



# New insights into the shock tube ignition of $H_2/O_2$ at low to moderate temperatures using high-speed end-wall imaging



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## ABSTRACT

In this work, the effects of pre-ignition energy releases on  $H_2-O_2$  mixtures were explored in a shock tube with the aid of high-speed imaging and conventional pressure and emission diagnostics. Ignition delay times and time-resolved camera image sequences were taken behind the reflected shockwaves for two hydrogen mixtures. High concentration experiments spanned temperatures between 858 and 1035 K and pressures between 2.74 and 3.91 atm for a 15%  $H_2/18\% O_2/Ar$  mixture. Low concentration data were also taken at temperatures between 960 and 1131 K and pressures between 3.09 and 5.44 atm for a 4%  $H_2/2\% O_2/Ar$  mixture. These two model mixtures were chosen as they were the focus of recent shock tube work conducted in the literature (Pang et al., 2009). Experiments were performed in both a clean and dirty shock tube facility; however, no deviations in ignition delay times between the two types of tests were apparent. The high-concentration mixture (15% $H_2/18\%O_2/Ar$ ) experienced energy releases in the form of deflagration flames followed by local detonations at temperatures < 1000 K. Measured ignition delay times were compared to predictions by three chemical kinetic mechanisms: GRI-Mech 3.0 (Smith et al.), AramcoMech 2.0 (Li et al., 2017), and Burke's et al. (2012) mechanisms. It was found that when proper thermodynamic assumptions are used, all mechanisms were able to accurately predict the experiments with superior performance from the well-validated AramcoMech 2.0 and Burke et al. mechanisms. Current work provides better guidance in using available literature hydrogen shock tube measurements, which spanned more than 50 years but were conducted without the aid of high-speed visualization of the ignition process, and their modeling using combustion kinetic mechanisms.

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## 1. Introduction

Hydrogen is used as a fuel in many combustion applications including rockets, power generation gas turbines, and ground vehicle transportation [5–9]. Hydrogen is also the simplest hydrocarbon fuel in terms of its combustion chemistry which has been studied for the last 60 years in various combustion experiments including shock tubes, flames, rapid compression machines (RCM), flow reactors, etc. [4,10–26].

Many researchers have hypothesized that inhomogeneities—impurities, particles and facility surfaces – can influence autoignition of hydrogen containing fuels at low temperatures (in the premixer,  $T \sim 600\text{--}1000\text{ K}$ ). Contamination on experimental surfaces

reduced  $H_2$  ignition delay times when compared to a clean tube [27]. Surfaces can also promote chemical induction by catalytic enhancements in  $H_2$  systems [28]. However, there is little knowledge on whether the effect of heterogeneous activity inhibits or promotes homogeneous ignition [29–32]. Impurities in facilities can come from many sources including the vacuum system, metal surfaces, etc. [33,34] and their levels are typically on the order of a few ppms. Unfortunately, the autoignition in  $H_2$  mixtures are greatly sensitive to the presence of impurities, for example, with the addition of just 10 ppm impurity (modeled as H atoms) the ignition time scales are drastically reduced from 4 ms to 1 ms at 4 bar and 1000 K (simulation with CHEMKIN-PRO [35] and AramcoMech 2.0 [3]). Hence the validity of ignition delay time measurements and their influence on facility cleanliness must be characterized. In this study, impurities in the form of diaphragm particles are assessed for their influence on hydrogen ignition.

Shock tubes have been used to study combustion chemical kinetics since the 1940–s [36–43] due to their advantages in

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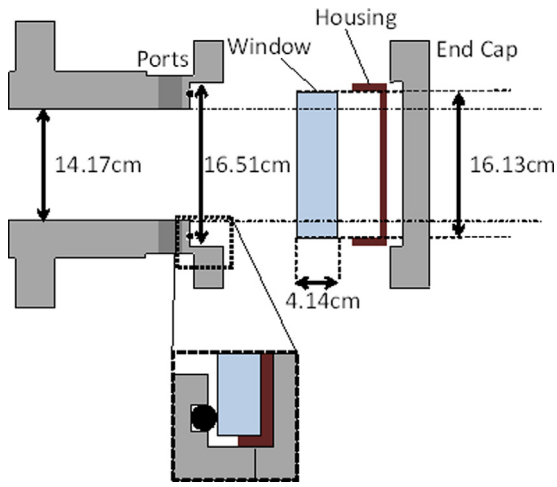


Fig. 1. Schematic of the shock tube and end-wall imaging setup.

providing experimental data that are de-coupled from the effects of heat transfer (i.e. adiabatic wall) and fluid mechanics (i.e. minimal boundary layer). It is important to use the correct thermodynamic assumptions while modeling shock tube data [44–46]. Typical assumptions are that the shock heated gas mixtures contain a homogeneously distributed quiescent gas behind the reflected shock wave and time scales for experiments are on the order of a few milliseconds. Therefore, experimentally obtained shock tube data are typically compared to kinetic model predictions determined under the assumptions of constant internal energy and volume (U, V). However, recent shock tube and rapid compression machine (RCM) studies suggest that some fuels may not exhibit uniform characteristics under certain conditions, and instead undergo pre-ignition [1,47–50], thus complicating the interpretation of measurements.

