

# **Auto-ignition of microparticles as mechanism for RCM combustion**

Vladimir V. Leschevich, Oleg. G. Penayzkov

Physical and Chemical Hydrodynamics Laboratory, Heat and Mass Transfer Institute 220072,  
Minsk, Belarus

## **Introduction**

The reaction kinetics plays a significant role in the ignition characteristics of fuel/air mixtures that control performance of many advanced combustion devices. Nonetheless, experimental ignition delay data for synthesis gas and hydrogen under practical conditions ( $T < 1000$  K,  $10 < P < 30$  atm) show considerable disagreement with what is predicted by state-of-the-art chemical kinetics models. The uncertainties in the rates of elementary reactions are not entirely explaining the model's failure. [1] It is now considered that chemical induction processes are strongly influenced by various experimental perturbations [2] which involves the correct experimental data interpretation. Numerous investigations of hydrogen mild ignition have shown that the reaction waves originate sometime earlier than chemical induction time from spatially distributed exothermic initiating centers, which can origin as a result of temperature fluctuations, fluctuations of activated molecules or radical concentrations, pressure waves, or catalytic generation on suspended particles [3-6]. In the rapid compression machine, the non-uniform ignition is usually attributed to the existence of temperature gradients generated by the compression process. However, the evidence that the effect of compression process is not important was received in the paper [7]. Authors stated that the presence of particles in the reactive gas mixture to be the major cause of non-uniform ignition process. This phenomenon was not investigated in further studies, although many researchers discussed it and observed the presence of particles in reaction chamber at visualization of mild ignition [8]. In order to get deeper understanding of mild hydrogen auto-ignition phenomenon and to observe the possibility of microparticles to force combustion the present study was conducted by means of rapid compression machine (RCM) using high speed visualization.

## **2 Auto-ignition of hydrogen/air mixture**

Experiments were carried out for stoichiometric hydrogen-air mixtures at temperatures 900 - 1000 K and pressures 0.8 - 1.8 MPa. Ignition times were measured by observations of OH radicals emission through quartz window mounted in the cylindrical wall of test chamber and connected with photomultiplier by optical fiber guide. Two narrowband interferometric filters

were installed in front of the photomultiplier photocathode to pass only emissions at  $\lambda = 308.9$  nm (transitions  $A^2\Sigma - X^2\Pi$ ). Ignition time was also defined by pressure measurements as the time difference between pressure peak at the end of compression stroke and the onset of pressure rise (5 % of maximal rise) caused by reactive mixture ignition. Fig. 1 shows typical pressure transducer (solid lines) and photomultiplier signals (dashed lines) recorded by digital oscilloscope during several runs. These experiments were performed at the same compression ratio and initial conditions which marked on the plot. The mixture temperature at the end of compression ( $T_c$ ) was calculated from measured pressure history using assumption of the adiabatic process and temperature depended specific heat ratio.

