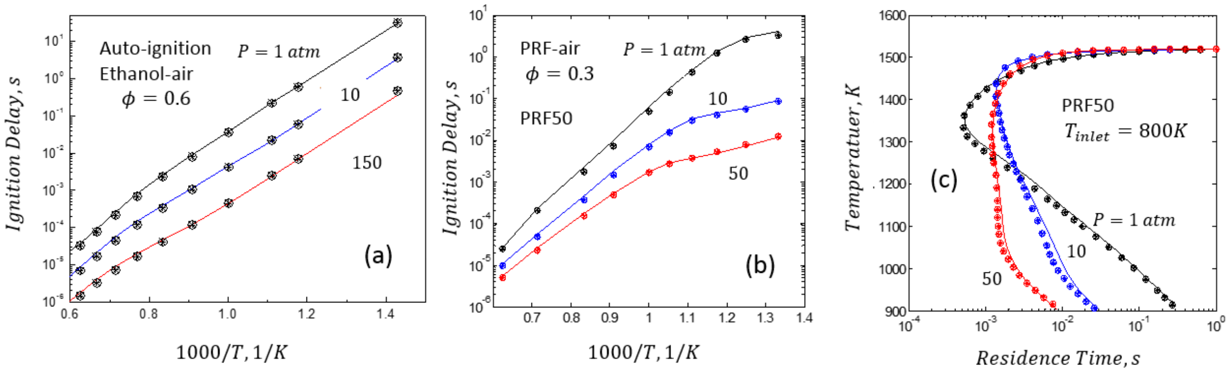

Figure 1. Comparison of the (a) 28-species reduced mechanism for ethanol/air [7], and a 171-species skeletal and 116-species reduced mechanisms with the detailed mechanism for (b) constant-pressure ignition delays, (c) extinction temperatures in perfectly stirred reactors (PSR) [1]. Lines: detailed, Circles: skeletal, Asterisks: reduced.

For simulations of lifted jet flames at diesel engine conditions, a detailed mechanism for *n*-dodecane was updated, reduced and integrated into diesel engine simulations (collaboration with Dr. W.J. Pitz at Lawrence Livermore National Laboratory and Dr. S. Som at Argonne National Laboratory) [6]. The updated detailed mechanism consists of more than 2000-species involving the NTC chemistry. A 106-species skeletal mechanism [6] was derived and employed to simulate a lifted diesel jet flame. The skeletal mechanism was comprehensively validated in 0-D, 1-D and 3-D combustion systems [6]. Good agreement was observed in the flame lift-off length (LOL) as shown in Fig. 2, which compares the simulated temperature isocontour and experimentally measured OH chemiluminescence data [6]. The *n*-dodecane mechanism was selected by the engine combustion network (ECN) for their diesel spray simulations [iii].

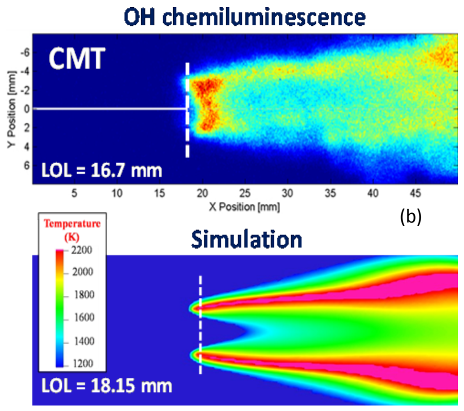

Figure 2. OH chemiluminescence image and simulated temperature contour plot for a lifted jet flame of *n*-dodecane into heated air (RANS simulation by S. Som) [6]

b. Linearized error propagation (LEP) model

Rigorous error control is critical for model reduction methods while the challenges result from the highly nonlinear couplings among different reaction pathways. While the DRG method can effectively control the worst-case error in skeletal mechanisms, error propagation in further reduced models can be highly complex. A linearized error propagation (LEP) model based on Jacobian analysis is developed to quantify the reduction error propagating into high concentration species due to the elimination of the species in low concentrations. The reduction is based on 0-D steady-state perfectly stirred reactors (PSR) involving both ignition and extinction states. Validation result in Fig. 3 shows that the LEP based reduction method can effectively control the worst-case errors in major species of interest. Furthermore, it is shown that the skeletal mechanism derived from PSR can be extended to more complex flames in that the PSR *S*-curve covers both ignition and extinction chemistry that are both relevant to turbulent combustion.

