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► **To cite this version:**

Hugo Ajrouche, Ob Nilaphai, Camille Hespel, Fabrice Foucher. Impact of nitric oxide on n-heptane and n-dodecane autoignition in a new high-pressure and high-temperature chamber. Proceedings of the Combustion Institute, 2019, 37 (3), pp.3319-3326. 10.1016/j.proci.2018.07.102 . hal-02132939

HAL Id: hal-02132939

<https://hal.science/hal-02132939v1>

Submitted on 21 Oct 2021

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Impact of nitric oxide on n-heptane and n-dodecane autoignition in a new high-pressure and high-temperature chamber

Hugo Ajrouche^{a,*}, Ob Nilaphai^a, Camille Hespel^a, Fabrice Foucher^a

^a *Univ.Orléans, INSA CVL, PRISME, EA 4229F45072 Orléans, France*

Corresponding and Contact Author:

Hugo AJROUCHE
Université d'Orléans- Laboratoire PRISME
Polytech Orléans
8 rue Léonard de Vinci, 45072 – Orléans Cedex 1
Phone: +33-02-38-49-47-28
Fax.: +33-02-38-41-73-83
Email: hugo.ajrouche@univ-orleans.fr

Colloquium Topic Area: Spray, Droplet, and Supercritical Combustion

Total paper length (method 1): 5954 words

Main text: 2929 words (MSWord 2010 word count)

References: (30 references + 2) x (2.3 line/reference) x (7.6 words/line) = 559 words

Figure 1: [63 mm + 10 mm] x 2.2 words/mm x 2 column + (14 words in caption) = 335 words
Figure 2: [50 mm + 10 mm] x 2.2 words/mm x 2 column + (14 words in caption) = 278 words
Figure 3: [62 mm + 10 mm] x 2.2 words/mm x 2 column + (40 words in caption) = 357 words
Figure 4: [37 mm + 10 mm] x 2.2 words/mm x 2 column + (42 words in caption) = 249 words
Figure 5: [37 mm + 10 mm] x 2.2 words/mm x 2 column + (42 words in caption) = 249 words
Figure 6: [37 mm + 10 mm] x 2.2 words/mm x 1 column + (49 words in caption) = 152 words
Figure 7: [37 mm + 10 mm] x 2.2 words/mm x 1 column + (49 words in caption) = 152 words
Figure 8: [46 mm + 10 mm] x 2.2 words/mm x 2 column + (45 words in caption) = 291 words

Figure (total): 2063 words

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Table 1: (11 + 2 lines) x (7.6 words/line) x (1 columns) = 99 words

Table 2: (18 + 2 lines) x (7.6 words/line) x (2 columns) = 304 words

Table (total): 403 words

Submitted for consideration to the 37th International Symposium on Combustion, the Convention Centre Dublin, Ireland, 29 July-3 August 2018.

Abstract

A New One Shot Engine (NOSE) was designed to simulate the thermodynamic conditions at High Pressure-High Temperature like an actual common-rail diesel engine in order to study the compression ignition of spray. The volume of the combustion chamber provided with large optical windows simplified the implementation of various optical diagnostics. The advantage of this kind of set-up in comparison to pre-burn or flue chambers is that the initial gas mixture can be well controlled in terms of species and mole fraction. The purpose of this work was to investigate the impact of nitric oxide (NO) on ignition delay (ID) for two fuels with different cetane numbers: n-heptane, and n-dodecane. In the thermodynamic conditions chosen (60 bar and over 800–900 K), NO had a strong effect on ID, with increases in NO tending to reduce the ignition delay. Results showed that ID and Lift-Off Length (LOL) presented the same trend as a function of temperature and NO concentration. Experimentally, at 900 K the ignition of n-dodecane was promoted by NO up to 100 ppm, whilst higher NO levels did not further promote ignition and a stabilization of the value has been noticed. For n-heptane, stronger promoting effects were observed in the same temperature conditions: the ignition delays were monotonically reduced with up to 200 ppm NO addition. At a lower temperature (800 K) the inhibiting effect was observed for n-dodecane for [NO] greater than 40 ppm, whereas only a promoting effect was observed for n-heptane. The experimental results of LOL showed that NO shortened LOL in almost all cases, and this varied with both the NO concentration and the mixture temperature. Thus, fuels with shorter ignition delays produce shorter lift-off lengths.

Keywords: Nitric oxide, Engine Combustion Network (ECN), Spray and Combustion Characterization, Lift-Off Length, Ignition Delay.

