Plasma Assisted Cool Flames in a Rapid Compression Machine

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Abstract

In order to reduce pollutant emissions in conventional engines, lean mixtures and low temperature conditions can be used. Plasma discharges have attracted more and more attention in the purpose of ignition and flame stabilization. The aim of this work is to study the interaction between the plasma generated species and the low temperature combustion mechanism using n-heptane/air mixtures. ICCD imaging shows that the discharge morphology depends on the pressure and emission spectroscopy experiments show that the emission of the plasma discharge is dominated by N_2 emission whereas the emission from the cool flame is mostly due to formaldehyde emission.

Introduction

For a long time plasma discharges have been considered as an efficient tool to extend the ignition limits and improve to combustion stability. Nanosecond discharges provide low temperature non-equilibrium plasma, that is plasma for which the electron temperature exceeds the gas temperature. Energetic electrons produce ions, radicals and excited species through impact dissociation, excitation, and ionization of the gas molecule. Nanosecond discharges have attracted more and more attention in the purpose of ignition in different experimental configurations: in shock tubes [1], in flat flames [2], in flow reactors [3] and in internal combustion engines [4] .

There is currently a need for experimental Plasma Assisted Ignition (PAI) data in the low temperature, lean mixture and high pressures conditions of new engine technologies. These will help to better understand the interactions between the nanosecond scale plasma chemistry and millisecond scale combustion chemistry associated with engine phenomena.

Most of the recent studies focused on the effect of the plasma at pressures inferior or equal to the atmospheric pressure, with a temperature range from 400 K to 2200 K [2], [3]. These modeling studies showed that the effect of the plasma is associated with the fast development of chain reaction mechanisms due to the production of O and H atoms by electron impact dissociation of molecules in the discharge. These studies also showed that O and H atoms yield higher concentrations of the OH radical in comparison to comparable combustion.

Other studies in flame conditions [5] showed that at low pressures, using non-equilibrium plasma discharges in DME/oxygen/helium mixtures the plasma could activate the low temperature combustion mechanism. CH_2O PLIF experiments showed that a significant amount of CH_2O is formed at low temperatures following the discharge.

Nanosecond dielectric barrier discharges (DBD) are considered as an efficient source of non-equilibrium and low temperature plasma because they offer the advantage of multi-point ignition of combustible mixtures at initial ambient temperatures and atmospheric pressures [6]. In the surface dielectric barrier discharge (SDBD) configuration, one electrode is placed above the dielectric surface while another electrode is placed below the dielectric surface. The advantage of this configuration is that it limits the current and prevents the transition of the discharge to an arc.

Rapid Compression Machines (RCMs) are efficient tools to study ignition phenomena in conditions close to those reached in internal combustion engines. Their operational domain spans temperatures ranging from 600K to 1000K and pressures from one to tens of bar.

RCMs usually provide accurate measurements of ignition delays, but can also be equipped with optical diagnostics, or sampling apparatuses. The corresponding experimental data forms a detailed validation target for reaction mechanisms.

The combination of a Rapid Compression Machine with a Surface Dielectric Barrier Discharge is therefore a good tool to provide experimental data on the kinetics and application of PAI in internal combustion engines. The aim of this work is to study the effect of a nanosecond SDBD discharge on the low temperature reactivity of a stoichiometric n-heptane/O_2/N_2 mixture.

EXPERIMENTAL SETUP

The Plasma Assisted Ignition experiments were carried out in the University of Lille1 RCM, which is has been described before [7]. Only a brief description will be given in this paper. This RCM has a right-angle design, which ensures that the volume is kept strictly constant at the end of compression. At top dead center (TDC), the RCM combustion chamber is a cylinder, with a diameter of a 50 mm and height of 16.16 mm. The stroke length is 200 mm. The combustion chamber is equipped with two side optical accesses with a diameter of 14 mm. Temperatures in the range 600-1000 K and pressures from 1 to 16 bar were obtained by compressing gas mixtures in the combustion chamber.

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A creviced piston head is used to ensure maximum homogeneity of the temperature field at the end of the compression [8] following the design suggested in [9].

The nanosecond SDBD was generated by a electrode system described in [10]. It is composed of a coaxial high voltage electrode connected to a copper disk, 1 mm in thickness and 20 mm in diameter. The inner diameter of the low voltage aluminum electrode is equal to the outer diameter of the high voltage electrode. The outer diameter of the low voltage electrode is equal to 46 mm. A dielectric PVC layer is placed between the two electrodes to avoid short circuit. Figure 1 presents a scheme of the electrode system and its coupling with the RCM combustion chamber.