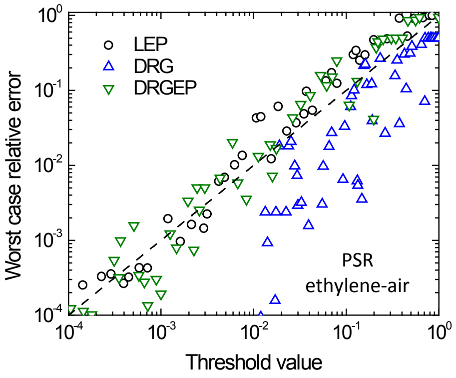

Figure 3. Worst case relative error in target species in PSR for 1-30atm, ϕ=1, Tin = 1000K, calculated using local skeletal models for ethylene/air based on USC-Mech II, derived with LEP, DRG, and DRGEP, respectively.

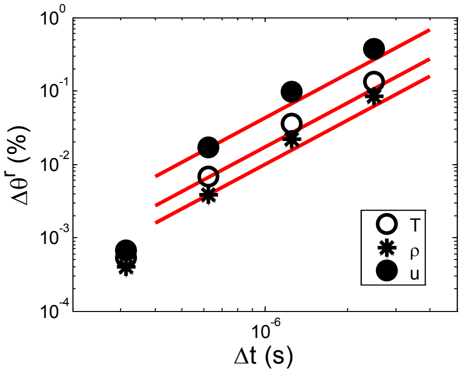

Figure 4. Demonstration of the second-order temporal convergence of the Strang splitting schemes with DAC for temperature, density and velocity in a 1-D premixed flame of methane/air [5]. The solid lines are lines of slope two.

c. Dynamic adaptive chemistry (DAC) for flame simulations

While the size of chemical kinetics of practical fuels can be large, not all the species and reactions typically are simultaneously important in different flame zones and time instances. To exploit this nature of detailed chemistry, a DAC method is developed to expedite time integration of chemically reacting systems based on the DRG method [3,5,11], which rigorously controls the worst-case incurred errors. The DAC method is studied with the Strang splitting schemes, which is widely adopted in computational fluid dynamics (CFD) simulations involving stiff chemistry, and a second order accuracy in time-integration is achieved as demonstrated in Fig. 4 for a 1-D freely propagating premixed laminar flame of methane/air [5]. Furthermore, the DAC and *in situ* adaptive tabulation (ISAT) [iv] are combined and compared. It is found that the performance of DAC is mostly independent of the nature of combustion simulations, e.g., steady or unsteady, premixed or non-premixed, and its efficiency increases with the size of the chemistry. A speedup factor of about 30 is achieved using DAC for a simulation of HCCI combustion of iso-octane/air in a partially stirred reactor (PaSR) [3]. Even larger speedup factors are achieved by combining DAC with ISAT. DRG-based DAC is further extended to the method of species time scale and rate analysis (TSRA) by modeling error propagation in major species. The TSRA method is applied to auto-ignition of methane/air and *n*-heptane/air mixtures over a wide range of initial temperatures and pressures. Ignition is accurately predicted, including the NTC behavior for *n*-heptane.

d. Reduction of multicomponent diffusion model

Multicomponent diffusion is involved in most practical combustion devices. For high-fidelity flame simulations, multicomponent diffusion models involve inversion of linear problems and can be time consuming when the number of species is large, say above 200 species. A systematic strategy is developed to reduce the multicomponent diffusion models by accurately accounting for the species whose diffusivities are important for flame responses, while approximating the diffusivities of the less important species [11]. The reduced models are investigated in planar premixed flames, counterflow diffusion flames, and ignition of droplet flames, showing significantly higher accuracy than that of the mixture-averaged model, while the computational cost was reduced by a factor of approximately 5 compared with the detailed multi-component model for an *n*-heptane with 88 species.

