Homogeneity of Propane/Air Ignition in Shock Tubes: Ignition Delay Times and High-Speed Imaging

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1 Introduction

Non-ideal gas-dynamic effects in shock tubes can have a non-negligible impact on the study of chemical kinetics. In particular, they can induce premature ignition (or pre-ignition) at different locations in the shock tube prior to the main ignition and thus can affect the accuracy of measured ignition delay times (IDTs) and their interpretation. Such inhomogeneities are especially important at long ignition delay times, and thus practically relevant cases of low-temperature ignition (< 1000 K) and low dilution may lead to erroneous IDTs because of strong heat release coupled with gas dynamic effects. Overall, non-ideal effects can affect measured data, making them less suitable for mechanism development as the naïve interpretation of such data can lead to bias in the resulting mechanisms. Therefore, understanding and avoiding non-ideal effects and the reasons for inhomogeneities during ignition in shock tubes has become a major task in the last decade for the combustion community.

To study inhomogeneous ignition in shock tubes, various methodologies have been applied, such as pressure-history measurements with piezo-electric transducers [1], schlieren imaging [2], and high-repetition-rate chemiluminescence imaging [3, 4]. High-repetition-rate luminescence imaging through the end-wall was used to visualize localized ignition and to monitor hot particles [3, 5]. Tulgestke et al. studied homogeneous and inhomogeneous ignition of real and surrogate fuels at low pressures and highlighted that when using helium as diluent gas, the number of hot particles initiating inhomogeneous ignition was reduced [3]. Nativel et al. also used helium as diluent gas in stoichiometric ethanol/air mixtures and could partially mitigate inhomogeneous ignition [4]. Besides, they explain that the ignition is not only influenced by the dilution level but also the type of carrier gas (e.g., N_2 or Ar). Figueroa-Labastida et al. [6] studied ethanol pre-ignition using high-repetition-rate imaging up to 4 bar. They visualized ignition events and concluded that pre-ignition is more likely in mixtures with high ethanol concentration, and that pre-ignition energy release is more pronounced at low temperatures. Furthermore, Nativel et al. [4] explained that in Ar-diluted mixtures, pre-ignition does influence the main ignition event compared to N_2-diluted mixtures and attributed this to the lower thermal diffusivity and heat capacity of Ar (vs. N_2).

Among other hydrocarbons, propane (C_3H_8) was found to be prone to pre-ignition. It was found that ignition delay times of C_3H_8 measured in shock tubes deviate from simulations based on detailed mechanisms at temperatures below 1000 K and at nominal pressures ranging from 6 to 40 bar [6]. In this context, collaborative work between the University of Duisburg-Essen (UDE) and Texas A&M University (TAMU) has been undertaken with multiple aims. In a first part of this work, conditions where inhomogeneous or homogeneous ignition events are encountered are identified by measuring ignition delay times in several C_3H_8/O_2/diluent mixtures over a wide range of conditions in the high-pressure shock-tube (HPST) facility at UDE. Second, the effect of helium on the ignition is studied by substituting N_2 and Ar diluents with He (up to 20 %). Finally, some conditions were reproduced at TAMU and investigated by high-speed imaging technique to visualize the ignition events through the shock tube endwall prior and during the ignition. A comparison between the experimental results and the simulation performed using the kinetics model of the CRECK Modeling Group (C1–C3) [7] is also provided.

2 Methods / Experimental

The HPST-UDE was presented in previous publications [4, 8]. It has a constant inner diameter of 90 mm, and the lengths of the driver and driven sections are 6.4 and 6.1 m, respectively. The two sections are separated by an aluminum diaphragm (1.5 mm thick). The maximum test time can be extended up to 15 ms by driver-gas tailoring (adjusted He/Ar mixtures). Four pressure transducers (PCB 112A05 or 112A03) are used to determine the speed of the incident shock wave from the arrival time of the pressure rise at the known locations of the transducers. Post-shock conditions were calculated using the 1D shock-wave equations with an uncertainty on T_5 of about 15 K. The measurement plane is 16 mm from the endwall and has four ports in the sidewall. In the bottom position, the fourth pressure transducer (PCB 112A03, insulated with RTV silicone (to minimize heat transfer effects) was mounted to record the pressure history. For this study, a total of three photomultiplier tubes (PMT, Hamamatsu 1P28) are mounted in combination with quartz windows at the end- and side wall of the shock tube for spatially-integrated detection of OH* and CH* chemiluminescence selected by 310±5 and 430±5 nm FWHM bandpass filters, respectively. In the top position and in the left side wall (view from the endwall), two PMTs are mounted for OH* detection (PMT 1 and 2, respectively). A third PMT is located at the center of the endwall (PMT 3) for CH* detection. Such a configuration helps in identifying the location of ignition events by detecting them from different perspectives (end- and side-wall detections). Spatially integrated chemiluminescence signals were transmitted via fiber bundles to PMT 2 and 3. IDTs were determined as the time between the rise of the reflected shock wave from the fourth pressure transducer intersection of the tangent to 0 of the steepest rise in OH* signal received from PMT 1.