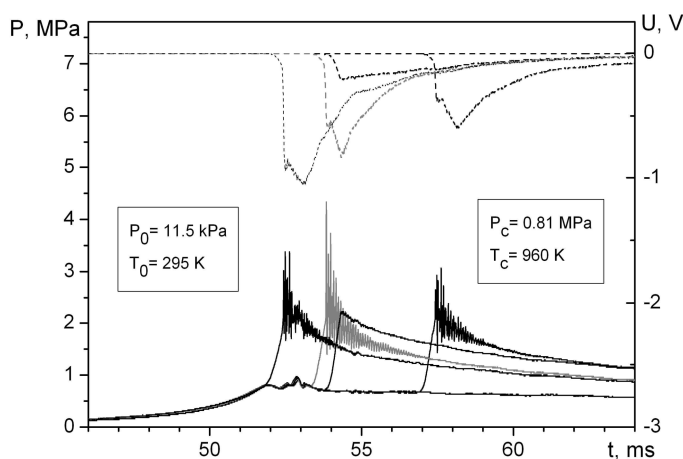


Figure 1. Pressure (solid lines) and OH emission (dashed lines) registrations

It was found that ignition delay time could be rather different. Additionally knocking phenomena were sometimes developed in the burning mixture. However, the main question is the existence of experiments when auto-ignition was not observed or appeared before the end of compression (anomalous behavior). We suggested that such behavior of reactive mixture at these conditions can be connected with non-uniform ignition, started before the end of chemical induction time at the spatially distributed initiating centers. As discussed previously the origination of these centers can be caused by microparticles suspended in reactive mixture. In order to clean reactive mixture the 80 nm filter was installed between the storage vessel and the combustion chamber. This increased the amount of experimental runs during which auto-ignitions were not observed. Nevertheless, the ignition delay time measurements were not repeatable and significant different from zero-dimensional homogeneous chemical kinetic calculations. As there are no experimental data in literature obtained by RCM at these conditions, we performed several runs at higher pressures (about 3 MPa) and compared our measurements with others [9-11]. The rather good agreement was found in general, but in rare experiments, auto-ignition was also started too early and even before the end of compression. This comparison proved applicability of our experimental setup for measurements of ignition delay time with the assumption of a correct data interpretation. Nevertheless, this did not give understanding of anomalous experiments. To investigate the possible effect of particles presented in reactive mixture on induction time high-speed digital imaging was used. Auto-ignition visualization was made by intensified CCD camera system "PCO DiCAM-PRO" in double trigger mode, which allows obtaining two frames with minimal exposure time and interval between two exposures equal to 500 ns. Camera was installed in front of compression cylinder with quartz window equal to its diameter (50 mm). The recording time was synchronized with ignition process by signal from photomultiplier.

The example of two serial images exposed through neutral optical filter at the onset of hydrogen auto-ignition is presented on Fig.2. The initial pressure in this experiment is 22 kPa and temperature is 290 K. The postcompression conditions (compression ratio 24.5:1) have to be about 1.68 MPa and 960 K, but the ignition in this run was started a little before or even at the end of compression event. The first frame was exposed during 300  $\mu\text{s}$  from the moment of light appearance in the test volume, the second during 100  $\mu\text{s}$  with 500 ns delay after the first. Due to long exposure time the narrow bright tracks that can be left by moving burning particles were registered on the background of the propagating reaction front. It is difficult to make an assumption from these images about the location and reason of the earlier auto-ignition. Therefore, this experiment was repeated under other camera settings. The frames were exposed during 50  $\mu\text{s}$  through interferometric filter (650 nm) with delay time between frames 100  $\mu\text{s}$  (Fig. 3).

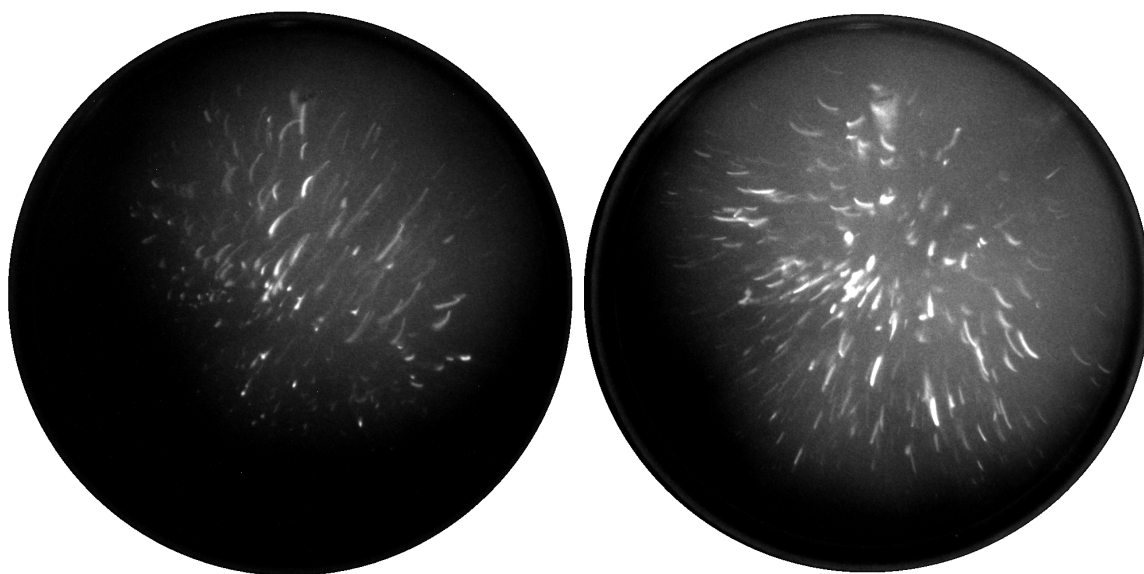


Figure 2. Imaging sequence of hydrogen/air mixture auto-ignition.

Auto-ignition started before the end of the compression stroke at pressure about 1.4 MPa and mixture temperature about 930 K. An evolution of the combustion from a single initial center was registered on images. The visible velocity of combustion expansion is about 40 m/s. The origination of this ignition center may be caused by a catalytic effect on the surface of a foreign particle or by its auto-ignition in heated gaseous atmosphere. To check this hypothesis and to establish the ability of small unknown particles spontaneously ignite under such conditions, several experiments were conducted with non-reactive gas mixtures.