B. Computational flame diagnostics with chemical explosive mode analysis (CEMA) and bifurcation analysis (BA)

a. Development of Bifurcation analysis

Ignition and extinction are critical limit flame phenomena and of primary concern in many combustion applications. It is important to systematically identify the underlying chemical processes controlling ignition and extinction to understand and predict relevant flame behaviors at turbulent conditions.

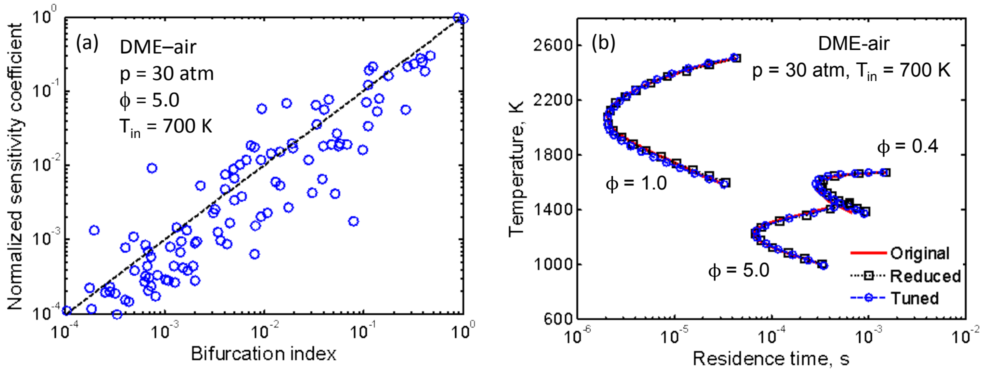

Figure 5. (a) Correlation of the bifurcation index (BI) with the normalized sensitivity coefficients, and (b) strong flame extinction calculated with reactions with large BI values with and without rate parameter tuning, for DME/air in PSR.

A method of bifurcation analysis (BA) [8] was developed based on the description of extinction and ignition states as bifurcation points on the *S*-curves of steady state combustion. Reactions important to ignition and extinction are identified based on their contributions to the bifurcation points, defined as the bifurcation index (BI) [8]. The BA method is validated against and compared with the brute-force global sensitivity analysis, which is probably the most widely used method to numerical identify important species and reactions for ignition and extinction. Figure 5 shows that the BI values are strongly correlated with the global sensitivity as demonstrated by the extinction state of DME/air in PSR. Compared with the global sensitivity analysis, the BI values carry clear physical meanings and are significantly more efficient to evaluate in complex flows.

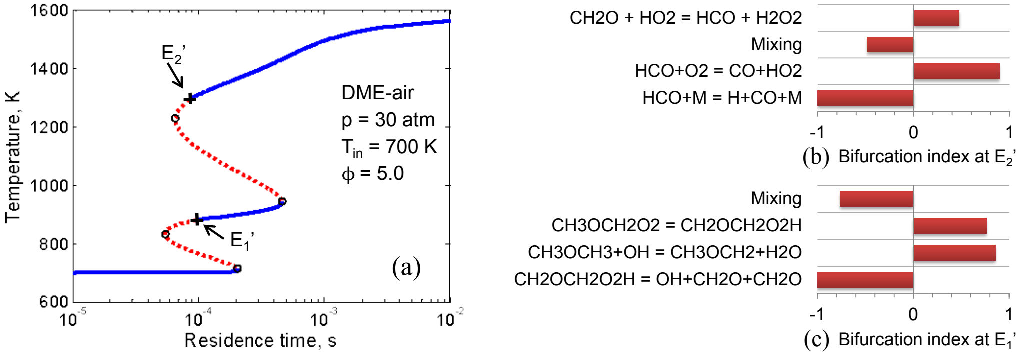

Figure 6. (a) The *S*-curve for a rich DME/air mixture in PSR (solid lines: stable, dashed: unstable). (b) and (c) the important reactions identified with bifurcation indices for the extinction of strong and cool flames, respectively.

