Dynamics of Low-Temperature Autoignition Kernels in Hydrogen-Air Mixtures

S. P. Medvedev, S. V. Khomik, O. G. Maximova, G. L. Agafonov, A. M. Tereza, A. S. Betev
Semenov Institute of Chemical Physics, Russian Academy of Sciences
Moscow, Russia

H. Olivier
RWTH Aachen University
Aachen, Germany

1 Introduction

Autoignition is one of the most intriguing phenomena in gas combustion. Autoignition dynamics is generally expressed in terms of ignition delay τ. Experimental data on ignition delay play a key role in the validation of reaction mechanisms describing combustion processes in internal combustion engines, gas turbines, and other devices that use heat released by fuel oxidation. Since an early work of Voevodsky and Soloukhin [1], it has been known that the classical concept of homogeneous autoignition does not hold for hydrogen and other fuels in the low-temperature range of $T < 1100$–1200 K. Under these conditions, autoignition typically occurs locally, originating in a single or multiple hot spots. This mode of autoignition is more pronounced in the practically important high-pressure range of $P > 0.5$ MPa, as demonstrated by experiments in different setups including shock tubes [1, 2], rapid compression machines [3], and flow reactors [4]. The delay time before the onset of hot-spot ignition of hydrogen mixtures is a parameter of particular importance with regard to engine combustion and nuclear power safety issues. In many practical situations, ignition occurs in a complicated turbulent flow field, and reaction-front propagation from a hot spot is therefore significantly influenced by turbulence. A detailed study of autoignited combustion should use an optical technique. A low-temperature, high-pressure, turbulent environment imposes special requirements on the experimental setup. In the present work, we use a laboratory shock tube operated in an over-tailored mode. This makes it possible to extend observation time and simultaneously create sufficient turbulence due to multiple interactions between the reflected shock wave and the contact surface. As shown in [5], using an over-tailored condition provides ignition delay data that can be compared directly with flow reactor results.

2 Experimental setup

Correspondence to: podwal_ac@yahoo.com

The experimental setup is schematized in Fig.1. The length of high-pressure section *1* can be varied between 2.6 m and 4.2 m. Low-pressure section *2*, separated from the high-pressure section by burst diaphragm *3*, has a cross section of 40 × 40 mm^2 and a length of 2.6 m. Near the tube end, the sidewalls are equipped with two optical windows. The photo in the left bottom of Fig. 1 shows the test section with windows removed. The schlieren/shadow imaging system is equipped with Telemar-7 telephoto lenses spanning a field of view of 140 mm. Images are recorded by a Mikrotron-1362 high-speed digital video camera at a frame rate of up to 20000 fps. Pressure is recorded by Kistler 603B transducers located at distances of 18 mm and 98 mm from the end wall. The onset of ignition is detected by a silicon photodiode. The hydrogen–air mixtures under investigation were prepared in a separate mixer using a partial pressure technique. Argon or nitrogen was used in the driver section to operate the shock tube in an over-tailored mode. The observation time, limited by the arrival of the reflected rarefaction wave, was approximately 15 ms when the shorter (2.6 m) driver section was used. Temperature at autoignition varied between 700 and 850 K and pressure reached 2–3 MPa.

