

2009 Fall Technical Meeting  
Organized by the Eastern States Section of the Combustion Institute  
and Hosted by the University of Maryland College Park  
October 18-21, 2009

## The Role of Chemical Explosive Mode in Flames

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Chemical explosive mode (CEM) is the eigenmode associated with a positive eigenvalue of the Jacobian of the chemical source term. CEM plays a critical role in combustion systems, especially in limit phenomena, e.g. ignition, extinction and flame propagation, that involve burned and unburned mixtures. A chemical mixture with CEM is intrinsically explosive or unburned and otherwise non-explosive or burned. In the present work, the role of CEM in auto-ignition, perfectly stirred reactors and premixed flames are studied and CEM analysis (CEMA) is employed to classify burned and unburned mixture for the purpose of flow segmentation. For zero-dimensional reactors, it was found that the transition of the eigenvalue associated with the CEM from positive to a negative value is a major factor that determines ignition and extinction. CEMA is further applied to analyze a 2-D slice from a 3-D turbulent lifted ethylene jet flame issuing into coflowing heated air, computed with direct numerical simulation. It was found that auto-ignition is the dominant stabilization mechanism for the lifted flame. The partially premixed flame fronts were accurately located, and the diffusion flame kernel was identified on the CEMA-visualized flame structure.

### 1. Introduction

In conventional flow diagnostics, individual scalars, such as temperature or a species concentration, are frequently employed for detection of critical features in flames such as ignition, extinction, and flame fronts. Such approaches may be subject to counter-examples in general flow analysis. This is because chemical reactions in most combustion systems involve multiple species. Many behaviors of the system are therefore determined by the interaction of multiple variables, including species concentrations and thermodynamic states.

Recently, a method of chemical explosive mode (CEM) analysis (CEMA) was proposed for flow diagnostics involving detailed chemical kinetics [1-2]. CEMA is based on eigenanalysis on the Jacobian matrix of the chemical source term. In combustion research, eigenanalysis has been primarily adopted in analysis and reduction of detailed kinetic mechanisms, with two prominent examples being intrinsic low-dimensional manifold (ILDM) [3] and computational singular perturbation (CSP) [4]. The effect of explosive CSP modes on ignition was studied in igniting systems [5-6], and the existence of explosive modes in heated pre-mixtures was confirmed.

Compared with CSP modes, which may be affected by transport, CEM was defined on the chemical Jacobian, such that CEM is purely a chemical property. Therefore major differences exist in the application of CEMA and CSP modes.

For simple homogenous systems, while it is known that explosive mode plays an important role in ignition, the role of CEM in extinction applications, such as perfectly stirred reactors (PSR), is less well understood. The effect of transport on CEM and the role of CEM in diffusive flames will be further explored in the following.

## 2. CEM in zero-dimensional systems

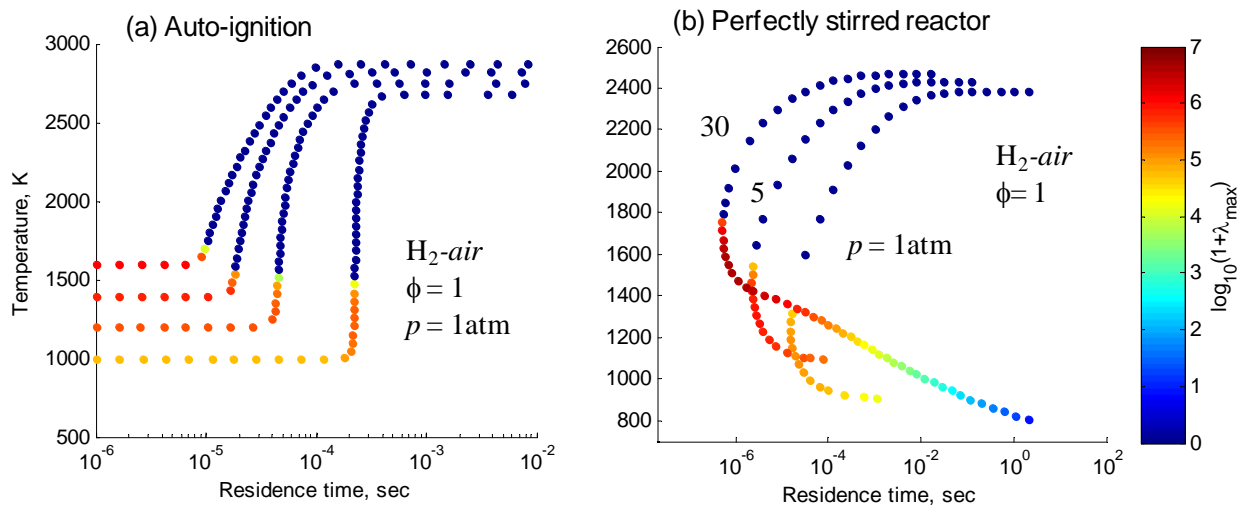
After discretization, the differential equations of a reacting flow can be expressed as:

$$\frac{D\mathbf{y}}{Dt} \equiv \boldsymbol{\omega}(\mathbf{y}) + \mathbf{s}(\mathbf{y}) \quad (1)$$

where  $D/Dt$  is the material derivative,  $\mathbf{y}$  is the vector of dependent variables, such as species concentrations and temperature,  $\boldsymbol{\omega}$  is the chemical source term, and  $\mathbf{s}$  includes all the non-chemical sources such as diffusion. The local chemical information is encoded in the chemical Jacobian matrix  $\mathbf{J}_{\boldsymbol{\omega}}$ . A chemical mode is defined as the eigenmode of  $\mathbf{J}_{\boldsymbol{\omega}}$ , and each mode is associated with an eigenvalue and a pair of (left and right) eigenvectors, assuming that a complete set of eigenvectors exist for  $\mathbf{J}_{\boldsymbol{\omega}}$ . A CEM is the chemical mode associated with a positive eigenvalue, i.e.

$$\lambda_{\text{exp}} > 0 \quad (2)$$

The existence of a positive  $\lambda_{\text{exp}}$  indicates the existence of CEM and consequently an explosive mixture, which typically leads to ignition in spatially homogeneous and lossless environments. To demonstrate the existence of CEM in various homogeneous systems, the time scales of CEM is shown in Fig. 1 for auto-ignition and PSR.



**Figure 1: CEM in (a) auto-ignition and (b) PSR for hydrogen-air. Dark blue indicates the absence of CEM, and red hues indicate the existence of fast CEM, or explosive/unburned mixtures.**

Fig. 1a shows the temperature profiles of stoichiometric hydrogen-air at various initial temperatures, superimposed with the color that indicates the reciprocal time scale,  $\lambda_{\text{exp}}$ , of the CEM. Red indicates the existences of CEM and blue indicates the absence of CEM. The inflection point on each temperature profile is the widely accepted definition of the ignition

point. It is seen that CEM exists in mixtures prior to the ignition points but not in the post-ignition mixtures. Therefore the eigenvalues of the CEMs crosses zero at the ignition points, the separatrix between unburned (explosive) and burned (nonexplosive) mixtures.

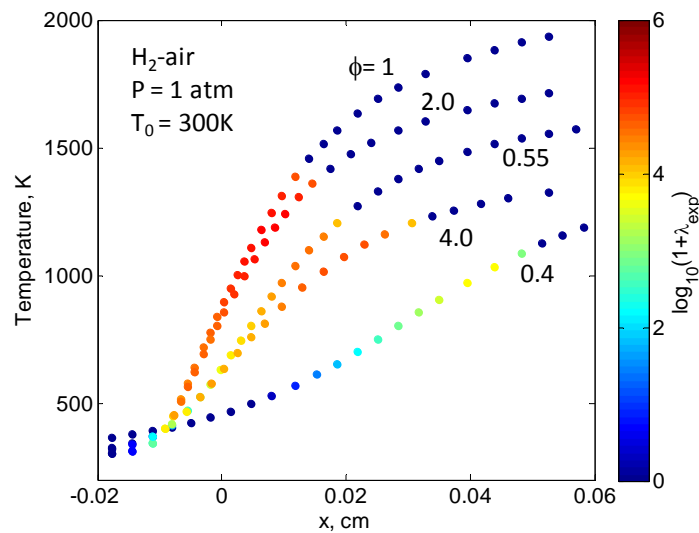
Figure 1b shows the temperature profiles under various pressures for stoichiometric hydrogen-air in PSR, a typical extinction application in steady state. Each profile consists of the upper and middle branches in the typical S-curve of steady state combustion systems [7]. The turning point on each curve is the extinction point since a flame can not be sustained with a shorter residence time beyond this point. The role of CEM in PSR is shown by the imposed color on the temperature profiles. It is seen that above the extinction points, CEM quickly disappear before the mixture approaches the chemical equilibrium as residence time increases. Below the extinction point, CEM exists because the chemical reaction is incomplete due to the decreased temperature and chemical reactivity, such that substantial amount of reactants are unconsumed and the mixture is explosive. Therefore CEM also plays a critical role in both ignition and extinction applications, and it can be utilized to classify burned and unburned mixtures.