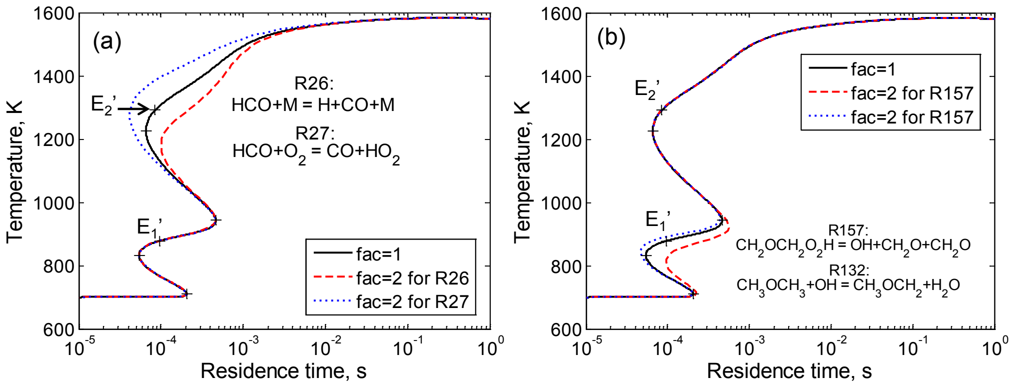

Figure 7. *S*-curves for DME/air with different perturbations in the “A”-factors of the important reactions for (a) the extinction of strongly burning flames, E_2’ and (b) the extinction of cool flames, E_1’. “fac” represents the factor multiplied to the “A”-factors.

Figure 6a shows the *S*-curve for a rich DME/air mixture with NTC behaviors in a steady state PSR. The actual flame ignition and extinction states, which may be different from the conventionally regarded turning points, were rigorously detected by bifurcation analysis as the transition states between the stable (solid lines) and unstable (dashed lines) states. Controlling reactions for the ignition and extinction states were identified by the bifurcation indices as shown in Fig. 6b for the extinction state of the strong flames (E2’ in Fig. 6a) and that of the cool flames

(E1'), respectively. It was found that the strong flame extinction was primarily controlled by the reactions relevant to CO formation, while the extinction of the cool flames involves the NTC chemistry, as expected and shown in Fig. 6(c). The importance of the reactions in Fig. 6b identified with the bifurcation analysis was verified by perturbing the "A"-factors and recalculating the extinction states with the perturbed mechanisms. It was found that uncertainties (or perturbations) in the "A"-factors of such identified reactions have significant effects on the corresponding extinction states as shown in Figs. 7, and thus their rate parameters should be determined with high accuracy in detailed kinetics. In reduced chemistry, the "A"-factors of these reactions can be slightly tuned to obtain highly reduced mechanism that can accurately predict ignition, extinction and onset of flame instabilities.

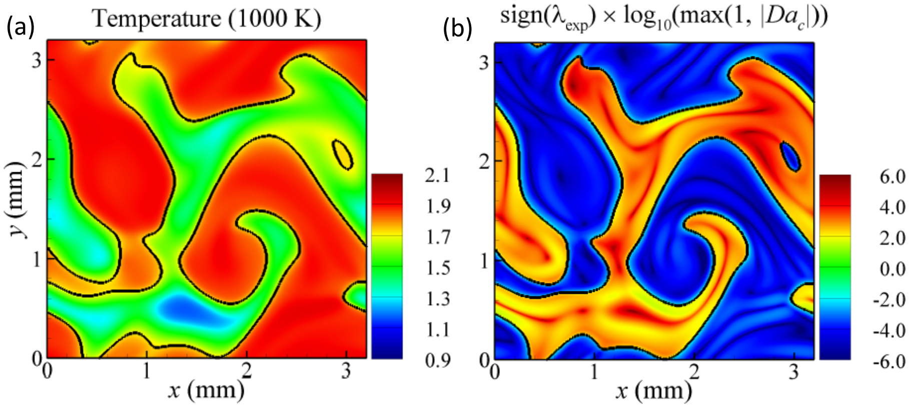

Figure 8. Isocontour of (a) temperature, and (b) a Damköhler number defined on CEMA, for a 2-D DNS of HCCI with PRF/air at $\tau/\tau ig = 0.95$ with 15K RMS initial temperature fluctuations (Simulation by C.S. Yoo) [1].