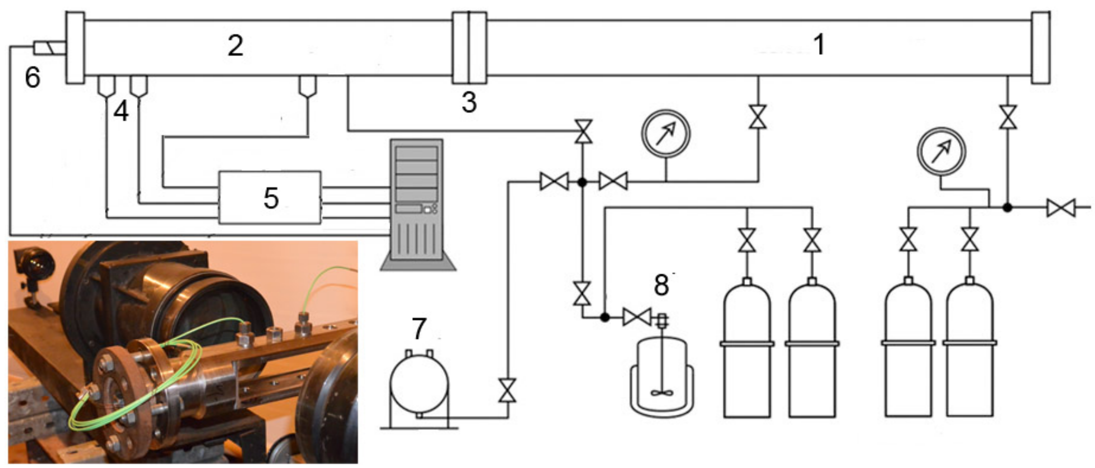

Figure 1. Experimental setup: *1* – driver section; *2* – test section; *3* – burst diaphragm; *4* – pressure transducers; *5* – charge amplifiers; *6* – photodiode; *7* – vacuum pump; *8* – mixer.

3 Results and Discussion

The results of an experiment consist of pressure traces and a series of shadow images. Consider Fig. 2 as an example of data obtained under over-tailored conditions. The pressure trace in the upper panel illustrates multiple shock interactions during an autoignition event. The stages of the process are enumerated in the figure caption. The incident shock wave reflects from the end wall, and the reflected shock propagates towards the oncoming contact surface. Interaction between the reflected shock and the contact surface gives rise to a pressure wave propagating back towards the end flange. Repeated interactions of this kind lead to further increase in pressure and temperature in the end-wall region of the test section. The contact surface spreads out and pressure increases gradually (during several milliseconds) in a similar manner as in rapid compression machines. An additional pressure rise occurs after autoignition. Finally, a rarefaction wave arrives and pressure drops. Following [5], the temperature history at the stage of pressure rise behind the reflected shock can be calculated by the

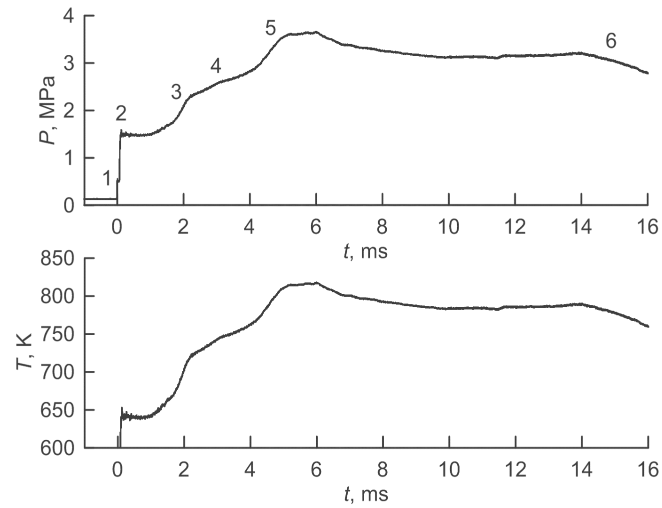

Figure 2. Pressure and temperature histories, 15%H_2 + 85%Air, P_0 = 0.13 MPa: *1* – incident shock wave; *2* – reflected shock wave; *3* – multiple shock–contact surface interaction; *4* – autoignition; *5* – pressure rise due to autoignition; *6* – rarefaction wave arrival.

isentropic relation $T(t) = T_R \cdot \left(P(t)/P_R\right)^{\frac{\gamma-1}{\gamma}}$, where T_R and P_R are temperature and pressure behind the reflected shock wave, and γ is the mean ratio of specific heats. The appearance time and location of a nascent ignition kernel are determined from images such as those presented in Fig. 3.

