Laser based study of auto-ignition of sprays in a continuous flow reactor

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Abstract
Auto-ignition phenomena with relevance to lean premixed prevaporised (LPP) combustion have been investigated with laser-optical methods. A generic mixing reactor with optical access was set up, offering the possibility to observe the reaction within a droplet laden hot air flow (up to 1100 K) at elevated pressure levels (0.8 MPa). The measurements aim at a detailed understanding of the local conditions under which auto-ignition and combustion occur in the mixing duct. The onset of chemical reactions in the fuel-air mixture was imaged by planar laser-induced fluorescence (LIF) of formaldehyde (CH₂O). Formaldehyde is formed during reaction progress leading to auto-ignition and consecutively consumed during combustion. Simultaneous imaging of droplet Mie scattering was applied to study the correlation of their location with the ignition. Furthermore, the chemiluminescence signal of the flame was analysed, using a high-speed ICCD camera system, in order to track the propagation of the individual ignition spots.

Introduction
The reduction of peak combustion temperatures by means of lean combustion is the most promising concept for reducing the NOₓ emissions of gas turbine combustors. Presently, the lean premixed prevaporised (LPP) combustion is not applicable in aero engine gas turbines due to unresolved challenges like flame instability, flash back and auto-ignition. In contrast to industrial gas turbines, the transient conditions and also the use of liquid fuel still inhibit the successful and safe implementation of this technology. The study of auto-ignition of liquid fuel sprays is the topic of the present paper.

Knowledge of the ignition delay time and its dependence on global conditions (pressure p, temperature T, equivalence ratio Φ) is indispensable for combustor design. In this context, the auto-ignition delay times of homogeneous fuel/air mixtures as well as of partially-premixed ones and of single droplets [1] have been studied extensively in devices like shock-tubes [2, 3], rapid compression machines [4, 5] and also some in continuous flow reactors [6, 7, 8]. Compared to the auto-ignition of gas mixtures, spray auto-ignition is much more complicated, since in a droplet laden turbulent flow, in general inhomogeneous distributions of temperature as well as of species concentrations are present. Additionally, the reaction paths and kinetics underlie a decisive temperature and pressure dependence in the technically relevant temperature regime between 800 K and 1100 K [9]. As a consequence, a pronounced negative temperature coefficient (NTC) region of the auto-ignition reaction delay as known for many homogeneous gas mixtures is no longer observed. For inhomogeneous compositions like single droplets and fuel sprays, ignition starts at distinct locations, where optimal conditions with respect to temperature and fuel concentration prevail [10]. Altogether, these effects lead to a complex behaviour being up to now not accessible for accurate modelling.

The measurements in this study are focussed on a detailed understanding of the local conditions that lead to auto-ignition and on the characteristics of ignition including the subsequent flame propagation in presence of residual droplets in a continuous turbulent flow reactor at elevated pressure.

The experimental setup was designed to avoid flame stabilisation in the mixing duct. Hence, the reaction which can be observed through the optical access can be attributed to continuous auto-ignition. For high temporal and spatial resolution, a high-speed ICCD system was used to observe the onset and progress of auto-ignition by imaging the chemiluminescence either over broadband spectral range or spectrally filtered for specific species. Furthermore, LIF of formaldehyde (CH₂O) was used to monitor the onset of ignition [11] in a laser-light-sheet. Simultaneously, the Mie scattering signal of residual droplets was recorded to investigate the spatial correlation between droplets and ignition onset.

Experimental set-up
The test rig was designed for investigation of auto-ignition of fuel sprays in a continuous flow reactor. It is integrated into the high pressure, high temperature facility of the Institute für Thermische Strömungsmaschinen (ITS), which provides a non-vitiated air at mass flow of up to \( m = 1.4 \text{ kg/s} \) at a maximum temperature of \( T = 1123 \text{ K} \) and pressure of \( p = 1 \text{ MPa} \).