### 3. CEM in flames

In strained systems such as diffusive flames, CEM can interact with diffusion or other losses. The relative strength of CEM and diffusion/loss can be quantified with a Damköhler number defined as:

$$D_a = \lambda_{\text{exp}} \cdot \chi^{-1} \quad (3)$$

where  $\chi$  is the scalar dissipation rate in strained flames, or other relevant reciprocal time scales of the loss.



**Figure 2: Temperature profiles of H<sub>2</sub>-air mixtures with various equivalence ratios, superimposed with the time scale of CEM.**

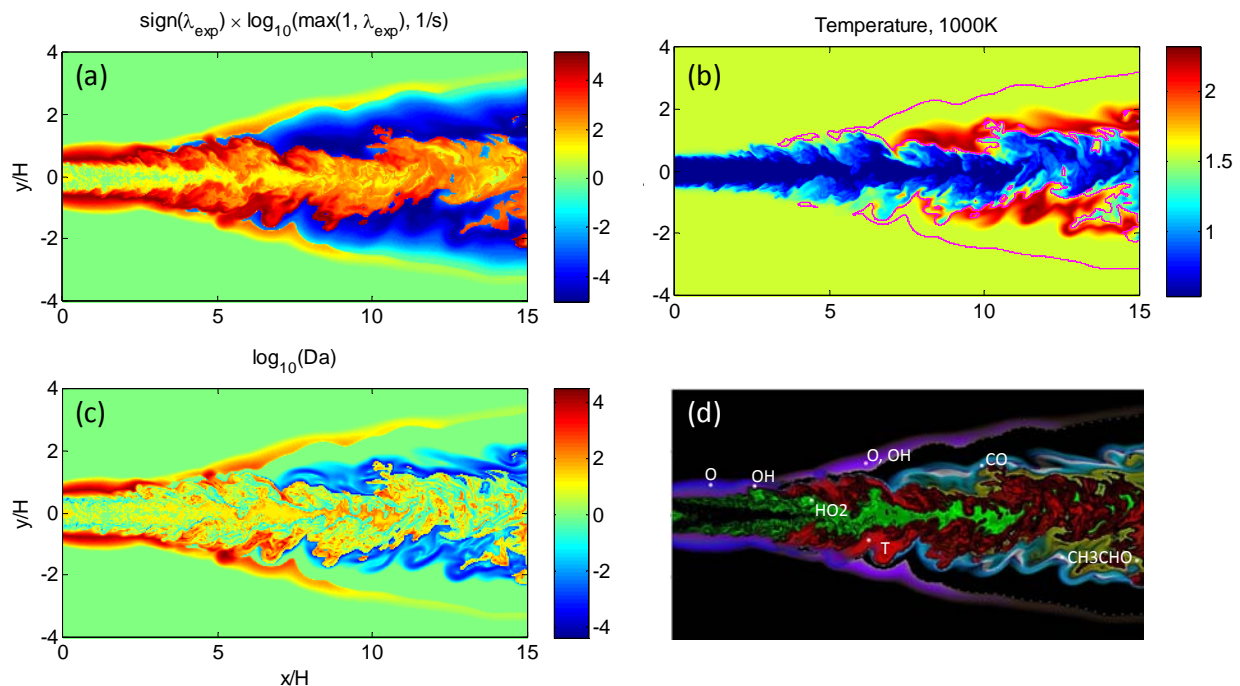
Figure 2 shows the temperature profiles for premixed flames of hydrogen-air under STP with various equivalence ratios. For each flame profile, the cold mixture enters the flame from the left boundary, then into the preheat zone, it subsequently ignites and eventually leaves the flame

while approaching chemical equilibrium. Again, the red hues indicate explosive mixtures with CEM, and it is seen that CEM exists only in the preheat zones, where the mixture is explosive or unburned. Furthermore, the transition from an explosive to non-explosive mixture is abrupt, and the sharp boundary accurately defines the location of the flame front that separates the burned and unburned mixtures. This feature of CEM in premixed flames will be exploited in the following discussion as a flow diagnostic in direct numerical simulations (DNS).