The TAMU Aerospace shock tube (AST) and the high-pressure shock tube (HPST-TAMU) facilities were used to test Mix 3 (see Table 1) at a fuel-lean equivalence ratio of $\phi = 0.5$. The AST is constructed of a 3.13-m long driver and a 7.33-m long driven section with a 16.20-cm inner diameter. The HPST-TAMU possesses a 2.51-m driver and a driven section that is 4.92 m in length. The internal diameter of the driver is 7.62-cm in both facilities. The driver and driven sections are separated by a breech assembly which houses the 1.02-mm polycarbonate (Lexan) diaphragms used to generate the pressure-driven shock waves. The incident shock wave velocity was detected using four PCB P113A pressure transducers, which are positioned at equidistant intervals along the last 1.44 m of the driven section in both tubes. The reflected-shock conditions were calculated using one-dimensional normal shock-wave relations, as the initial conditions of the test gas were recorded for each test. To ensure purity of the gas mixtures, the shock tubes were evacuated to near-vacuum pressures of ~1×10^{-6} mbar in between experiments. Ignition events were monitored using both sidewall pressure and OH* chemiluminescence diagnostics. Ignition delay time was defined as the time difference from the steepest rise in sidewall pressure and the arrival of the reflected shock wave at the endwall. The OH* signal was used to identify unusual ignition events (pre-ignition, two-stage ignition) and to define ignition if the rise in pressure was minimal in the lower-temperature experiments. More details regarding the design of these facilities can be found elsewhere [9, 10]. A high-speed endwall imaging system was set up on the HPST-TAMU. The endwall window assembly consists of a 17.8-cm diameter, 25-mm thick sapphire window placed at the end of the driven section of the shock tube. The window is held in place inside a 304 stainless steel housing using an ABS 3D-printed shim and stainless-steel spacers and endcap. High-speed imaging is made possible by implementing a high-speed camera (Photron, Fastcam SA-Z) coupled with a LaVision high-speed intensified relay optics (HS-IRO) S20 unit; a UV camera lens (Halle, OUC 2.50); and an optical bandpass filter for OH* centered at 315±7.5 nm (Semrock, FF01-315/15-50). The camera is placed at 90° to the shock tube, and the use of a mirror allows for viewing down the length of the shock tube. Images for this study were taken at a frame rate of 20 kfps.

A total of six mixtures were prepared manometrically using the partial pressure method. The mixtures are presented in Table 1. N_2 and Ar were used as diluents, with the ratio of diluent and O_2 being maintained at 3.76:1. Two equivalence ratios (ϕ = 0.5, 1.0) were investigated for each diluent at ~6 bar from 900 to 1400 K. He was used in Mix 5 and Mix 6 to substitute Ar with 10.0 and 20.0%, respectively, taking as reference Mix 3 and keeping constant the ratio of diluent/O_2 at 3.76:1.

Table 1: Composition of investigated reactant gas mixtures.

Number	ϕ	Composition (mol%)
Mix 1	0.5	2.06 % C_3H_8 + 20.57 % O_2 + 77.37 % N_2
Mix 2	1.0	4.03 % C_3H_8 + 20.15 % O_2 + 75.82 % N_2
Mix 3	0.5	2.06 % C_3H_8 + 20.57 % O_2 + 77.37 % Ar
Mix 4	1.0	4.03 % C_3H_8 + 20.15 % O_2 + 75.82 % Ar

Simulations were performed with Chemical Workbench [11] using a "predefined pressure profile" reactor model which account for the measured pressure increase d*p*/d*t* for a non-reactive run. A constant pressure was observed after shock contact-surface interaction. In this manner, just facility-related, non-ideal effects are corrected for but not the pressure rise due to pre-ignition. The reaction mechanism from the CRECK Modeling Group (CRECK C1–C3) [7] was used for the simulations. Ignition was determined from the time of the steepest rise in simulated OH* concentrations.