Figure 1 shows a detailed sketch of the test rig. It mainly consists of three components: fuel atomiser, flow reactor and plenum section. After mass flow

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measurement ($m_{\text{air}}$) by a venturi tube (not shown), the hot air enters from the left and passes the atomiser section. Here the temperature $T_{\text{air}}$ of the hot air is measured. The fuel is injected coaxially via a constant pressure spill-return atomiser. The atomising pressure was set below 3.5 MPa to increase the number of residual droplets in the plenum section. Due to the short residence time of the flow within the reactor, the conductive heat losses are negligibly small compared to the enthalpy of the flow.

After entering the optically accessible plenum where the auto-ignition zone is located, the hot air/fuel-mixture is shielded by a coaxial flow of cold air, which serves to flush the corners of the plenum as well as to inhibit flame stabilisation in the boundary layer. The velocity of the cold air is always kept lower than the mean velocity of the reacting flow which results in a fast dilution of the fuel/air mixture due to high shear stresses between the hot and cold flow. In order to ensure that a reaction is inherent from continuous ignition and not caused by flame propagation, the flow velocities were kept significantly above that of the turbulent flame speed. Optical access to the plenum section is provided via four fused silica windows (130x60mm²) mounted perpendicularly to each other. A water cooled bluff-body detaches the plenum from the afterburning section and ensures downstream mixing and afterburning.

Jet-A1 kerosene and heptane were used as fuels in this investigation. The variation of the auto-ignition delay time with temperature and pressure is basically compensated by adjusting the flow velocity ($35 \text{ m/s} < \overline{v}_\text{air} < 120 \text{ m/s}$) in the rig accordingly. All measurements presented in this paper were accomplished at an absolute plenum pressure of 0.8 MPa. The overall equivalence ratio was $\Phi = 0.55-0.65$, resulting in a maximum thermal load of 0.6 MW for kerosene at maximum air temperature.

Results and Discussion

The discussion of the results is subdivided into two parts, referring to the different measurement techniques applied. First, we will focus on high-speed sequences of the chemiluminescence emitted from the reaction zone.

Chemiluminescence imaging

The screenshot of a video camera in figure 2 gives an impression of the continuous auto-ignition reaction. Due to the long exposure time of 15 ms, spots of high intensity appear as streaks. At higher injection pressure these yellow streaks would disappear and the auto-ignition zone shows up as a blue flame.

In figure 3, exemplary sequences of high speed chemiluminescence imaging are shown. Apparently, the auto-ignition originates from individual ignition events. Consecutively, the appearing flame kernels are growing and expanding into the unburned mixture, sometimes coalescing with each other. At the same time, the flame kernels are convected downstream and a “reaction-cone” is formed (see figure 2). Additionally, it is also observable that very intense spots appear within the spreading flame kernels. The number of spots increases when the atomising pressure is decreased. However, if heptane is used as fuel, almost no spots and a blue flame are visible, even for low atomisation pressures. This finding must be attributed to residual droplets, since the boiling point of heptane is much lower than that of kerosene components. In fact, applying the Mie technique when operating with heptane, signals from residual droplets could hardly be found.

In a further analysis, the chemiluminescence was separated into contributions from different species by performing simultaneous shots with a multiple ICCD-camera system (HSFC-Pro (pco) and DynaMight).
(LaVision) [with UV objective]). By using band pass filters, the raw signal was split into fractions originating from \( \text{OH}^* (308 \text{ nm}) \), \( \text{CH}^* (430 \text{ nm}) \), and by a 600 nm long pass filter the luminosity originating from soot precursors was recorded [4].

This results from residual droplets which burn quite rich within the expanding flame zones.

The mode of auto-ignition, however, could not be determined from 3D-imaging. The mode describes whether auto-ignition is initiated around residual droplets or even groups of droplets or if the onset is somewhere in the gas phase with no direct relation to the coexisting droplets.