Energy releases that are present before homogeneous combustion are extremely unfavorable, as they have been observed to increase the pressure and temperature of the unburned portion of the fuel mixture [50], which can lead to decreases in ignition delay time. There has been a push in recent years to understand this phenomenon through images that show the uniformity of combustion. Walton et al. [50] studied iso-octane at temperatures between 903 and 1020K and pressures between 8.7 and 16.6 atm in an RCM and captured high-speed images through a window that allowed optical access to the test manifold. Herzler et al. [51] captured a single ICCD image of n-heptane ignition through an end-wall in a low-pressure shock tube. Wang et al. [49] examined the pre-ignition to super-knock transition for iso-octane mixtures in an RCM. Fieweger et al. [52] examined the self-ignition of several spark-ignition engine fuels through shadowgraph images captured along the length of the test section in a square shock tube. Vermeer et al. [53] also captured shadowgraph images of n-heptane and iso-octane combustion behind post-reflected shockwaves through the length of the test section. Troutman et al. [54] captured the combustion of n-heptane via high-speed, end-wall OH\* chemiluminescence images in a shock tube. Very recently, Pryor et al. [38] examined methane ignition delay times in shock tubes under heavy bifurcation in CO<sub>2</sub> diluted mixtures using multiple diagnostics. It was shown that for mixtures with large amounts of CO<sub>2</sub> dilution the ignition was no longer homogeneous and the bifurcation of the shock wave was extreme leading to increased uncertainty in the measurement.

For hydrogen, one recent study by Pang et al. [1] observed severe ramp-like increases in pressure prior to ignition, which is a strong indicator of non-idealities in the post-reflected shock region.

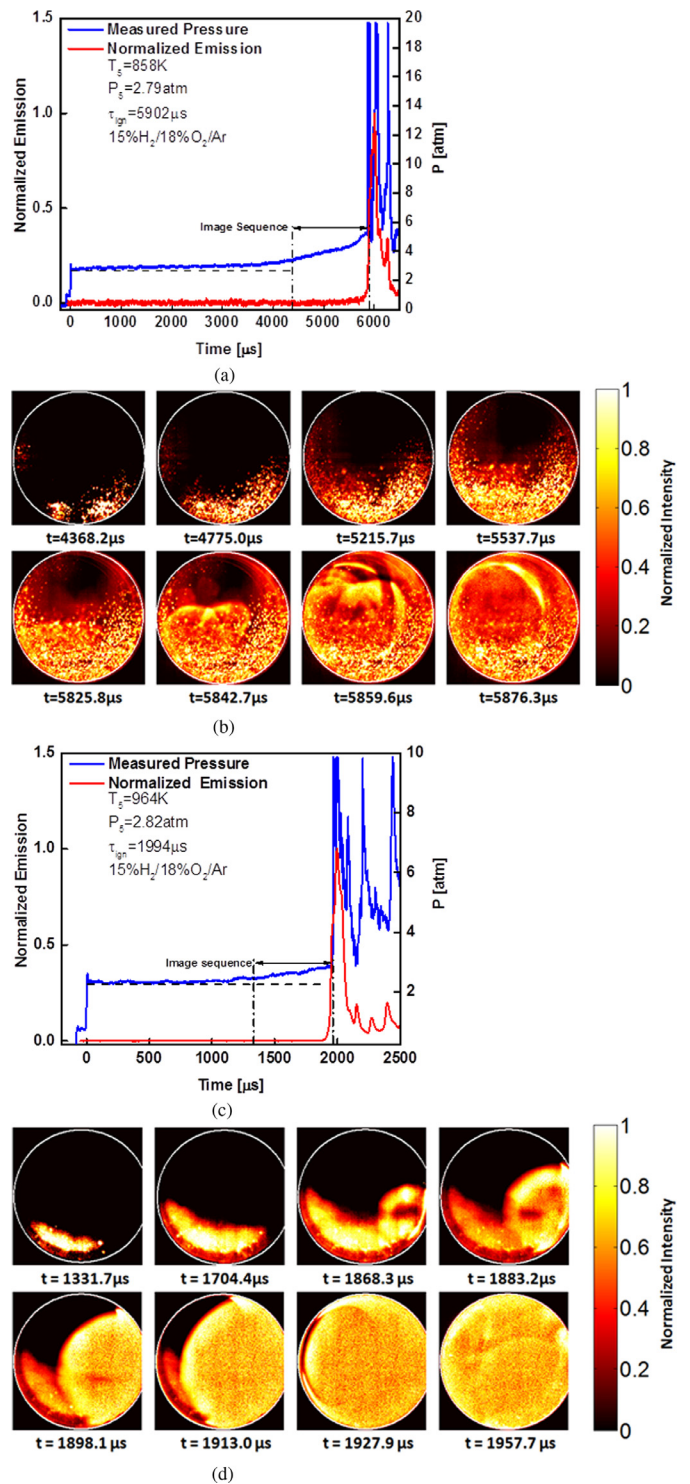


Fig. 2. Typical pressure, emission, and image sequences for 15% $H_2$ /18% $O_2$ /67%Ar in the lower temperature regime ( $<1000$  K). (a) Pressure trace showing mild ignition in a dirty shock tube, (b) corresponding image sequence for dirty tube, (c) pressure trace for run with clean shock tube and (d) corresponding image sequence for clean tube.

The assumption of constant (U, V) is invalid under such conditions. Of the two mixtures that were studied (high concentration – 15%  $H_2$ /18%  $O_2$ /Ar; low concentration – 4%  $H_2$ /2%  $O_2$ /Ar) only significant pressure ramps were observed in the high concentration mixture. In the lower temperature regime ( $T < 1000$  K), ignition delay times for this mixture differed substantially from current models

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