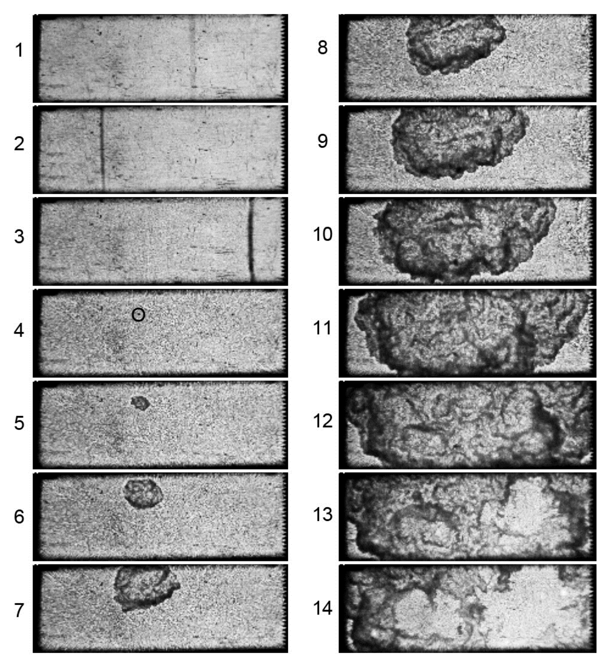

Figure 3. Shadow images for 15%H_2 + 85%Air. Autoignition occurs at P = 2.5 MPa, T = 760 K. Time between frames 1–3 and 4–14 is 0.188 ms. Exposure time = 2 μs. End wall is on the left.

The incident shock in frame *1* propagates from right to left towards the end flange. Frames *2* and 3 in Fig. 3 illustrate the propagation of the reflected shock. Ignition starts at frame *4*, where a nascent kernel is enclosed in a circle. Initially, the ignition kernel looks like a flame ball. Comparing the series of frames with the pressure and temperature histories, we find that the pressure at autoignition is 2.5 MPa and the corresponding temperature is 760 K. An analysis of image sequences shows that the location of the ignition kernel varies randomly from test to test within a range of about 100 mm in a region adjacent to the end flange.

The data obtained from the shock tube experiments are consistent with those reported in [5], where the tube was of 54 × 54 mm^2 cross section and twice as long. The use of a longer tube benefits the study of autoignition of very lean mixtures.

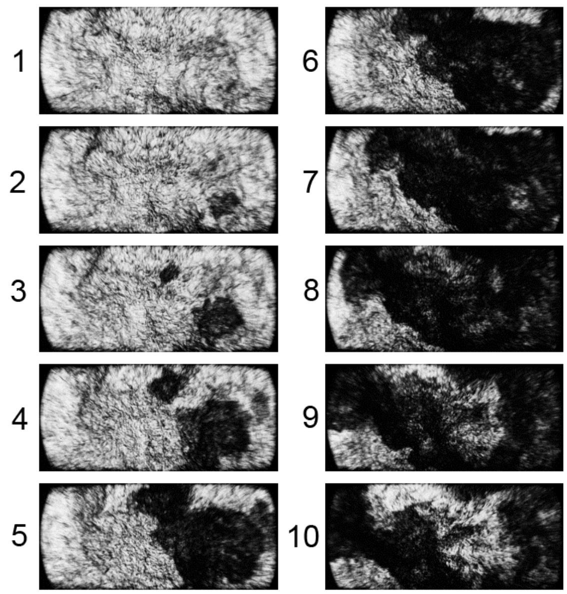

Figure 4. Shadow images for 6%H_2 + 94%Air. Autoignition occurs at P = 1.0 MPa, T = 860 K. Time between frames = 1 ms, exposure time = 64 µs. End wall is on the right.