1. Introduction

During the last decade, several new combustion chambers were developed to reach the thermodynamic conditions of current common-rail diesel engines [1–3], for example: Constant-Volume Preburnt (CVP) chamber, Constant-Pressure Flow (CPF) chamber, Rapid Compression Machine (RCM) or Optical Research Engine (ORE) [2,3]. Moreover, a new kind of RCM, called “Rapid Cycling Machine (RCYM)” [4,5], seems to be an interesting option to study spray in high pressure and high temperature conditions, due to the absence of combustion products as in the CVP and the possibility of covering a full range of diesel engine working conditions (up to 100 bar). The New One Shot Engine (NOSE) was designed to simulate these thermodynamic conditions at high pressure and high temperature [6,7] in order to study the diesel spray and combustion relevant to engines that use a moderate rate of Exhaust Gas Recirculation (EGR). The advantage of this kind of set-up in comparison to pre-burn or flue chambers is that the initial gas mixture can be well controlled in terms of species and mole fraction. For example, spray combustion can be achieved in a more realistic environment without H_2O and CO_2 or with species representing the burnt gases.

In piston engines, the reacting mixture always contains a certain amount of residual gases resulting from the incomplete combustion of species such as carbon monoxide (CO) and unburned hydrocarbons in the previous combustion cycle. One of these leftover combustion species is nitric oxide (NO) which has an impact on the autoignition kinetics. Some studies have reported that the appearance of small quantities of NO typically occurs inside the CVP at a concentration of a few dozen ppm after mixing with the fresh charge [6–8]. No studies are yet available confirming the absence of NO during premixed burn in this kind of chamber. However, the existence of these burnt products has been reported in few studies to have a significant impact on autoignition in SI engines [6–8] and HCCI engines [9–13]. Ignition delay (ID) is a key combustion property for fuels used in compression-ignition (CI) engines, and is commonly used as the metric for the validation of

combustion simulations. A promoting effect of NO was often observed, particularly at high temperatures and low NO concentrations, whereas inhibiting effects have also been reported in other conditions [14–16].

The purpose of this work was to study the influence of NO on ID for different fuels in NOSE. Pure n-heptane and pure n-dodecane were the fuels used. First, to determine the ability of fuels to auto-ignite, the ignition delays of these fuels without NO were characterized and compared. Experiments were conducted at the Spray A operating conditions of ECN in NOSE. The engine set-up was also improved to extend conditions to different ambient temperatures (800 K and 850 K). To provide a homogeneous temperature field at 800 K or 900 K two specific compression ratios were tested 12.6 and 15 respectively. In the thermodynamic conditions chosen (60 bar and over 800–900 K), the effect of NO on ID and LOL was studied by adding 0-200 ppm NO to the engine intake. To measure ID, different techniques such as Schlieren visualization and OH* chemiluminescence were used to analyze the cool flame and the high temperature ignition (hot flame). The Lift-Off Length (LOL) was measured by OH* chemiluminescence images. The observed influence of NO addition on ID and LOL was analyzed. The related chemical reasons responsible for the effects are then discussed.

2. Experimental approach

2.1. Experimental facility

The NOSE set-up is based on a single-cylinder engine (155 mm bore diameter and 177.8 mm stroke), driven by an electric motor (Figure 1). The speed of the electric motor is controlled as a function of the crank angle, so that the time for the compression stroke (40 ms), a plateau near the Top Dead Centre (TDC) (15 ms) and the expansion stroke are completely known during one rotation (Figure 2). For each run, the engine is used during a single compression-expansion stroke. The original cylinder

head was replaced by a dedicated chamber, designed by extending the combustion chamber to enable optical measurements and the study of Diesel sprays. There are 4 quartz windows, 25 mm thick, 25 mm wide, and 80 mm long to support several kinds of optical measurement. The operation of NOSE starts by setting the initial conditions such as initial pressure, initial temperature and gas composition at Bottom Dead Center (BDC). After creating a vacuum, different gases (N_2 , O_2 , Air and bottle of NO at 200 ppm mixed into N_2) were injected into the chamber by using mass flow controller (Brooks instruments). The initial temperature was specified by controlling the water temperature around the liner and 4 heaters in the cylinder head, and the temperature of the injector was maintained at the same temperature as the cylinder head ($90\text{ }^\circ\text{C} \pm 0.5\text{ }^\circ\text{C}$ for all experiments). Two type-K thermocouples were placed inside the chamber and casing (near the injector) to measure the gas temperature inside the chamber and the casing temperature, respectively. A high pressure pneumatic pump (MAXIMATOR M189 DVE-HD) maintained the injection pressure around 1 500 bar (± 20 bar) before injection. The injection pressure was measured by a piezo-electric pressure sensor (KISTLER Type 6533A11) with a charge amplifier. As this new device compresses ambient gases during only one cycle from BDC to TDC, it is called a “New One Shot Engine.”

