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Hot Surface Ignition of Ethylene-Air mixtures: Selection of Reaction Models for CFD Simulations

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The present study investigated hot surface ignition of C₂H₄-air mixtures. The ignition thresholds from a stationary commercial glow plug (active heated area is 9.3 mm in height and 5.1 mm in diameter) were characterized using two-color pyrometry and high-speed interferometry. The minimum ignition temperature decreases from 1180 K to 1110 K as the equivalence ratio increases from $\Phi=0.4$ to $\Phi=4$. The performance of eight reaction models have been quantitatively evaluated using a comprehensive database on auto-ignition. The reaction model of Mével C₁-C₃ and the GRI-mech 3.0 were reduced for inclusion in two-dimensional numerical simulations. Simulations performed with the GRI-mech demonstrate better quantitative agreement for rich mixtures but predicts an opposite trend with Φ as compared to experiments. Simulations with Mével's model demonstrate quantitative agreement for lean conditions, and decreasing performance for $\Phi \leq 1$. However, it reproduces the correct evolution of ignition threshold as a function of Φ .

1 Introduction

The risk of accidental ignition of flammable mixtures by a hot surface and subsequent flame propagation is of particular importance for industry. Such a scenario is relevant to commercial aviation, chemical processes, nuclear energy production, and mining, among other industrial activities. For example, considering typical jet fuels and operating conditions, the fuel tank of an aircraft will contain a flammable mixture during parts of the flight envelope. A scientific understanding of the hot surface ignition process is needed which requires accurate measurements and detailed modeling of chemical reaction kinetics and fluid dynamics. In previous studies from our laboratory [1–3], ignition of n-hexane- and hydrogen-based mixtures has been examined and in this study we are using similar techniques for C₂H₄-air mixtures. Recent studies of C₂H₄-air thermal ignition have also been performed by Beyer and Markus [4] and Roth et al. [5].

The goal of the present study is obtain high quality data on hot surface ignition thresholds for ethylene-air mixtures and quantify the predictive capability of two-dimensional numerical simulations at reproducing the experimental ignition threshold using current chemical kinetic mechanisms.

2 Materials and methods

2.1 Experimental setup and diagnostics

Ignition experiments were performed in a 2.2L vessel. The hot surface, a cylindrical Autolite 1110 glow plug made of stainless steel 316, was mounted vertically in the lower section of the vessel.

The active surface area of the glow plug is 9.3 mm in height and 5.1 mm in diameter. The setup has been described previously in [2,3]. The vessel was filled with ethylene, oxygen and nitrogen using the method of partial pressures with a 10 Pa accuracy. The gases were mixed using a circulation pump and left to settle and reach quiescent conditions. Initial conditions were $P_o = 101.3$ kPa and $T_o = 296$ K. The glow plug was heated by electric current until ignition occurred.

Ignition was characterized in terms of surface temperature at ignition (ignition threshold), and gas temperature field in the glow plug vicinity. Surface temperature was measured by two-color pyrometry (uncertainty +50K/-75K). Gas temperature fields (uncertainty <10%) and ignition dynamics were captured by Mach-Zehnder interferometry using a Spectra-Physics Excelsior-532-200-CDRM 532 nm laser and a Phantom V7-11 high speed camera. Further details on the diagnostics will be given in future publications.

2.2 Reaction models and validation database

Eight reaction models from the literature which include C_2H_4 chemistry were selected: Caltech-Mech (1106 Reactions (R); 169 Species (S)) [6], Dagaut (925 R, 128 S) [7], Galway (689 R, 120 S) [8], GRI-mech 3.0 (325 R, 53 S) [9], JetSurf (2163 R, 348 S) [10], Konnov (1200 R, 127 S) [11], Mevel C_1 - C_3 (920 R, 115 S) [12, 13], Mével C_1 - C_6 (2628 R, 531 S) [14].

To test the predictive capability of the different reactions models, a comprehensive database on C_2H_4 - O_2 -diluent mixtures auto-ignition has been assembled from the literature as summarized by Schultz and Shepherd [15] and Cymbalist [16]. We also considered the data from the Stanford shock-tube database [17]. The database, which includes 964 shock-tube experiments, covers the following ranges: $\Phi=0.125$ -4; $X_{Diluent}=0.685$ -0.99; $P=20$ -4130 kPa; $T=882$ -2339 K. The kinetic targets are: OH^* , CH^* , C_2^* , OH , C_2H_4 , $[O] \times [CO]$, $[CO] + [CO_2]$, pressure, and temperature. Data of Suzuki et al. [18] obtained below 1080 K were removed due to apparent non-idealities in their experiments (strong decrease of activation energy typically seen when pre-ignition compression is taking place during shock-tube experiment [19]).