#### 4. CEM for flow diagnostics in DNS

CEMA is then applied to DNS of a lifted ethylene jet flame in a heated coflow of air, simulated by Yoo et al using Sandia's S3d DNS code [8-9]. The DNS required 14 million CPU hours on a Cray XT4 at ORNL with 30,000 CPU cores using a 22-species comprehensive non-stiff reduced mechanism, and generated 240TB of field data and 50TB of particle data. The inlet (center-left) fuel jet consists of 18% ethylene and 82% nitrogen by volume under atmospheric pressure at a temperature of 550K with a jet velocity of 204m/s. The air coflow is at 1550 K with a velocity of 20 m/s. The jet Reynolds number is 10,000 based on the width of the slot:  $H=2$  mm. The size of the computational domain is 30mm by 40mm by 6 mm in the streamwise ( $x$ ), transverse ( $y$ ) and spanwise ( $z$ ) directions, respectively. 1.28 billion grid points were required to resolve both the Kolmogorov length scales and the structure of flames and ignition fronts.

The 2-D center slice ( $z = 0$ ) in Fig. 3 was taken at a time of 2.21ms, when the simulated flame has attained statistical stationarity.



**Figure 3: Structure of the lifted ethylene jet flame in a heated air coflow. (a) time scale of CEM. red indicates unburned mixture ( $\lambda_{\text{exp}} > 0$ ) and blue indicates burned mixture ( $\lambda_{\text{exp}} < 0$ ). (b) isocontour of temperature; the magenta isocontour indicates the premixed flames detected by CEMA. (c) the Damköhler number (eq. 5) defined with CEM, red color indicates auto-igniting zones, blue indicates the diffusion flame kernel. (d) controlling species at different flame zones identified with the explosion index (eq. 6) in CEMA.**

Fig. 3a shows the time scales of CEM. Consistent with the previous discussion, unburned mixtures are shown in red while burned mixtures are in blue. The transition layer between the burned and unburned mixtures is therefore a (partially) premixed flame front. To compare CEMA with the conventional methods based only on temperature, the flame front detected by CEMA is shown as the magenta isocontour in Fig. 3b, superimposed on the temperature field. It is seen that the lean flame front (located well into the heated coflow shown in green) detected by CEMA is indiscernible on the temperature plot due to the high ambient temperature (1550K) in the coflow, and consequently, the extremely lean flammability limit. Furthermore, CEMA detects burned mixtures substantially further upstream compared with the axial location where temperature increase can be observed. This is similar to the observations in a lifted hydrogen jet flame studied previously [2, 8].

Figure 3c shows the distribution of the Damköhler number defined in eq. (3). A large positive  $Da$  indicates that the CEM dominates transport, such that the mixture is auto-igniting, indicated by the red hues in Fig. 3c. A large negative  $Da$  indicates a diffusion flame kernel where the fast chemistry is exhausted and transport is rate-controlling. That auto-ignition is the dominant reason leading to the burned regions downstream of the stabilization points can readily be seen.

To further show the interaction of the chemical species in flames an explosion index is defined in a similar manner to the radical point in CSP [10-11]:

$$\mathbf{EI} = \frac{|\mathbf{E}|}{\sum_{j=1}^N |E_j|}, \quad \mathbf{E} = \text{diag}(\mathbf{A}_{\text{exp}} \mathbf{B}_{\text{exp}}), \quad (4)$$

where  $\mathbf{A}$  and  $\mathbf{B}$  are the right and left eigenvectors, respectively, associated with the CEM. The explosion pointer, EI, is a normalized quantity indicating how parallel a species is to the CEM. The dominant species in different regimes of the flame as identified with the explosion index is shown in Fig. 3d.

## 5. Concluding Remarks

The role of chemical explosive modes in flow diagnostics was demonstrated through several examples including both homogeneous and diffusive systems. It is emphasized that CEM is a chemical property of the local mixture, and as such, CEMA can be efficiently applied in large scale simulations including 3-D DNS or LES where it can be applied on each grid point independently. Nevertheless, transport may interact with CEM, and therefore, the CEM obtained on each local grid point has the transport effect embedded. While qualitative effects of transport on CEM can be estimated with the  $Da$  defined in eq. (3), more accurate quantification requires extended analysis that includes the transport terms. This will be investigated in the future.

## Acknowledgments

The research at University of Connecticut was supported by NSF-PetaApps. The work at Sandia National Laboratories (SNL) was supported by the Division of Chemical Sciences, Geosciences, and Biosciences, Office of Basic Energy Sciences of the U. S. Department of Energy, and the U. S. Department of Energy SciDAC Program. SNL is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the U. S. Department of Energy under contract DE-AC04-94AL85000. The simulation used resources of the National Center for

Computational Sciences (NCCS) at ORNL, which is supported by the Office of Science of the U.S. DOE under contract DE-AC05-00OR22725.

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