b. CEMA based diagnostics to detect critical flame features

The DNS data obtained for the ethanol, DME, PRF and biodiesel flames are systematically diagnosed with CEMA to extract critical flame features, such as premixed flame fronts, local ignition and extinction. In Fig. 8, the DNS results of a 2-D DNS of HCCI with PRF (collaboration with C.S. Yoo at UNIST) diagnosed with CEMA are plotted. The auto-igniting mixtures (red), near-equilibrium products (blue), and propagating premixed fronts (black isocontour line) are clearly identified with CEMA based a Damköhler number as shown in Fig. 8b. The relative importance of auto-ignition and premixed flame propagation in controlling the HCCI combustion rate was quantified. Different premixed flame propagation modes, i.e. the propagation of auto-igniting fronts and the canonical deflagration wave, were further identified with CEMA. A flame structure identified with CEMA is demonstrated in Fig. 9 for HCCI of biodiesel [9].

In Fig. 10, results on 1-D laminar flames show that mixtures in the reaction zone start to show explosive behaviors on the stoichiometric surface (P2), characterized by a positive eigenvalue of the chemical Jacobian, when the flame approaches the turning point from the upper branch of *S*-curve, while only non-explosive mixtures are present in strongly burning flames at low strain rates (P1). Thus, the presence of explosive mixtures is a necessary condition to detect flame extinction. This criterion is employed to study the DNS data in [v]. Local flame features, including strongly burning non-premixed, near-extinction, post-extinction zones and premixed reaction fronts are identified as shown in Fig. 11 and compared with 1-D premixed and non-premixed flamelets.

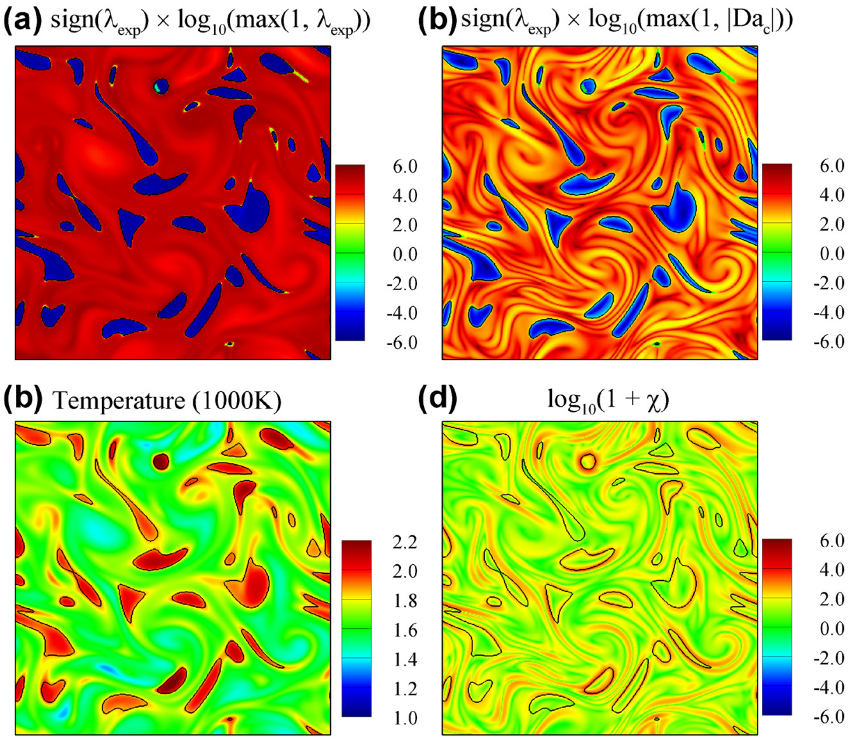

Figure 9. Isocontour of (a) chemical explosive mode timescale, (b) Damköhler number, (c) temperature, and (d) scalar dissipation rate for a 2-D DNS of HCCI combustion of biodiesel/air [9].