2.3 Numerical simulations

The motion, transport and chemical reaction in the gas surrounding the glow plug were modeled using the low Mach number, variable-density reactive Navier-Stokes equations with temperature-dependent transport properties [20]. A detailed description of the model can be found in [21]. The governing equations were solved in an axisymmetric 2-D geometry using the OpenFOAM toolbox [22]. Our implementation of the code is well validated as it has been used successfully in various ignition studies comprising different geometries, modes of heat transfer (e.g. forced and natural convection), and ignition timescales [21, 23–26]. The computational domain was discretized with 200,000 cells, compressed near the wall of the glow plug with a minimum cell size of $60 \mu m$, to resolve the thermal and hydrodynamic boundary layers. The initial conditions were $P_o = 101$ kPa, $T_o = 300$ K, $U_o = (0,0)$ m/s, and mass fractions Y_i corresponding to equivalence ratios, Φ , of 0.4, 0.5, 1.0, and 4.0. No-slip boundary condition and constant temperature $T_{wall} = T_o$ were imposed on the vessel walls. On the glow plug surface, a time dependent boundary condition given by $T_{surf}(t) = 300 \text{ K} + \alpha t$ was imposed with a heating rate of $\alpha = 220 \text{ K/s}$.

3 Results and discussion

3.1 Chemical kinetics modeling

Figure 1 a) to c) show typical examples of experimental and calculated characteristic times of reaction, referred to as delay-time in the following, for various C_2H_4 - O_2 -diluent mixtures. The predictions of the different reaction models vary significantly. This aspect is also illustrated in Figure 1 d) for a stoichiometric mixture at $P=100$ kPa and $T_1=300$ K. Above 1200 K, the adiabatic

