



Chemical kinetic analysis of detonability-enhancing strategies for ethylene–oxidizer mixtures



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ABSTRACT

Four detailed chemical kinetic mechanisms are used in conjunction with an empirical detonation cell width model to numerically assess strategies to increase the detonation sensitivity of ethylene–oxidizer mixtures. Using this method, reasonable agreement is achieved with computed cell width and the available experimental data. Elevated initial pressures significantly reduce cell width for a wide range of equivalence ratios, yielding 80% reduction at stoichiometric conditions for a tenfold increase in pressure. Elevated initial temperatures have almost no effect on the cell width at stoichiometric conditions, but yield 80% reduction at lean conditions when the initial temperature is doubled. Reduced nitrogen dilution within the oxidizer dramatically reduces the cell width for the entire computed range of equivalence ratios. Introducing hydrogen as a fuel additive yields mild improvement to detonation sensitivity at stoichiometric conditions, but requires relatively high H₂ concentrations and is ineffective when coupled with elevated initial pressures. Introduction of supplemental oxygen and increasing the initial reactant pressure appears to be the most effective approach to enhance detonability for ethylene–oxidizer mixtures.

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1. Introduction

Detonation is a supersonic mode of combustion, consisting of a shock wave coupled to a reaction zone. The shock wave is sufficiently strong to initiate chemical reactions due to the high temperature and pressure in the post-shock environment. Precursor, chain-branching reactions drive radical production and culminate in exothermic, termination reactions. Due to strong compression provided by the leading shock, detonation yields a net pressure rise from reactants to products. This type of unsteady “pressure gain” combustion holds the potential for improved thermodynamic efficiency and engine concepts with reduced complexity and size [1].

Experimentally observed detonations exhibit a complex three-dimensional structure, as recorded by the diamond-shaped cellular patterns imprinted onto smoked foils during detonation passage [2]. This structure arises due to nonlinear coupling between shock compression and subsequent heat release, which excites a complex, oscillatory response in the detonation front [3]. This instability manifests macroscopically in the form of strong transverse perturbations, propagating orthogonal to the detonation front [4]. As these transverse waves periodically collide with

one another, a cellular pattern emerges, with the width of the detonation cell (λ) corresponding to the average transverse wave spacing [5]. While the local transverse wave spacing can be highly irregular, especially for fuel–air mixtures, the published data typically report average cell width from these patterns.

The cell width is the most important dynamic parameter of detonations, and is a direct measure of the detonation sensitivity of a mixture [5,6]. It is directly related to the mixture composition and thermodynamic state, with smaller cell width indicating higher detonability (e.g. detonation sensitivity). Transverse wave interactions are necessary for detonation propagation for most mixtures, thus limiting the detonation structure to a minimum size for which these transverse features are supported. For successful onset and propagation, the channel height (h) must exceed one cell width for rectangular geometries ($h > \lambda$) [7], or the tube diameter (D) must exceed one cell width for circular geometries ($D > \lambda$) [8]. For a given confinement, detonations will fail if the cell width exceeds these geometric limits, restricting the successful operating range. Enhancing the detonation sensitivity of the mixture (i.e., reducing λ) extends the limits of detonation propagation for a given confinement, and promotes the development of detonation. A recent numerical and experimental study explores the role of kinetics in the transition to detonation by utilizing a reduced, multi-step acetylene oxidation mechanism [9]. In addition to achieving a close match between experiment and simulation, those results indicate that equivalence ratio (e.g., mixture

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reactivity) is the most dominant factor in determining the regime of detonation formation, even for complex combustor geometries.

Due to the intrinsic link between chemical kinetics and the generation of the cellular structure, Shchelkin and Troshin [10] first postulated that the cell width is proportional to a representative chemical induction length (Δ), (e.g., $\lambda = A \cdot \Delta$, where A is a proportionality constant). By employing the one-dimensional, Zel'dovich-von Neumann-Döring (ZND) detonation model to estimate the post-shock state, the detonation kinetics can be evaluated with a chemical kinetic mechanism. Utilizing this approach, Westbrook and Urtiew [11] calculated the induction length for a wide range of mixtures and related it to a number of macroscopic detonation properties including cell width. Utilizing the experimental data of [12], the proportionality constant was evaluated for fuel-oxygen mixtures as $\lambda = 29 \cdot \Delta$. However, Shepherd et al. [13] reported significant discrepancies when using a single proportionality constant for a wide range of mixtures, suggesting that this relation was not universally valid. Recent studies by Gavrikov et al. [14] and Ng [15] proposed more complex forms of the original relation to achieve better agreement with experimental data. These express the proportionality constant (A) as a function of one or more variables calculated from the ZND solution. While these relations are particularly promising, Schultz and Shepherd [16] illustrate the significant impact of the chemical kinetic mechanism on the computed induction length (which varies by a factor of two, depending on the mechanism).