Figure 4 shows a sequence of shadow images obtained for a 6% H_2 in air mixture. In this case, the autoignition kernel grows much more slowly than in the 15%H_2 + 85%Air mixture. At least three autoignition kernels can be identified. A common feature of both experimental setups is that multiple pressure wave interactions create a relatively high turbulence level in the autoigniting mixture. In particular, a comparison between Figs. 3 and 4 demonstrates a transition from wrinkled flamelet to corrugated flamelet combustion. Interaction between a turbulent flow field and an autoignition kernel is an important problem that requires special consideration.

Schlieren/shadow imaging made it possible to evaluate root-mean-square turbulent velocity via schlieren (shadow) image velocimetry (SIV) technique [6]. Images were processed by using PIVlab software [7]. Figure 5 shows the result of applying the SIV technique to a selection of images presented in Fig. 3. We used built-in functions to determine the rms turbulent velocity components u'_x and u'_y in the plane of view as the rms deviations from the in-plane mean velocity. Assuming that turbulence is transversely isotropic in cross-sectional planes of the tube ($u'_z = u'_y$), we calculated rms turbulent velocity as $u' = \sqrt{\frac{1}{3}(u_x'^2 + 2u_y'^2)}$.

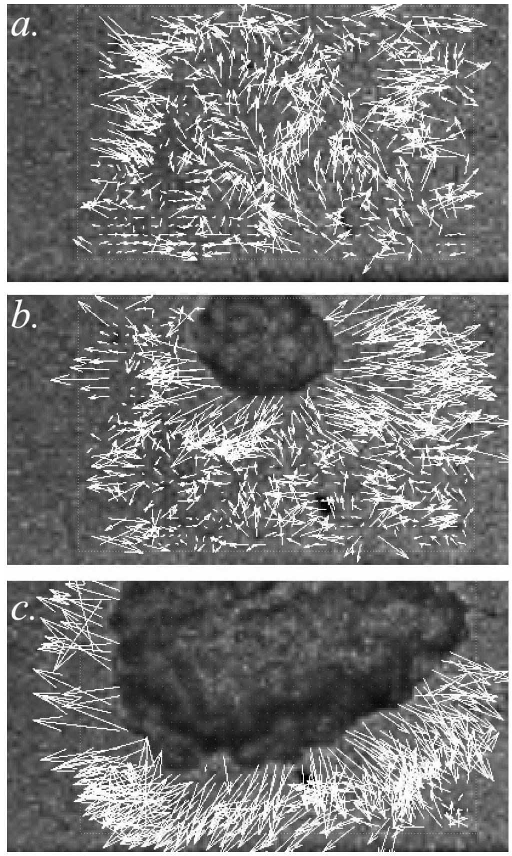

Figure 5. SIV results, 15%H_2+85%Air, *P*= 2.5 MPa, T=760 K. Frames are taken from Fig. 3: (a) frame *4*, u' = 14 m/s; (b) frame *6*, u' = 16 m/s; (c) frame *8*, u' = 29 m/s.

It is clear from Fig. 5 that the autoignition kernel does not affect the turbulent flow field at an early stage. The growth and acceleration of the kernel generates an intense flow, and the rms velocity u' increases twofold (from u' = 14 m/s to u' = 29 m/s). The fact that single kernels are typically observed in relatively sensitive mixtures can be explained by the impossibility of autoignition in stronger turbulence. In less sensitive (leaner) mixtures, the first autoignition kernel almost does not affect the adjacent flow field, and subsequent autoignition events become possible at different locations as illustrated by Fig. 4.

In summary, experiments performed in a shock tube operated in an over-tailored mode reveal characteristic features of low-temperature/high-pressure autoignition of turbulent hydrogen–air mixtures. It is shown that the formation and growth of an autoignition kernel (hot spot) can increase turbulence intensity in the unburned region. Further experimental work should be directed at determining local conditions relevant to autoignition in a turbulent environment.

Acknowledgments

This work was performed within the framework of the Program of Fundamental Research of the Russian Academy of Sciences for 2013-2020 on the research issue of the ICP RAS No. 49.23. State registration number of the Center of Information Technologies and Systems for Executive Power Authorities: AAAA-A18-118031590088-8.

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