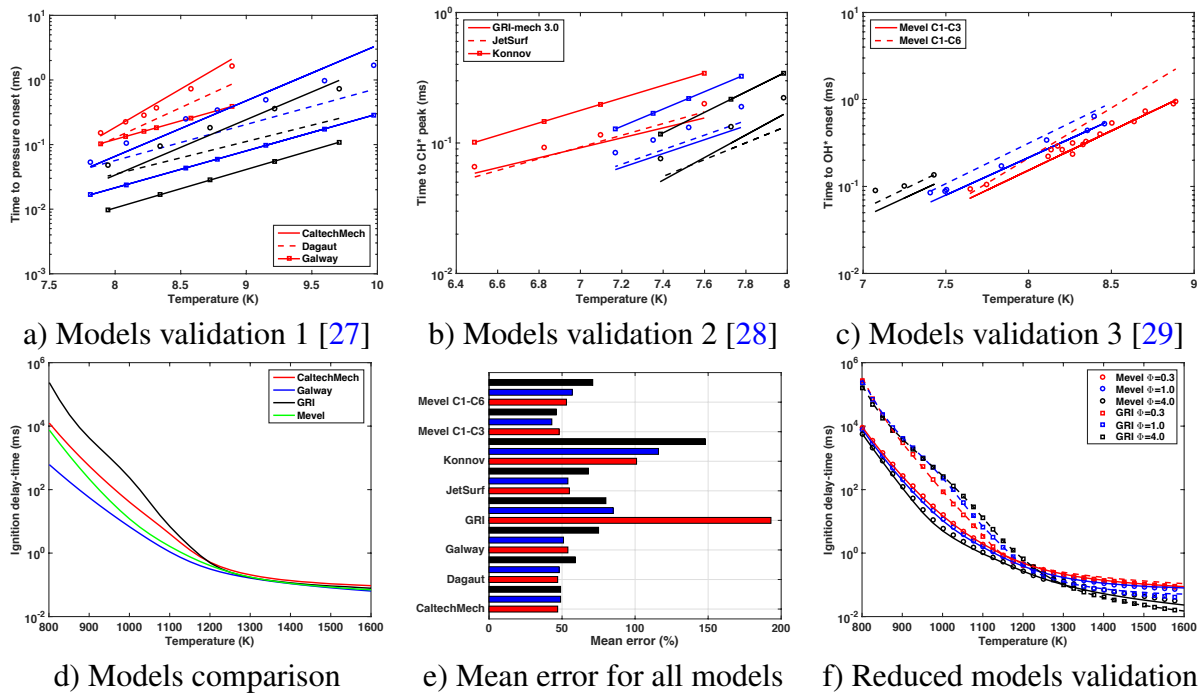


Figure 1: Kinetic modeling for C_2H_4 -air mixtures. In a), b), and c) experimental and numerical delay-times are compared. In a): $\Phi=2$; $X_{N_2}=0.693$; $P=100$ kPa (\circ), $P=1$ MPa (\circ), $P=2.5$ MPa (\circ). In b): $\Phi=1$; $X_{Ar}=0.92$; $P=100$ kPa (\circ), $P=200$ kPa (\circ), $P=400$ kPa (\circ). In c): $P=500$ - 800 kPa ; $\Phi=0.5$ and $X_{Ar}=0.951$ (\circ), $\Phi=0.75$ and $X_{Ar}=0.965$ (\circ), $\Phi=1$ and $X_{Ar}=0.972$ (\circ). In d), the delay-time (time to OH peak) predicted by four models are shown for $\Phi=1$ and $P=100$ kPa. In e), the mean error for all models is shown for three ranges of initial pressure: full range (red), data at $P<500$ kPa (blue), and data at $P<150$ kPa (black). In f), full (symbols) and reduced Mevel C_1 - C_3 (solid lines) and GRI-mech 3.0 (dashed lines) models are compared for mixtures with $\Phi=0.3$, 1, and 4 at $P=100$ kPa.

constant pressure (ADCP) delay-times predicted by all the models converge toward close values with low absolute differences. The range of ADCP delay-time predicted by the different models below 1200 K is bounded by the GRI-mech 3.0, longest delays, and Galway's model, shortest delays. The difference in prediction between these two limiting models can be illustrated by the difference observed at 1000 K. At this temperature, GRI-mech predicts a delay-time around 230 ms whereas Galway's model predicts about 7 ms. Such a difference may results in a significant difference in the predicted hot-surface ignition threshold. The accuracy of the reaction models has been quantified using the mean relative error with respect to the experimental data, as described in Chatelain et al. [13]. The error results are presented in Figure 1 e) for three ranges of initial

pressure: (i) full range, (ii) data at $P < 500$ kPa, and (iii) data at $P < 150$ kPa. The most accurate models were CaltechMech and Mevel C_1 - C_3 with a mean error of respectively 49% and 46% for data at $P < 150$ kPa. Note that these two models exhibit similar errors for the three ranges of initial pressure considered. Given that the Mevel C_1 - C_3 model demonstrates the lowest error, it has been reduced to 100 reactions and 30 species using the approach detailed in Davidenko et al. [30]. In order to provide an upper limit for the predicted hot surface ignition threshold, the GRI-mech has also been reduced to 55 reactions and 32 species. A lower limit for the threshold could be provided by using Galway's model and , we plan to reduce this mechanism as well to complete the present study. The comparison between the full and the reduced models is shown in Figure 1 f) under ADCP conditions. The reduced models reproduce the ignition delay-time from the full models within approximately 10% on average for $\Phi = 0.3$ -7, $P = 100$ kPa and $T_1 = 300$ K.

3.2 Experimental and numerical results

Figure 2 a) shows the typical evolution of the glow plug surface temperature during an ignition experiment. During the first part of the heating phase, $T < 700$ K, the signal is too weak to enable the measurement of temperature with the present pyrometry set-up. At $t = 20$ s, a discontinuity is observed in the temperature signal due to the ignition event. The temperature at this instant, 1165 K, is taken as the temperature threshold. Figure 2 d) shows the profile of the maximum