As mentioned in the introduction, the complex interaction of temperature and mixture fraction in the droplets’ boundary layers with ignition chemistry determines the time and location at which ignition starts. According to Tanabe et al. [1], for heptane this should lead to ignition closer to droplets at a certain (medium) range of initial temperatures, as the enthalpy of evaporation reduces the temperature and thereby allows for a shorter ignition delay. Thus, under these conditions, the “most reactive zones” are located in the fuel richer region closer to a droplet.

Figure 3: Chemiluminescence high-speed sequences (33x33mm²; Jet-A1; \( p_{\text{fuel}} = 2.5 \text{ MPa}, \Delta t = 100\mu\text{s} \))

In figure 4, two sample shots are shown as an example for many other similar shots. Obviously, there is a good correlation of the signals originating from \( \text{OH}^* \) and \( \text{CH}^* \). These excited radicals are formed in zones of exothermic reactions indicating the heat release of the hot flame [12]. In contrast, the small spots of high intensity within flame zones can be attributed to broadband radiation between 600 nm and 900 nm, which is an indication of young soot [4]. The soot radiation originates from residual droplets that are burned under fuel-rich conditions in ambiance of the hot reaction products, as the previous reactions in the flame front of the expanding flame kernels consumed most of the oxygen already.

The appearance of random but individual ignition spots within the turbulent flow field can be observed over the investigated temperature range. With reduced atomisation pressure and thereby worse spray quality, the luminosity of the flame zones is shifted to the red. The colour/intensity scales of the different chemiluminescence images are not comparable, because neither the amplification of the different cameras nor the effect of the filters can be quantified. Comparisons can therefore only be made within one picture or qualitatively referring to the flame shape.

Figure 4: Chemiluminescence signal - partially filtered for species with specific optical filters (36x36mm²; Jet-A1; \( p_{\text{fuel}} = 2.5 \text{ MPa} \))
Planar LIF and Mie scattering

In the second part of the investigation, images of planar LIF of CH$_2$O and simultaneous Mie scattering of droplets in the continuous flow were captured. The objective of this measurements was to study the interactions of droplets, ignition and flame in detail, with respect to the theoretical implications described above.

Two measurement techniques were combined as follows: The LIF system consists of a XeCl-excimer pumped dye-laser tuned to an absorption line of CH$_2$O at 339.3 nm and a 16-bit ICCD camera (DynaMight (LaVision)). Mie scattering from the UV-laser was blocked by an UV-filter. Mie scattering is excited by a Nd:YVO$_4$ laser at 532 nm, filtered by a band pass filter and captured by one of the cameras of the 4x-ICCD system (HSFC-Pro; pco). Both cameras were directed to opposite sides of the plenum and aligned in order to display the same light-sheet section. Formaldehyde was chosen as a candidate for the LIF technique because it is well suited as indicator for the auto-ignition / cool-flame progress [13]. A high amount of CH$_2$O is generated during the first reactions leading to ignition. In a two stage ignition process, CH$_3$O$^*$ chemiluminescence also contributes to the cool flame luminosity [4, 12]. Eventually, CH$_3$O is consumed in the final stage of the ignition.

Figure 5: Simultaneous LIF and Mie scattering with additional image of emitted chemiluminescence (36x36mm$^2$; Jet-A1; $p_{\text{fuel}}$=2.5 MPa)

Presently, this set up was used in a single shot configuration. However, it is planned to extend it to high-speed sequences of LIF/Mie pairs.

Figure 5 shows two examples of the LIF/Mie twin-shots ($\Delta t=1 \mu$s), with an additional picture of the emitted chemiluminescence. The width of the laser light sheets was only 18 mm. Although being not corrected for sheet inhomogeneity, the LIF-signal of CH$_2$O displays pronounced areas of higher and lower fluorescence intensities which can be attributed to different levels of reaction progress at different locations. For interpretation, however, one has to keep in mind that intensity correlates with reaction progress as well as with inherent reactant. This means, that the LIF-signal is influenced by concentration of formaldehyde which in turn depends on the reaction progress and the local equivalence ratio. Additionally, it is also quenched by spectroscopical influences. Nevertheless, zones of complete CH$_2$O consumption due to expanding flame structures can be distinguished by sharp gradients in comparison to the smooth transitions between zones of more and less CH$_2$O-LIF intensity. The images should therefore serve rather to localise the boundaries of the hot flame zones than to show a quantitative distribution of the CH$_2$O.