3 Results and discussion

N_2-diluted mixtures: Figure 1a shows IDTs of Mix 1 (ϕ = 0.5) and Mix 2 (ϕ = 1.0) performed at UDE in comparison with the simulation at p_5 ~ 6.5 bar and temperature from 900 to 1400 K. No ignition was observed at T_5 < 990 K and < 940 K for Mix 1 and Mix 2, respectively. The IDTs of Mix 2 are shorter than those of Mix 1 at T_5 < 1150 K, which is also predicted by the simulation. Good agreement between experimental and simulation results is observed for Mix 1 over the entire range of temperature. However, a large deviation between experiments and simulation is observed for Mix 2 for T_5 < 1050 K. It is to be noticed that relatively low d*p*/d*t* were observed for the N_2-diluted experiments (typically < 2.5 %/ms) and as mentioned in section 2, the simulation takes into account the gradual increase of pressure. It means that either the model does not work well at ϕ = 1.0, or inhomogeneous ignition has occurred. To understand the reasons for such a deviation, time-resolved information in terms of pressure, OH* and CH* profiles is important to consider.

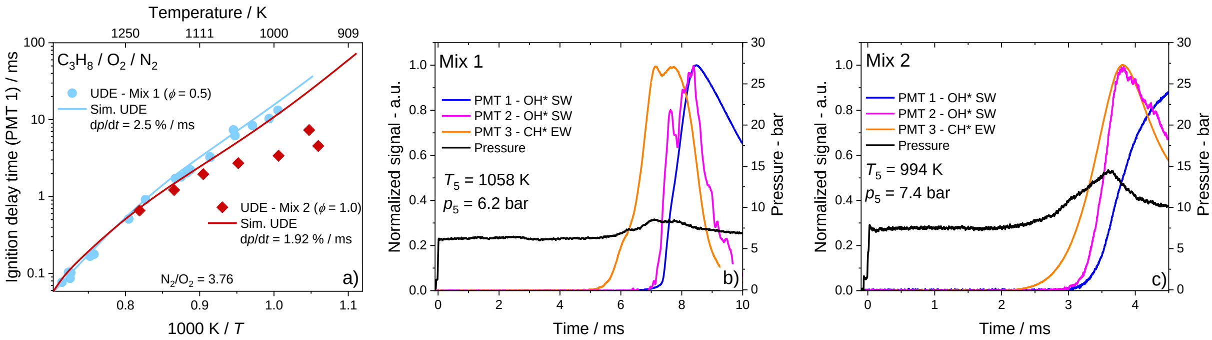

Figure 1: a) IDTs of Mix 1 and Mix 2. Simulation is based on the CRECK C1–C3 mechanism [7]. b, c) Representative signal traces where remote ignition for Mix 1 and Mix 2 (HPST-UDE) is observed.

Figures 1b and c present some time-resolved traces (pressure and PMTs signals) at 1058 and 994 K for Mix 1 and Mix 2, respectively. A timing mismatch is observed in these two cases. First, weak ignition from the pressure profile is observed which is starting at the same moment as the increase in the PMT 3 signal. Later (after 2–3 ms), the other PMT's signals (from the side wall) start to rise, which indicates that ignition starts remotely away from the endwall of the shock tube. This indication of remote ignition is surprising since there is no visible effect of inhomogeneous ignition in the IDT plot presented in Fig. 1a. In the case of Mix 1, similar remote ignition is observed coupled with a slightly stronger ignition. This observation can mean that the stronger the remote ignition, the larger the deviation between measurement and simulation.

Such analysis on the pressure and PMT profiles was performed for each case. It resulted in the observation that the lower the temperature, the more important is remote ignition, the more the deviation with model is seen. However, only strong ignition (homogeneous) events were observed for temperatures T_5 > 1100 K

Ar-diluted mixtures: Figure 2 shows the ignition delay times of Mix 3 measured at UDE and TAMU and Mix 4 measured at UDE together with the predictions of the simulation. The UDE data show a clear deviation from the model and for temperatures lower than 1050 K for both mixtures. The TAMU data obtained in the AST agree with the CRECK model well at higher temperatures but begin to diverge at temperatures lower than 1120 K, while data obtained in the HPST-TAMU agree with the model for temperatures in the range of 1048–1072 K. At lower temperatures, large deviations between measurement and simulation are observed in the HPST-TAMU with IDTs values reaching the one of the HPST-UDE. In general, there is good agreement between the UDE and TAMU data in terms of both IDT values and the occurrence of inhomogeneous ignition events.

