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Stability of rich laminar hydrogen-air flames in a model with detailed transport and kinetic mechanisms

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ABSTRACT

The diffusive-thermal stability of rich hydrogen-air combustion waves under ambient and elevated pressures is investigated numerically. The model includes the detailed transport and detailed kinetic mechanism of hydrogen oxidation. Three different kinetic mechanisms are employed: truncated GRI3.0, Warnatz and San-Diego. The critical conditions for the onset of pulsating instabilities are found in the equivalence ratio vs pressure parameter plane in each case. It is demonstrated that the boundaries of stability differ significantly depending on the specific kinetic mechanism. This suggests that finding the critical parameters for the onset diffusive-thermal instabilities experimentally offers a new way for the verification of kinetic mechanisms. A sensitivity analysis is undertaken and allows to identify the five key elementary reactions which are shown to be the same for both stationary and non-stationary flames thus implying that a successful reduction of the reaction kinetics is possible for non-stationary combustion regimes.

1. Introduction

It is known that deflagration in rich hydrogen-air mixtures can propagate steadily until certain critical conditions are reached in the space of parameters, beyond which the traveling combustion waves become unstable due to the pulsating diffusive-thermal instabilities. This can lead to the emergence of various wave patterns observed in experiments [1,2]. More generally pulsating deflagration was detected for the first time in [3], where tests on diluted acetylene flame propagating downward in a steel tube were performed. It was found that once certain critical pressure or dilution is reached flame front started to propagate in self-vibrational mode. It was also reported that the oscillations had behavior of radial and spinning waves. Pulsating radial and spiral waves were observed in flames propagating in tube in diluted mixtures of propane, heptane, and octane with oxygen [4,5]. The experiments were also repeated in microgravity conditions and confirmed that the oscillations were not buoyancy induced. The possibility of acoustic mode excitations was also ruled out since characteristic frequencies of pulsations were of the order of hundred hertz, which is substantially below the frequencies of fundamental acoustic modes in tube. Thus the authors conclude that these modes are direct manifestation of diffusive-thermal instability. In [1,2] spherically expanding flames of rich hydrogen-air/oxygen and lean butane–oxygen–helium mixtures under elevated pressures were experimentally studied. It is found that once the critical conditions are crossed in the parameter space there emerge either bulk oscillation, target pattern or spiral waves.

All the examples mentioned above represent the cases of deflagration waves in mixtures with the Lewis number of deficient component greater than one. The analytical criterion for the onset of pulsations was found in [6] using the activation energy asymptotics for the one-step Arrhenius reaction model as $Z(Le-1)=4+4\sqrt{3}$, where Z is the Zeldovich and Le is the Lewis number. The overall activation energy and Lewis number for hydrogen-air mixtures were calculated in [1], which allowed the authors to obtain the estimate of the critical conditions for the loss of stability based on the Sivashinsky criterion. In [7] the stability of premixed adiabatic flames in rich hydrogen-air mixtures at normal ambient conditions is studied using models with two-step global chain-branching and recombination reaction steps. It is demonstrated that the two-step models are capable of predicting the flame stability and frequency of pulsations of unsteady combustion waves.

In a number of papers [8–14] the detailed reaction mechanisms were used in order to analyze numerically the diffusive-thermal stability of rich hydrogen-air/oxygen flames. It was demonstrated that as the equivalence ratio is increased at certain critical conditions the traveling wave stability is lost and there emerge flame oscillations,

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which evolve to periodic pulsating combustion waves. The critical H_2 molar fraction in the hydrogen-air mixture at normal ambient conditions was found as 75–76% in [11], whereas in [12] larger value around 79% was obtained. In the latter paper it was shown that as the equivalence ratio was increased the amplitude and period of oscillations grew and at certain conditions, period doubling bifurcation occurred. The increase of hydrogen content to 84% resulted in flame extinction. In [10] the results summarized above were qualitatively confirmed, although the onset of pulsations was determined for mixtures with more than 75% of H_2. The flammability limit was also reduced to 82% of hydrogen in fresh mixture. The adiabatic and non-adiabatic flames with radiative heat losses were studied for normal conditions and elevated pressures in [8,9]. For normal ambient conditions in the adiabatic case the onset of pulsations was found at H_2 content about 75.6% and the solution of period two was observed at 76.1%. As the mixture gets richer the oscillations became relaxational and at certain stage flame exhibited extinction. As pressure was increased for the case of adiabatic flames the stability boundary shifted towards smaller values of equivalence ratio.

Thus it is seen that the numerical data for the critical parameters for the onset of pulsating instabilities is scattered. In the case of normal ambient conditions the boundary of stability in terms of the equivalence ratio and the H_2 content vary as 7.16–8.47 and 75–79%, respectively. The frequency of flame oscillations is also obtained within a broad range of uncertainty. In [8,9] it is reported to be around $80Hz$. The earlier papers [10,12] predict much smaller frequencies of about 12–30 Hz.

In this paper the diffusive-thermal stability of rich hydrogen-air combustion waves under normal and elevated pressures is investigated by using the model with the detailed transport and detailed kinetic mechanisms of hydrogen oxidation. The main goal of the paper is to delineate how different reaction schemes correlate in predicting the critical parameter values for the onset of pulsations and the frequencies of flame oscillations. By implementing the sensitivity analysis of the flame front velocity with respect to rates of elementary reactions the role of the chemical kinetics in establishing of oscillating regimes is studied.