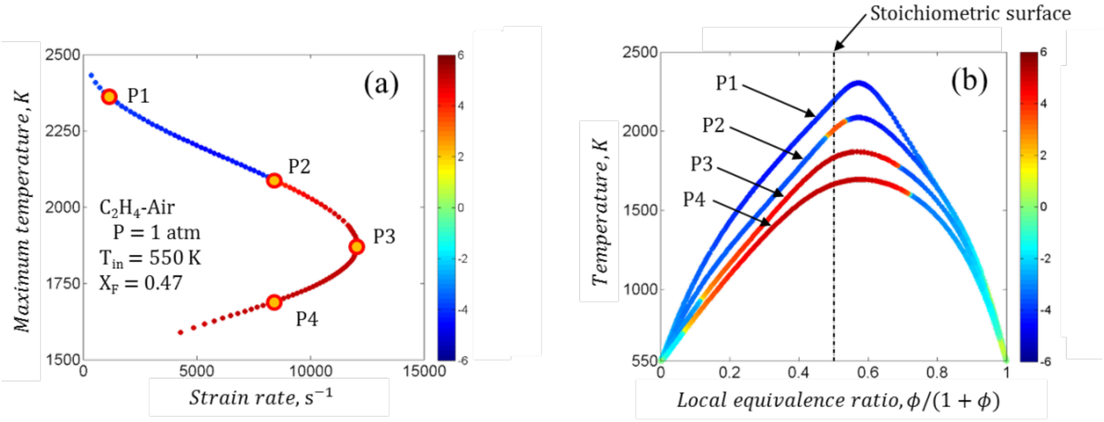

Figure 10. (a) Maximum temperature as function of strain rate in non-premixed flames of nitrogen diluted ethylene/air, and (b) temperature profiles at selected points on the *S*-curve in (a). Color indicates $sign(Re(\lambda_e)) \times log_{10}(1 + |Re(\lambda_e)|)$ at the stoichiometric surface, and λ_e is the eigenvalue of CEM.

Identification of extinction behaviors of 1-D laminar counterflow flames and a 3-D DNS of turbulent non-premixed ethylene/air flame [v] was investigated with CEMA to systematically identify local flame features involved during local extinction and re-ignition.