The match of flame structures of the LIF shot to the emission of chemiluminescence is quite obvious, especially in the right picture series, where the flame structures cover a major part of the light sheet plane and are also visible on intensified LIF image despite the very short exposure time. In addition, in this case of large flame dispersion, no more residual droplets can be detected from the Mie signal in contrast to the left series.

Figure 6: Simultaneous LIF and Mie scattering (14.7x18.2mm$^2$; Jet-A1; $p_{\text{fuel}}$=3 MPa)
The total measurement series comprises several hundred pairs of LIF/Mie images over the whole temperature range. Some more examples are shown in figure 6 and figure 7, representing the effect of temperature and of different fuels. The two pairs in figure 6 are acquired with Jet-A1, the ones in figure 7 with heptane. The major difference between Jet-A1 and heptane is the reduced number of droplets (highlighted by red circles) and also the lower signal level of the LIF image for heptane. This suggests that the LIF signal is enhanced by the aromatics contained in the Jet-A1. Nevertheless, the general image structure is quite similar for both cases.

Concerning the Mie scattering of the droplets, there is obviously no coherence with the progress of reaction or even the start of hot ignition. Droplets exist in areas of relatively high CH\textsubscript{2}O concentrations as well as in regions of low concentrations. For kerosene, fluorescent droplets can also be seen in the LIF images. The bright small spots which are present in regions where the CH\textsubscript{2}O is consumed (see figure 5) must be attributed to soot as discussed previously.

Figure 7: Simultaneous LIF and Mie scattering (14.7x18.2mm\textsuperscript{2}; heptane; \(p_{\text{fuel}}=3\) MPa)

Summarising the observations in this investigation, one can assert that the location of auto-ignition is not directly affected by the existence and position of any residual droplets. Bigger residual droplets are not able to follow the turbulent structures in the same manner as smaller ones. Furthermore, fuel vapour and radicals formed during the reaction progress are convected within the turbulent flow and interact also with the position of other residual droplets. Finally, the location with optimal conditions for fast auto-ignition is no longer directly influenced by the temperature and fuel boundary layer of a droplet, like postulated for a single droplet by Tanabe et al. [1]. The observed uncorrelated positions of droplets and flame structures might rather be explained by turbulent transport phenomena in the continuously auto-igniting droplet laden flow.

Conclusions

Despite the rough environment of the auto-ignition rig, the sensitive optical measurement techniques of laser induced fluorescence and Mie scattering of residual fuel droplets were successfully applied simultaneously. In this way, the course and behaviour of self igniting fuel spray in a continuous hot air flow under elevated pressure were investigated. The fuels used were Jet-A1 kerosene and heptane. Additional high-speed sequences of the chemiluminescence were captured.

The most important findings can be summarised as follows:

- Auto-ignition happens at random locations within the reacting flow and also with temporal fluctuations of the axial position, i.e. of the auto-ignition delay time.
- Spots of auto-ignition result in flame zones which are expanding whilst they are convected downstream.
- Residual droplets are burned quite rich within the propagating flame zones, emitting broadband radiation of high intensity which indicates young soot.
- The LIF images of CH\textsubscript{2}O show smooth transitions of higher and lower intensity indicating a different reaction progress within the flame zones as well as sharp gradients where CH\textsubscript{2}O is already consumed by the propagating flame.
- There is no observable spatial correlation of visible droplets and the progress of the chemical reaction zone within the turbulent flow field.

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References