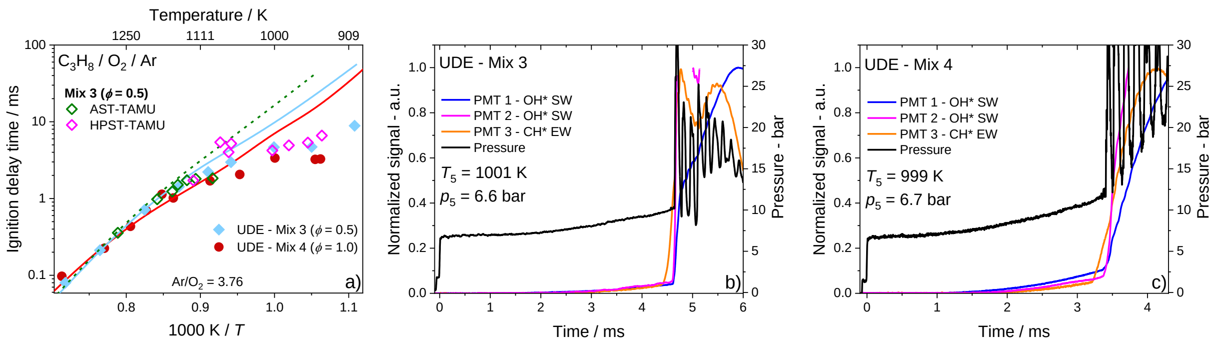

Figure 2: a) IDTs of Mix 3 (UDE and TAMU) and Mix 4 (UDE). Simulation (short green dashed-line for TAMU and red and blue full lines for UDE) is based on the CRECK C1–C3 mechanism [7]. b, c) Representative signal traces at ~1000 K for Mix 3 (left) and Mix 4 (right) in the HPST-UDE.

Non-ideal effects such as d*p*/d*t* and pre-ignition, especially at colder temperatures, are responsible for disagreements between the data and the model. These effects are consistent with the non-ideality observed in the UDE experimental traces resulting in reduced ignition delay times. The largest difference between the TAMU-AST data and the CRECK model is found at 1091 K. An analysis of the pressure and chemiluminescence profiles revealed that this difference was essentially caused by facility effects (d*p*/d*t*). Figures 2b and c show representative traces of inhomogeneous ignition observed during the UDE experiments for Mix 3 and Mix 4. Strong pre-ignition starting very early (at half-time of the IDT) is observed. The signals of both pressure and chemiluminescence are rising at the same time, indicating that the pre-ignition event likely occured at the endwall, discarding the remote ignition reason.

Effect of He on ignition: Helium can mitigate inhomogeneous ignition when added to a reactive mixture [3, 4]. Mix 3 was selected among the other mixture because it showed more inhomogeneities. Therefore, argon was substituted with He in a proportion up to 20 mol%. The results are shown in Fig. 3a.

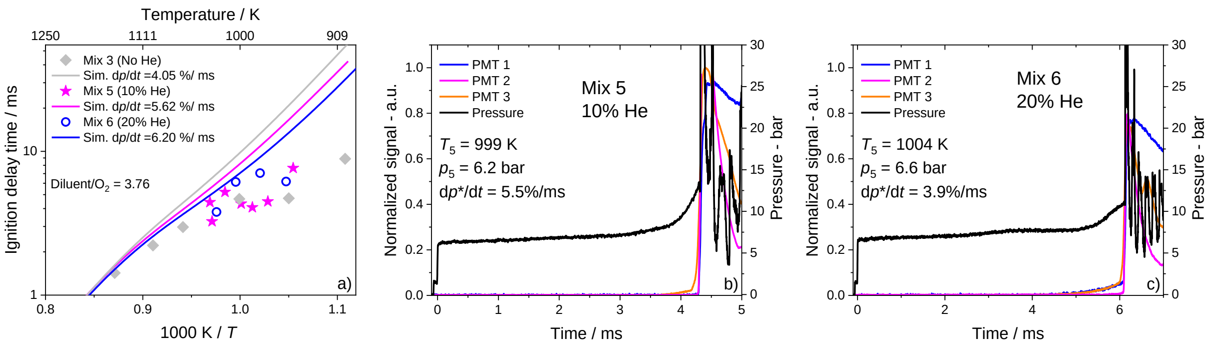

Figure 3: Effect of He on the IDT. a) Comparison between IDTs of Mix 3, 5, and 6 (UDE). Simulation is based on the CRECK C1–C3 mechanism [7]. b, c) Representative signal traces at ~1000 K for Mix 5 and Mix 6 recorded at UDE.