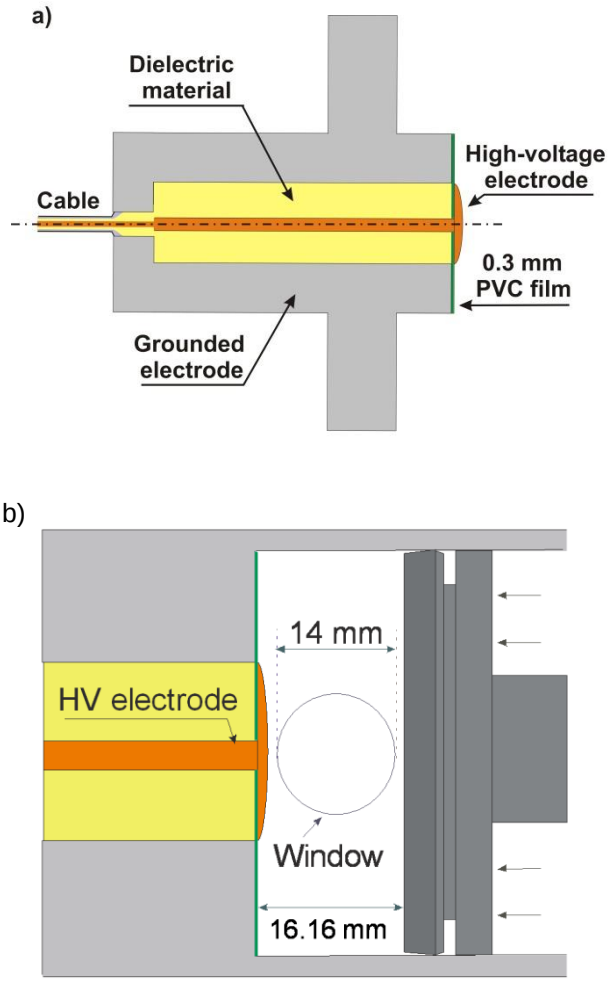

Figure 1: a) Scheme of the SDBD electrode; b) Side view of the RCM combustion chamber with the SDBD electrode mounted in place of the end plate.

The electrode system was installed in place of the end plate of the Rapid Compression Machine. The electrode system is connected to a High Voltage (HV) pulse generator through a 30 m long cable with a impendence of 50 Ω (RG 213U standard). This cable is equipped with two back-current shunts (BCSs). BCSs are electrical probes made of 14 resistors and provide information on the discharge timing as well as the corresponding oscillograms. The first BCS is located at the middle of the cable, while the second one is separated from the HV generator by a distance of 1 m. A FID Technology, FPG2003NM high voltage (HV) pulse generator was used in the experiments. It provides a ±(12-30) kV in the cable pulse amplitude, with a 20 ns pulse duration and 0.5 ns front rise time. A LeCroy WaveRunner 600 MHz oscilloscope was used to record the voltage oscillograms from the BCSs.

The discharge was initiated a few milliseconds after the end of the compression (maximum 3 ms) with help of the synchronization scheme displayed in Figure 2. A TTL signal from the RCM acquisition/command system was used to initiate the compression. The departure signal from the piston motion sensor was used to trigger a Stanford Research DG 535 delay generator set to the compression time, which in turn triggered the HV generator. As the high-voltage propagated through the cable and BCS2, it was transformed to a TTL pulse by an additional generator with an internal delay inferior to 10 µs. This allowed recording the timing of discharge initiation back in the RCM acquisition system.

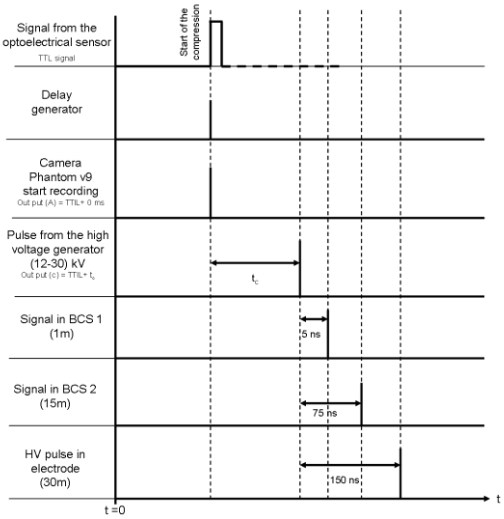

Figure 2: Descriptive chronogram of the synchronization scheme of the experimental setup.

Emission spectroscopy experiments were performed using a Acton Research Corporation SpectraPro 300i spectrometer connected to a Princeton instruments PIMAX 3 camera. The signal from the combustion chamber was reflected by an aluminum broadband mirror and collimated by two convergent lenses with a 200 mm focal length, as described in Figure 3. The ICCD camera is triggered by the delay generator in order to collect the emission spectrum at a specified time during the ignition delay. 150 line/millimeter and 1200 line/millimeter optical gratings were used for these experiments.