The mixture of 50%  $\text{O}_2$  and 50 %  $\text{N}_2$  was used in first experiment. It was filled to compression cylinder without filtration, in order to increase a quantity of extraneous particles in combustion chamber. The exposition of the first frame was started together with compression stroke and proceed during 75 ms. The second frame was exposed during 75 ms with 500 ns delay after the first. The maximum pressure (about 1.76 MPa) was achieved in the test chamber in 77 ms. The single short bright track is seen on the first frame (during compression stroke), but on the second image there are many tracks like in experiments with hydrogen/air mixture (Fig. 4). The photomultiplier recorded a high intensity light in the test

volume just after compression. The delay time of the appearance of this light, not significantly differ from ignition delay times measured for hydrogen/air mixture (about 1 ms).

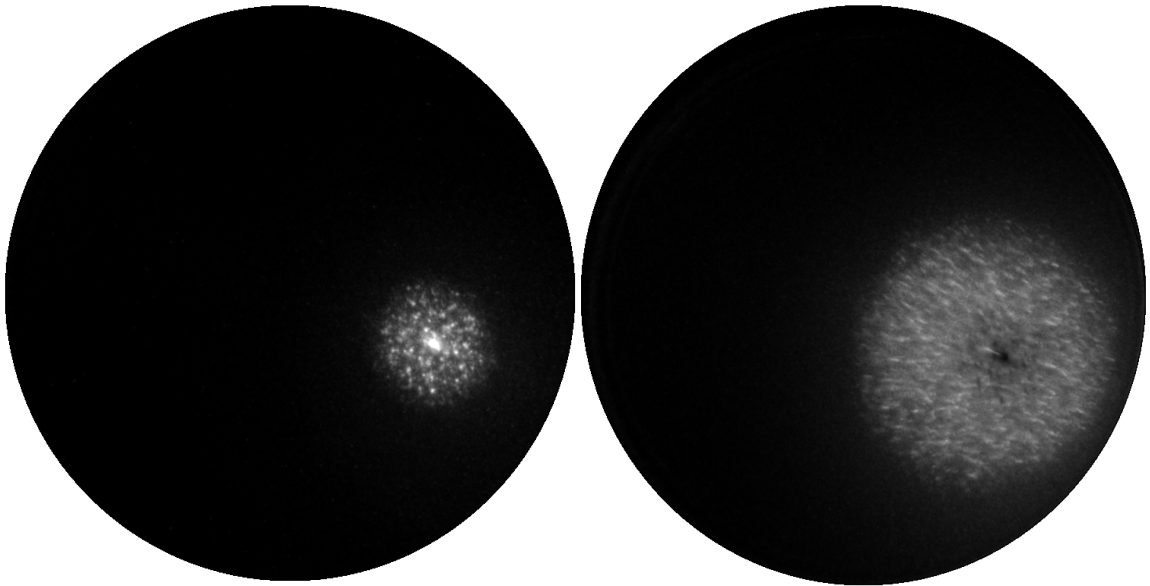


Figure 3. Imaging sequence of hydrogen/air mixture auto-ignition

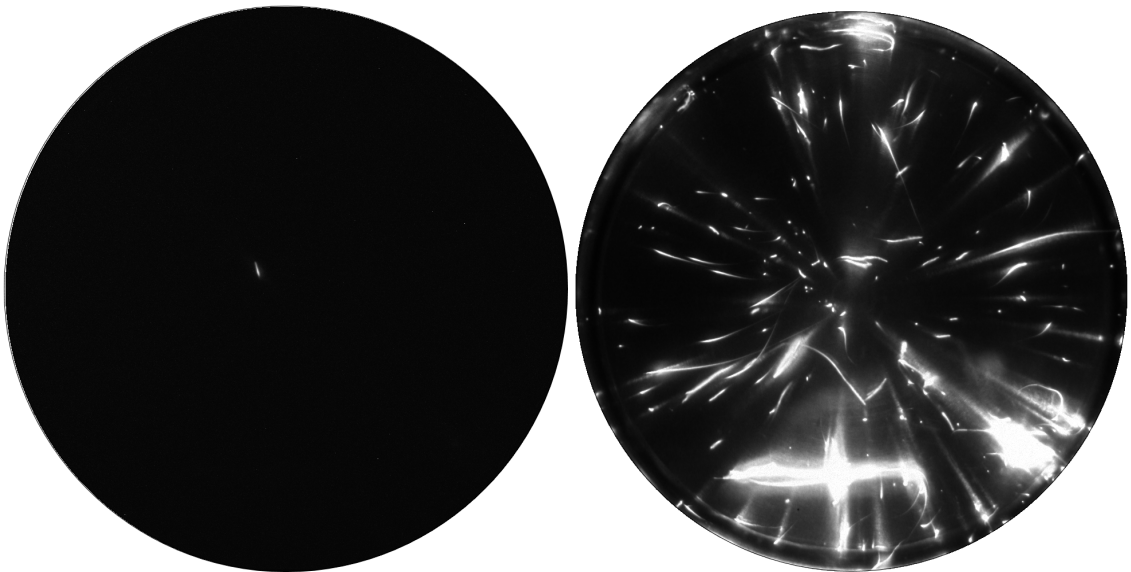


Figure 4. Imaging sequence recorded in experiment with mixture of 50% O<sub>2</sub> and 50 % N<sub>2</sub>.

The same experimental run was repeated for oxygen/nitrogen mixture filled to compression cylinder through 80 nm filter and no light was registered. It proves that tracks, registered in previous experiments, were produced by particles, which penetrated to the test volume together with gaseous mixture and were ignited in heated atmosphere of strong oxidizer. Nevertheless, filtration of reactive mixture is not enough procedure, which