If we compare just the experimental results, we observe that having 20 %He in the mixture results in longer IDTs. The other important information is about the simulation which was performed implementing the reactor model, the d*p*/d*t* of each mixture. For example, taking three cases at ~1000 K, good agreement is observed between the simulated values and the experimental results of Mix 6 when a deviation >50 % is noticed between Mix 3 results and the simulated results. Figures 3b and c show signal traces of two cases near 1000 K for Mix 5 and 6. We can observe that the pre-ignition is reduced as the amount of helium increases in the mixture (the case with 0 % He is shown in Fig. 2b). Finally, homogeneous post-shock conditions are shown in Fig. 3c with first a d*p*/d*t*, then a plateau (characteristic of driver-gas tailoring) and then a small pre-ignition followed by the main ignition event.

High-speed imaging: The images presented in Fig. 4 show the time histories of the shock tube experiments as well as the ignition sequence as captured by imaging through the endwall for an inhomogeneous case at ~1000 K in the HPST-TAMU. Starting in at *t* ~ 2.3 ms, there is evidence of OH* emission visible at three locations on the left and right sides of the shock-tube cross section. This visible OH* emission is a sign for pre-ignition before the main ignition event occurring at *t* ~ 4.2 ms.

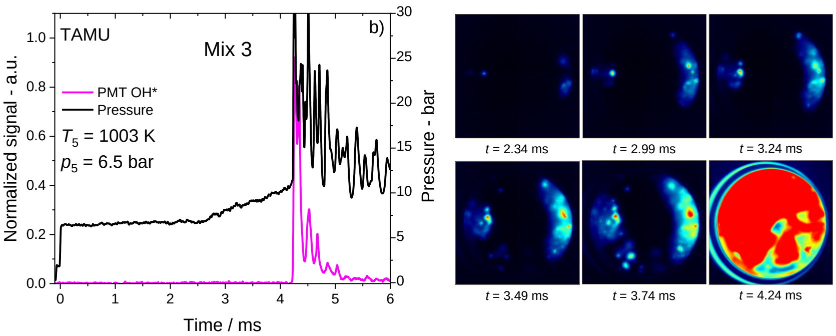

Figure 4: Inhomogeneous ignition case. Sidewall pressure and OH* emission data together with the corresponding endwall OH* emission imaging recorded in the HPST-TAMU for Mix 3.

4 Conclusions

Ignition delay times of propane/O_2/diluent mixtures were measured over a wide range of conditions in the UDE and TAMU shock tubes. N_2 and Ar were used as diluents, with the ratio of diluent and O_2 being maintained at 3.76. The temperature ranged from 900 to 1400 K, the nominal pressure was ~ 6 bar, and two equivalence ratios were investigated (ϕ = 0.5 and 1.0). These experiments allowed the team to identify conditions where inhomogeneous or homogeneous ignition events are encountered. It was found that the IDTs of the N_2-diluted mixture at ϕ = 0.5 agreed well with the simulation results even if remote ignition was detected looking at the time-resolved traces (pressure and PMT). The Ar-diluted mixtures presented inhomogeneous ignition for temperatures <1050 K (except for the large diameter tube HPST-TAMU). The inhomogeneities were attributed to large dp/dt for the AST shock tube and from a large dp/dt and pre-ignition for the HPST-UDE. The Ar-diluted (ϕ = 0.5) mixture was modified substituting Ar with He (up to 20 %) to potentially mitigate the inhomogeneities. At lower temperatures, a considerable discrepancy was noticed between the experimental results and the simulation performed using the kinetics model of the CRECK Modeling Group (C1–C3), but it was possible for some conditions diluted in He to mitigate inhomogeneous ignition. High-speed imaging performed in the HPST-TAMU revealed that local pre-ignition starting from the side wall is responsible for inhomogeneities when Ar dilution is used. In future work, the test conditions will be extended to ϕ = 2.0, and the effect of He as a diluent will be tested at different equivalence ratios together with the high-speed imaging setup.

5 Acknowledgments

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