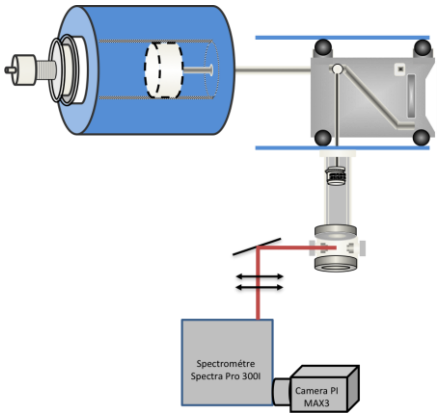

Figure 3: Scheme of the ULille RCM and the used emission spectroscopy facilities.

Emission spectra were recorded between 250 nm and 550 nm in order to observe the emission of CH_2O, OH, and N_2.

Results and Discussions

In a previous study [10], detailed experiments were performed in stoichiometric *n*-butane/O_2/inert, and stoichiometric and lean Methane/O_2/inert mixtures. The pressure and temperature range covered by this study was 600 K – 1000 K and 6 bar - 16 bar. Inert gases used were N_2 and Ar.

This study showed a well-pronounced voltage threshold between two separate regions. Below the threshold voltage the ignition delay is equal to the autoignition delay, and above this threshold the ignition delay measured from the pressure profile in the combustion chamber is 1 to 2 orders of magnitude lower. A tentative explanation provided in [11] relied on the transition from a quasi-uniform discharge mode to a filamentary discharge.

The effect of the temperature was also inspected and it was shown that the temperature had no visible effect on the ignition delay time in Plasma Assisted Ignition experiments, in comparison to autoignition experiments, even when the fuel presents a Negative Temperature Coefficient (NTC) region for autoignition experiments. Fast imaging experiments in this study also showed that the discharge in these gas density conditions was initiated at the surface of the electrode and induced a flat flame front propagation from the electrode system through the fresh gases. Numerical analysis of plasma action by dissociation of molecular oxygen was also conducted for a stoichiometric *n*-butane/O_2/Ar mixture at T_C= 725 K and P_{TDC} = 7.75 bar. It shows that once the initial atomic oxygen excess has been consumed to form an important radical pool, the main reaction pathways for n-butane and O remain unchanged when compared to similar stages for two-stage ignition.

To better understand the possible action of the discharge on the low temperature combustion mechanism, experiments were performed at lower pressures, where a transition is observed from a surfacic to a volumetric discharge, as pictured in Figure 4: The fuel was n-heptane, because of its well known low temperature reactivity [12]. Figure 4 displays images taken through the side window of the RCM at different pressures P_{TDC}, and for both polarities, at U=+50 kV and U=-55 kV.

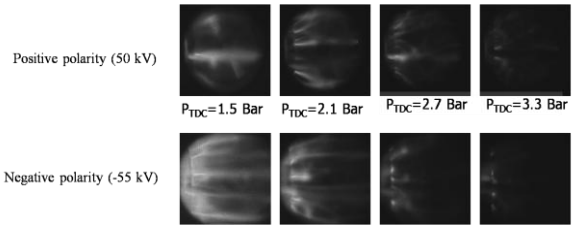

Figure 4: The ICCD imaging of the discharge inside a RCM combustion chamber at different P_{TDC} for both polarities. Left side: electrode, right side: RCM piston.

The results from Figure 4 are in accordance with the observations from [13] that show that the discharge morphology depends on the gas density, the chamber geometry and the nature of the gas mixture.

In these conditions, where the discharge travels across the combustion chamber, the chemical analysis of the experiment is easier than in the conditions where the discharge is surfacic. In this context, a case was observed, in which without discharge no ignition was observed, but in the case of Plasma Assisted Ignition two regimes were observed: fast ignition when a high voltage was applied, and a induced cool flame when a intermediate voltage was applied in the electrode system. Figure 5 shows the pressure profile as function of time in this case of a n-heptane/O_2/N_2 mixture with P_{TDC}=1.5 bar and T_C= 630 K.

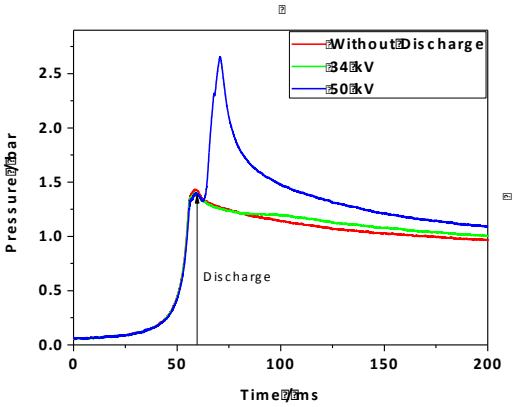

Figure 5 : Pressure profile as a function of time for the 3 situations : no ignition, cool flame, fast ignition. *n*-heptane/O_2/N_2 mixture, P_{TDC}=1.5 bar and T_C= 630 K.