Ethylene (C_2H_4) is commonly employed in detonation-based combustors [17], and serves as a surrogate fuel for larger, less detonable hydrocarbons. Previous studies have explored the effects of composition and thermodynamic state on the detonability of C_2H_4 -air mixtures. Elevated initial pressure (P_i) significantly boosts detonability [18], though experimental data is somewhat sparse, and numerical data is limited to stoichiometric mixtures at pressures of up to $P_i \leq 2$ bar [16]. The effect of elevated initial temperature (T_i) is less clear and has only received detailed treatment for stoichiometric C_2H_4 - O_2 -Ar mixtures [19], and stoichiometric C_2H_4 -air mixtures of up to $T_i \leq 373$ K (100 °C) [20]. Phylippov et al. [21] utilized a multi-step hydrocarbon oxidation mechanism to simulate the effect of temperature on detonation formation, and assert that there are competing kinetic and physical factors (e.g., promotion of certain chemical reactions, balanced by a reduction in density, and increased mixture sound speed). O_2 addition to the oxidizer (reduced N_2 concentration) has a potent detonability-enhancing effect [8,18,22], yielding reductions in cell width of up to two orders of magnitude for stoichiometric mixtures when pure O_2 is used as the oxidizer. The effect of H_2 as a fuel additive (substitution of various fractions of C_2H_4 with a stoichiometric equivalent of H_2) has only received numerical treatment [23]. For stoichiometric mixtures at ambient initial conditions, a significant effect is achieved for modest H_2 concentrations (50% reduction in the induction length for 10% replacement of the fuel with H_2).

The present study builds upon previous investigations to fully characterize the effects of initial pressure and temperature, oxygen concentration, and hydrogen addition on the detonability of baseline, ethylene-air mixtures. Applying the recently developed empirical approaches of [14,15] and four detailed chemical kinetic mechanisms, the ZND detonation model is used to estimate cell width. While previous studies report these effects for stoichiometric mixtures, this study explores the impact of these parameters for lean and rich mixtures as well. The effect of elevated initial temperature is explored for an extended range (up to $T_i = 600$ K) to resolve the impact on detonability. The effect of elevated O_2 content is explored for oxidizers ranging from pure O_2 to air to explore the benefit of moderate oxygen addition. The effect of hydrogen substitution is explored for a range of initial pressures to

determine whether the significant benefit reported by [23] is preserved at these conditions. The central objective of this work is to assess the effectiveness of these detonability enhancement strategies for C_2H_4 -oxidizer mixtures. This information can be used to augment the operating range of existing detonation combustors, or improve scaling capabilities for future combustors. Conversely, some approaches can be used in reverse to reduce the detonation sensitivity in environments where detonation is undesirable or hazardous.

2. Numerical method

Solutions are computed for the steady, one-dimensional ZND detonation equations in the form developed by Fickett and Davis [24]. This stiff system of ordinary differential equations is solved using MATLAB with a prescribed relative error tolerance of $1e-10$. These computations employ the Shock and Detonation Toolbox developed by [25], and the open-source chemical kinetics software Cantera [26]. The reaction rates and thermodynamic information are evaluated from a detailed chemical kinetic mechanism.

Four chemical mechanisms are employed in the present study: GRI 3.0 [27], San Diego Mech [28], AramcoMech 1.3 [29], and USC Mech II [30]. These detailed mechanisms have been developed for hydrocarbon oxidation and validated against a range of flame and shock tube data. Recently published ethylene-air shock tube data at elevated temperatures and pressures [31,32] allows for additional validation of these mechanisms at conditions representative of detonations. Because of the test procedure employed in these studies, the post-shock pressure within the shock tube varies over the range of temperature ($P = 10.7$ -16.5 atm). An average pressure (P_{avg}) is used to describe the post-shock condition for each data set. Ignition delay time data from [31] is determined simultaneously from the OH emission trace and the pressure history, while ignition delay time from [32] is reported for a number of methods, and the OH emission results are used here. Constant volume simulations are performed for the four mechanisms, using the peak in the OH emission profile to calculate ignition delay time. Three equivalence ratios are simulated for a range of reciprocal temperature ($1000/T = 0.65$ -0.95 K^{-1}) at an average post-shock pressure of $P_{avg} = 13.5$ atm.

The two sets of experimental data, despite slight differences in pressure, exhibit good agreement. The GRI 3.0 mechanism was developed and optimized primarily for methane combustion, but also includes a submechanism for ethylene oxidation. However, it deviates significantly from the other mechanisms at low temperatures, and does not adequately capture the general trend of the data. USC Mech II overpredicts the ignition delay for lean, high temperature conditions, but shows better agreement for rich, high temperature conditions. The slope of the curve (which is proportional to global activation energy [16]) is lower than the slope indicated by the experimental data. San Diego Mech and AramcoMech 1.3 show comparable behavior over this range, capturing the general trend of the data, but underestimating ignition delay for stoichiometric and rich conditions. Approaching the high temperature limit of the simulated range (the minimum value of reciprocal temperature), ignition delays for the four mechanisms agree within a factor of two, but the general behavior of San Diego Mech and AramcoMech 1.3 offer the best agreement with the experimentally observed trends.

Thermicity (σ), as defined by Fickett and Davis [24], is a non-dimensional parameter that "represents transformation of energy obtained from breaking chemical bonds and from the change in the number of particles or molecular sizes by chemical reaction, into energy of heat and motion". Thermicity coefficients corresponding to each species are multiplied by species production

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