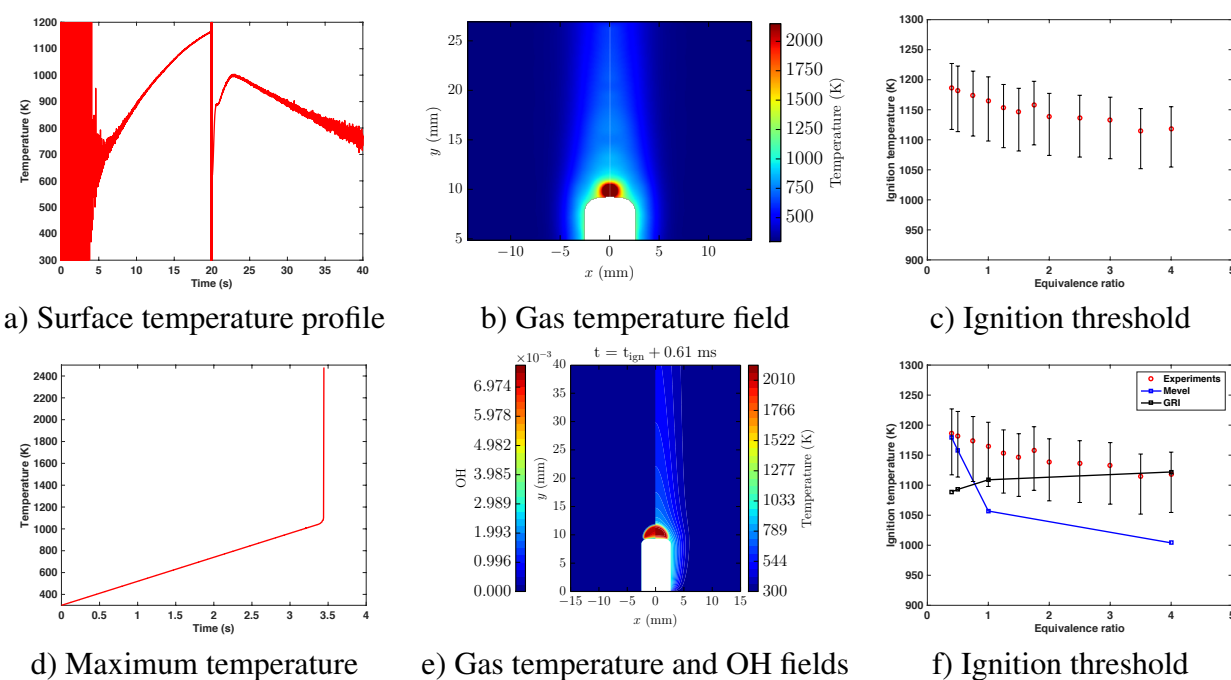


Figure 2: Experimental (top) and numerical (bottom) temperature profiles, temperature fields, and ignition thresholds for hot surface ignition of a stoichiometric C_2H_4 -air mixture initially at $T = 295$ -300 K and $P = 101$ kPa. Calculations were performed with the reduced Mevel C_1 - C_3 model.

temperature in the computational domain. At $t < 3.5$ s, the maximum temperature equals the surface temperature. At $t = 3.5$ s, a sharp increase of the temperature is observed due to gas ignition at a surface temperature of 1057 K. The heating rate is an order of magnitude higher in the simulation

to limit the computational time. The validity of this approach for the present configuration has been established in [21]. As ignition is taking place, a flame kernel develops just above the top surface of the glow plug as shown in Figure 2 b) (experiment) and e) (simulation). The dynamics of the subsequent flame propagation has been studied in detail by Boettcher [2]. Figure 2 c) shows the evolution of the ignition threshold with equivalence ratio over the range $\Phi=0.4-4$. Within this range, the threshold is seen to decrease from 1180 K to 1110 K as Φ is increased. Under similar conditions, Beyer and Markus [4] and Roth et al. [5] measured ignition thresholds in the range 1400-1850 K for laser-heated ceramic spheres of 750 and 800 μm in diameter, respectively. Given the large sensitivity of the ignition threshold to surface size and material, the present results seem consistent with the literature results. The threshold for C_2H_4 -air appears to be in between the thresholds we previously measured for H_2 -air (~ 1050 K) and $n\text{-C}_6\text{H}_{14}$ -air (~ 1275 K) [23] which is consistent with their respective ADCP ignition delay-time, i.e. shorter ADCP delay times at a given temperature imply a lower hot surface ignition temperature. Figure 2 f) compares the experimental thresholds with the predictions of the numerical simulations using the two reduced models. The simulations made using the reduced Mevel's model predict a decrease of the ignition threshold with Φ increase, as observed experimentally, whereas the simulations made with the reduced GRI-mech 3.0 predict an opposite trend, inconsistent with the experiments. The evolution of the threshold for the two reduced models is consistent with the ADCP ignition results shown in Figure 1 f). Quantitatively, the simulations made with the former model reproduce the experimental results within 25 K for the lean mixtures but demonstrate discrepancies for $\Phi=1$ and 4 with more than 100 K under-prediction, whereas the simulations made with reduced GRI-mech 3.0 exhibit opposite trends, i.e. ~ 100 K difference in the threshold for $\Phi < 1$, ~ 60 K for $\Phi=1$, < 10 K for $\Phi=4$.

4 Conclusion

Hot surface ignition of ethylene-air mixtures was characterized by measurements of the ignition threshold using a commercial glow plug over the equivalence ratio range of 0.4 to 4.4. The trends are consistent with previous results for C_2H_4 -air. The quantitative characterization of eight detailed reaction models against a large database of auto-ignition delay-time has revealed significant discrepancies between the experimental and the calculated delay-times. Two-dimensional numerical simulations performed with two reduced models demonstrated agreement only for specific mixture compositions, whereas none of the models could predict thresholds quantitatively across the entire investigated range of equivalence ratio.

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