As already observed in [5], this shows that the discharge can modify the limits of cool flame occurrence in the S diagram. It also shows that the plasma discharge can induce the low temperature combustion chemistry which is characterized by the cool flame. Working in these conditions will help us understand the interactions of the plasma and combustion chemistry. Emission spectroscopy

experiments were performed in the plasma induced cool flame conditions. They showed strong emission of N_2 ($C^3\pi_u$—> $B^3\pi_g$) during the discharge, as well as emission by excited formaldehyde CH_2O* during the induced cool flame. Figure 6 shows two emission spectra.

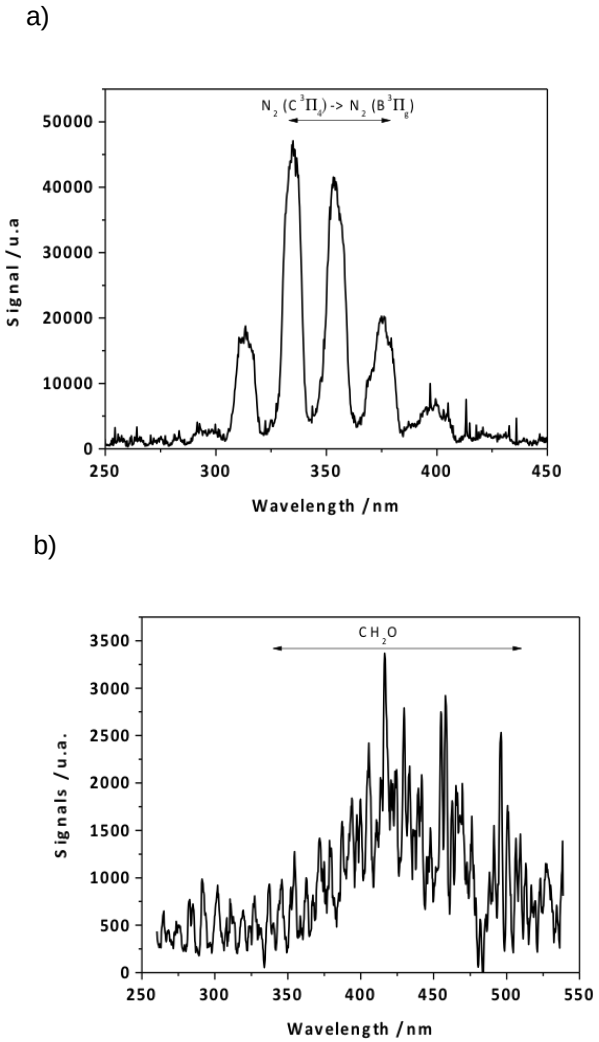

Figure 6: Emission spectra at two timings: a) During the the discharge and b) during the cool flame: *n*-heptane/O_2/N_2 mixture with P_{TDC}=1.5 bar and T_C= 630 K.

The formation of excited formaldehyde in the combustion systems is often believed to originate from the following reaction [14]:

$$CH_3^{\cdot} + {}^{\cdot}OH \rightarrow CH_2O^* + H_2O$$

A recent emission spectroscopy study [15] in engine conditions suggests that during spark ignition the emission was due to excited OH, NH, CN, NH and N_2, while during the flame the emission spectra are predominated by the emission of excited OH, CO_2, CH and soot precursors. The observed differences underline the non-equilibrium character of gas excitation by pulsed nanosecond discharge.

Conclusions

Emission spectroscopy experiments were performed in the case of Plasma Assisted Ignition in Rapid Compression Machine conditions. The discharge morphology depends on the gas density inside the combustion chamber with transition from surface discharge to a volumetric discharge as the pressure decreases.

In certain conditions a case was observed where:

-Without discharge no ignition was observed,

-With an intermediate voltage a cool flame was observed, without subsequent ignition,

-With a high voltage, fast ignition was observed.

The contributions of N_2 ($C^3\pi_u$—> $B^3\pi_g$) and excited formaldehyde were identified in the emission of the discharge and of the cool flame respectively.

Sampling experiments of the reactive system during the plasma induced cool flame are currently under way. Analyses with help from GC / MS techniques will help identify intermediate stable species formed during the plasma induced cool flame.

Acknowledgements

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