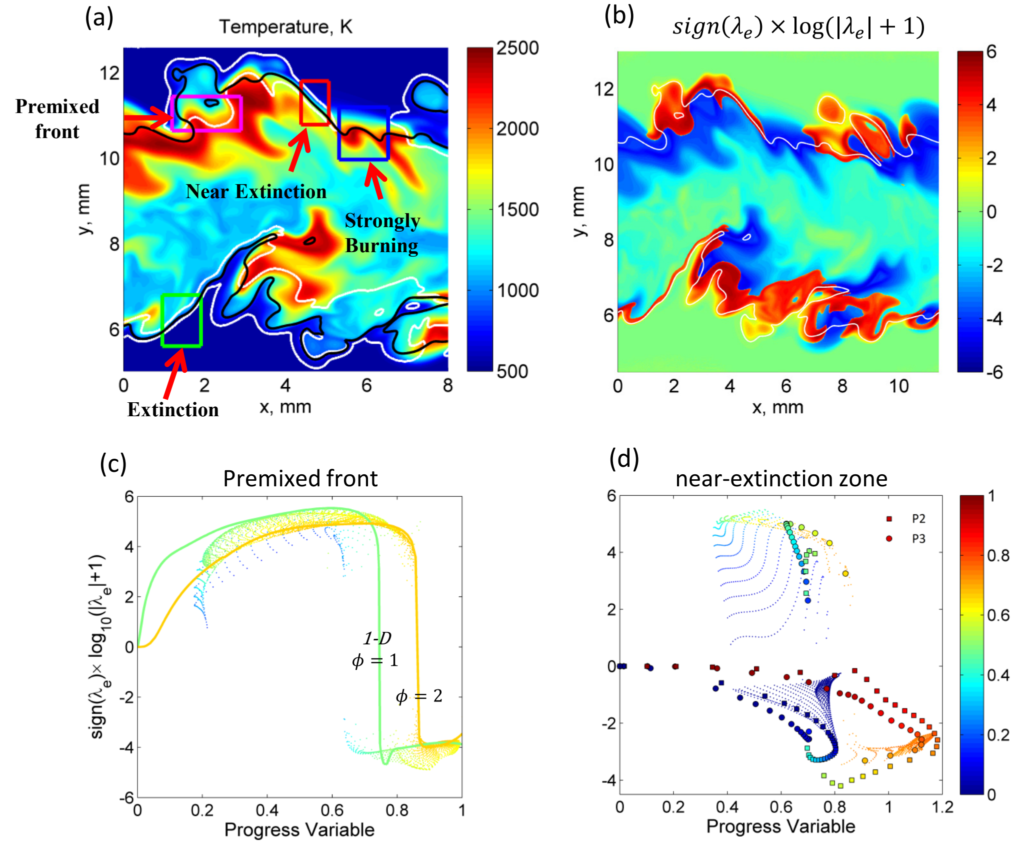

Figure 11. (a) Isocontour of temperature, (b) reciprocal explosive mode timescale, λ_e, (c) scatter of λ_e vs. progress variable in the premixed front zone, the pink box in (a), and d) scatter of λ_e vs. progress variable in the near-extinction zone, the red box in (a), on the 2-D center plane at $t = 0.2\ ms$. The white isoline in (b) indicates the stoichiometric surface. The solid lines in (c) are unstrained 1-D premixed flame solutions, showing the presence of local premixed fronts in the DNS data. The large symbols in d) indicate 1-D non-premixed flame solutions, showing the signature of non-premixed near-extinction flamelets.

c. An explicit CEMA formulation for on-the-fly computational diagnostics

While CEMA can identify critical flame features in both premixed and non-premixed flames, it involves time consuming eigen-decomposition of the chemical Jacobian and thus is difficult to be applied on-the-fly in large-scale flame simulations. To address this issue, a semi-analytic explicit criterion is developed to provide an efficient and robust replacement of CEMA to predict local limit phenomena in complex flow fields. The reactions dominating the CEM are first identified by decomposing the eigenvalue, such that the reactions with negligible effects to the CEM can be eliminated. The coefficients between the reaction timescales, which can be evaluated analytically, and the CEM eigenvalue are then tabulated as function of local thermodynamic quantities, e.g. temperature and local mixture fraction. The explicit CEMA formulation is validated in large eddy simulations of non-premixed *n*-dodecane jet flame into heated air, as shown in Fig. 13. It is seen that the explicit CEMA formulation can accurately capture the zero-crossing of the CEM eigenvalue, which is critical for the diagnostics of limit flame phenomena, while the computational overhead of the explicit formulation is negligible compared with the overall computational cost of practical 3-D flame simulations.

d. CEMA based zone-adaptive modeling of turbulent flames

Turbulent combustion modeling is challenging when complex flame configurations are involved, e.g. being hybrid of premixed and non-premixed. Most current models are typically specific for a particular type of flame, e.g. either premixed or non-premixed flamelet, or either auto-ignition or stirred reactors. CEMA is a reliable diagnostic to identify different flame features in complex flow fields, and it is extended to enable zone adaptive modeling for complex turbulent flames.

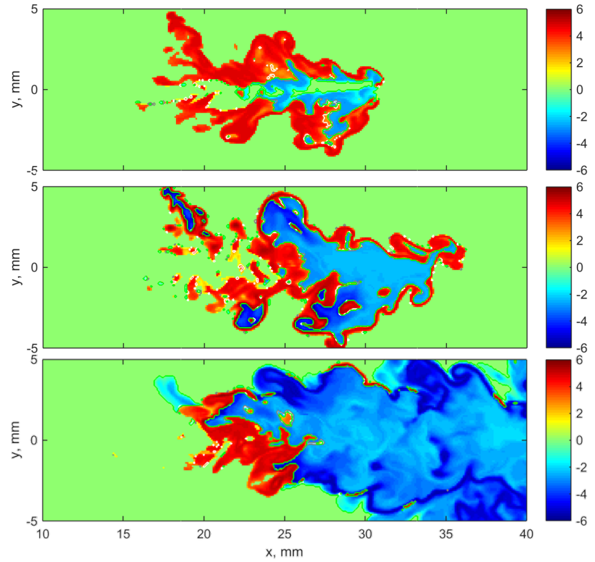

Figure 13. (a) Timescale of CEM calculated with full and semi-analytic explicit CEMA formulation, respectively, for a non-premixed *n*-dodecane jet into air at Spray A condition at different time. Isocontour: full CEMA, isoline: explicit formulation.