can provide “clean” experimental measurements in rapid compression machine. We performed experimental runs with carefully cleaned air (synthetic mixture of oxygen and nitrogen) and also registered many bright spots on frames exposed at the maximal CCD camera sensitivity

(Fig. 5). As in previous cases, the first light appeared before the end of compression stroke, but the maximal photomultiplier signal was registered in about 1 ms after compression.

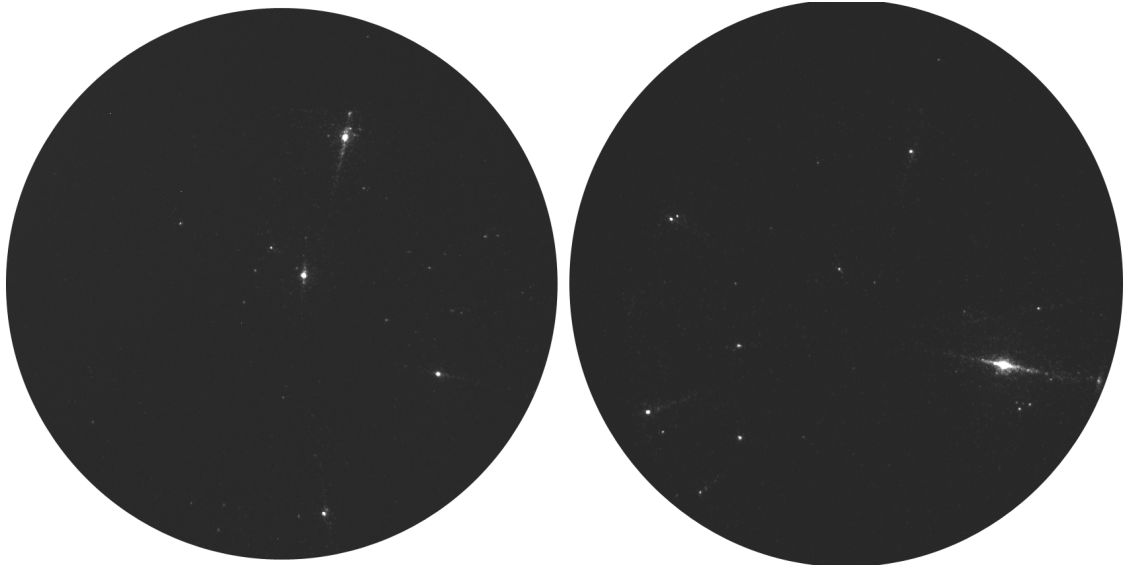


Figure 5. Imaging sequence recorded in experiment with filtrated air. Left: frame was exposed from the moment of light appearance in the test volume during 50  $\mu$ s. Right: frame was exposed with 1 ms delay after first during 50  $\mu$ s.

## Conclusion

The ignition of ultrafine particles can explain a big scattering of experimental data and discrepancy of measurements with results of zero-dimensional homogeneous kinetic calculations at temperatures lower than 1000 K. In our experiments, the absence of these particles explained the experiments, in which auto-ignition of hydrogen/air mixture was not observed. Nevertheless, obtained information is not enough to conclude that the non-uniform early hydrogen auto-ignition is caused only by presence of microparticles. As the suspended particles were not completely excluded from our experiments we can only conclude that any experimental data on ignition delay times are need in a careful analysis before application for homogenous chemical kinetic models validation and updating.

## References

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