2. Methods

The mathematical model and numerical scheme for detailed system integration used in this work has been reported in e.g. [15,16]. It is based on the numerical simulation of the one-dimensional system of conservation equations using detailed chemistry mechanisms and detailed transport models. More specifically, three detailed chemical reaction mechanisms are considered:

- Warnatz's mechanism [15,17] accounts for 38 elementary reactions between 9 - $(H2, H, H2O, H2O2, HO2, N2, O, O2, OH)$ species. Table A.1 shows all elementary steps for the sensitivity analysis.
- The so-called San-Diego mechanism [18] has 9 (the same as in the Warnatz's mechanism) and 42 elementary reactions.
- The truncated GRI3.0 reaction mechanisms [19], where the reactions for hydrogen oxidation are considered only, comprises 56 elementary reactions and 10 species (Argon Ar is present in the mechanism as an additional inert specie).

The detailed molecular transport including thermal diffusion is taken into account (see e.g. [15]). In this case a transport model based on the Curtiss–Hirschfelder approximation is chosen [20,21].

The system is described by mass, energy and species conservation equations for freely propagating flame [15,16]. In order to simplify the mathematical treatment the conservation equations are considered in primitive variables with the density - ρ, the velocity - v, the temperature - T and the species mass fractions w_i, as dependent variables. Furthermore, a Lagrangian type coordinate transformation is applied to reduce the order of the system by dropping the continuity

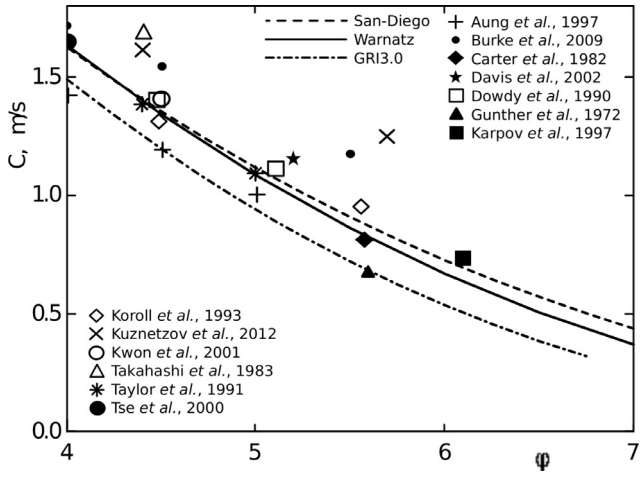

Fig. 1. Flame speed as function of equivalence ratio, ϕ, for normal ambient conditions, $298K$ and $1atm$. The symbols show various experimental [25–36] and numerical [11] data from the literature. The dashed, solid and dash-dotted curves represent the numerical results obtained here with San-Diego [18], Warnatz [15], and GRI3.0 [19] reaction mechanisms, respectively.

equation [15,22]. The quasi-uniform pressure assumption is used due to the low Mach number conditions, which is a very good approximation for the problems considered in this paper.

The numerical solution is performed by the method of lines. Spatial discretization on statically adapted grids leads to large systems of coupled ordinary differential and algebraic equations, which are solved by an implicit extrapolation method with an internal order, step-size control and error-estimation [23,24]. Due to the implicit integration the suggested numerical scheme allows us to simulate transient combustion regimes very accurately. Additionally, a sensitivity analysis is performed by solving the sensitivity equations simultaneously to the system solution.

A sensitivity analysis of the mass flow velocity (ρv) on the rates of elementary reactions (k_i) is performed (see the Appendix). Scaling invariant sensitivity coefficients

$$S_{\rho v,i} = \frac{\partial log(\rho v)}{\partial log(k_i)}, \ (i = 1, \ldots, n_r) \tag{1}$$

are calculated for the stationary regime and along several stable periods of the oscillatory regime of the flame propagation.

3. Results

3.1. Flame speed and structure

In Fig. 1 the normal flame speed, $c = \rho v/\rho_0$, in m/s is plotted against the equivalence ratio for normal ambient conditions i.e. $T_0 = 298K$ and $P = 1atm$. The flame speed is calculated with respect to the fresh mixture and only hydrogen rich mixture compositions are considered. Different symbols show various experimental [25–36] and numerical data [11] known from the literature. The experimental data for adiabatic combustion wave speed is available for equivalence ratio as high as 6. It is generally quite significantly scattered and the uncertainty can reach up to $30-45\%$. The results of numerical analysis is also presented in Fig. 1 with continuous lines. Three different reaction mechanisms are selected for numerical calculations: San-Diego [18], Warnatz [15,17], and GRI3.0 [19] which are shown with the dashed, the solid and the dash-dotted lines, respectively. It is seen in Fig. 1 that the numerical results for all mechanisms lie within the range of experimental data. For values of ϕ around 4–5 there is a very good quantitative agreement between the predictions of San-Diego and Warnatz reactions mechanisms, while the GRI-mech3.0 underestimates the velocity of the combustion wave by 10%. As ϕ