The zone adaptive modeling is investigated *a posteriori* in large eddy simulation of a transient lifted *n*-dodecane jet flame into hot air at Spray A condition. The explicit CEMA formulation was employed to identify premixed reaction fronts, and subsequently pre-ignition zone, and post-ignition zones in the flame. The premixed fronts were identified as the zero-crossing of the CEM eigenvalue, pre-ignition zones are identified by the presence of CEM and post-ignition zone was identified by the absence of CEM. Figure 14 shows the temperature isocontour obtained using full chemistry and the progress variable model, a variation of the non-premixed flamelet model, respectively, for post-ignition mixtures identified using the explicit CEMA. Accurate result was obtained using a single progress variable because CEMA accurately identifies the applicable range of the highly simplified progress variable model. Figure 15 further shows the scatter plots of temperature and CO concentration for the full solution and modeled post-ignition zone, respectively. It is seen that the scatter of post-ignition states in the full solution (blue) are accurately captured by the model (green). CEMA-based zone adaptive modeling is shown capable to integrate existing models specific to certain flame conditions by rigorous identification of the applicable zones of such models in complex flow fields.

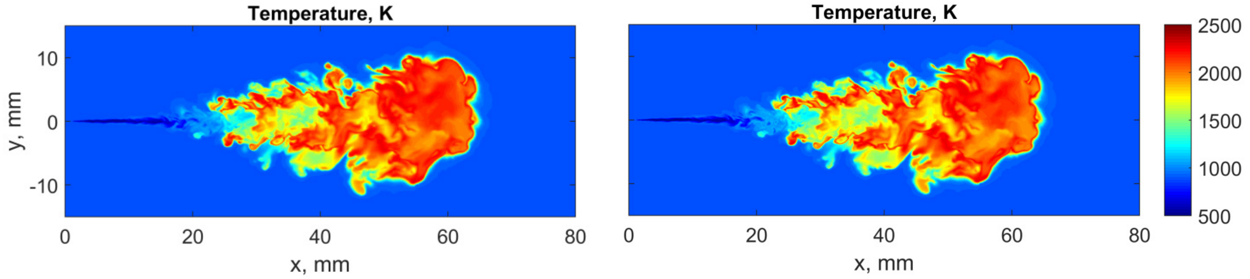

Figure 14. Temperature isocontour of a lifted *n*-dodecane jet flame at Spray A condition solved with full chemistry (left) and a progress variable model for the post-ignition zone identified with CEMA (right).

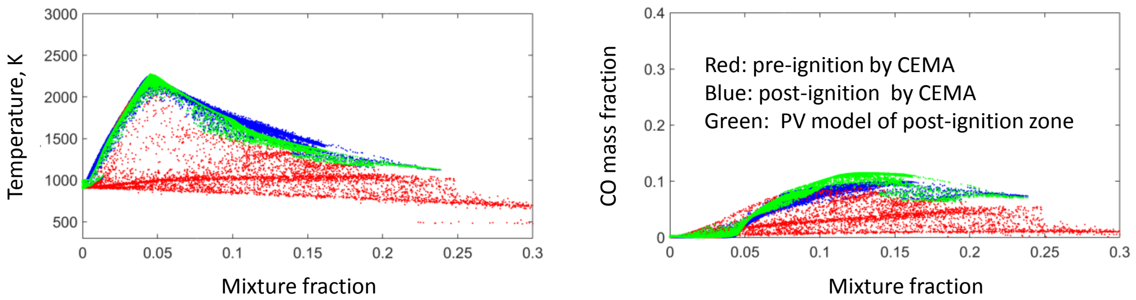

Figure 15. Scatter plots of temperature (left) and CO mass fraction (right) for the lifted *n*-dodecane flame in Fig. 14.

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