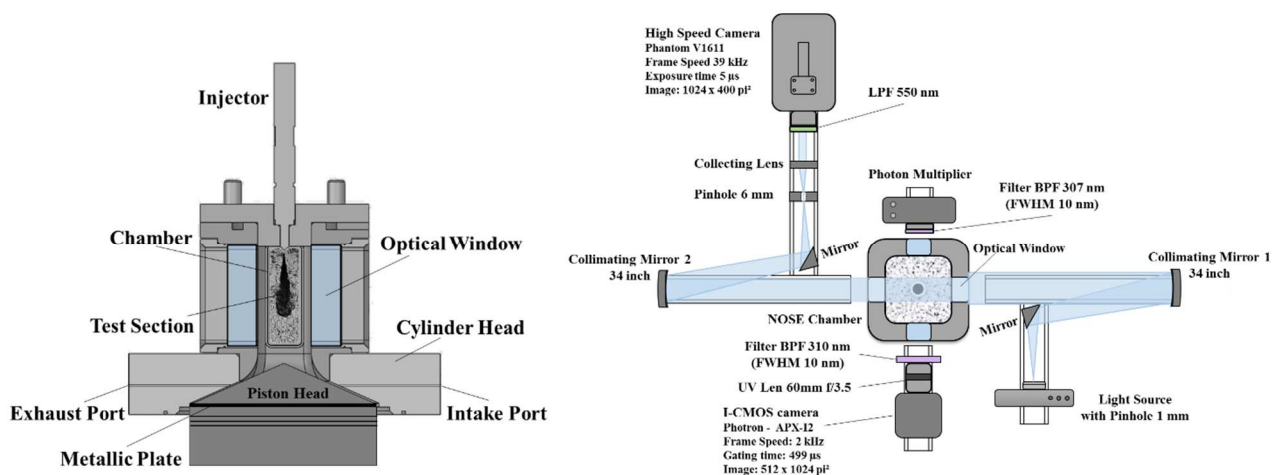


Figure 1: The NOSE chamber: cross-section (left) and experimental scheme of the optical set-ups (right).

The cylinder pressure, temperature inside the vessel, injector driving current, photomultiplier signal and crank angle were recorded as a function of time with a National Instrument CompactRIO at 250 kHz. A high response piezo-electric pressure sensor (KISTLER 7001) was installed at the chamber head and coupled with a charge amplifier (KISTLER 5011). All the fuel injection system, comprising the Common-Rail, injector, high pressure tube, and injector, are the same specification as ECN Spray A injection devices [17]. Further details about the chamber design, thermal characterization chamber, specification of the injection system, and data acquisition system were presented in our previous studies [6,18,19].

2.2. Spray condition

Spray-A condition (Table 1) was the chosen reference standard condition. The operating conditions (800 K and 900 K ambient gas temperature, about 60 bar cylinder pressure and 22.8 kg/m³) are typical of the real operating conditions of engines that use a moderate rate of EGR.

Table 1.

Standard Spray-A operating conditions of ECN [20].

Parameter	Value
Ambient gas temperature	800 and 900 K
Ambient gas pressure	about 60 bar
Ambient gas density	22.8 kg/m ³
Ambient gas velocity	Near-quiescent, less than 1 m/s
Fuel injection pressure	150 MPa (1 500 bar)
Fuel	n-dodecane (Cetane number = 80 [21], Boiling temperature =489 K) and n-heptane (Cetane number = 56 [21], Boiling temperature =372 K)
Fuel temperature at nozzle	363 K (90°C)
Injection duration	1.5 ms

To achieve standard Spray-A conditions for the lower temperature (800 K), the CR was changed from 15:1 to 12.3:1 by modifying the metallic plate of the piston (see Figure 1-left), thus changing

the volumetric compression ratio. The initial pressure at the BDC made it possible to achieve 800 K and 22.8 kg/m³ at the end of compression.

2.3. Optical diagnostics

Table 2 summarizes the optical set-ups for the different techniques needed to characterize macroscopic combustion parameters, following the ECN recommendations to ensure accurate experimental results. LOL was measured by OH* chemiluminescence images, and ID from high speed Schlieren images, and the OH* chemiluminescence temporal signal recorded by the photomultiplier as shown in Figure 1.

Table 2.

Summary of the optical set-ups

Parameters	Lift-Off Length		Ignition Delay
	OH*Chemiluminescence	Schlieren	OH*Chemiluminescence
Camera/Detector	Photron - APX-I2	Phantom-V1611	Newport Photomultiplier tube 70680
Sensor type	Intensified-CMOS	CMOS	-
Light source	-	LED (white) with 1 mm pinhole	-
Lens	UV 60 mm f/3.5	-	-
Mirrors	-	2 Parabolic Mirrors 34 inch	-
Pin hole	-	6 mm	-
Filter	BPF 310 nm , FWHM 10 nm(Asahi ZBPA310)	LPF ≤ 550 nm (Asahi XVS0550)	BPF 307 nm, FWHM 10 nm (Ealing 35-7939)
Frame speed	2 kHz	39 kHz	-
Exposure time	499 μs	5 μs	-
Image size	512 x 1024 pix ²	1024 x 400 pix ²	-
Magnification	18.2 pix/mm	12.3 pix/mm	-
Repetitions	10 tests	10 tests	10 tests

LOL is defined as the distance between the injector tip and the stabilized flame of the high temperature reaction zone [5,6]. As is well known, at high temperature the chemiluminescence spectrum of OH* is centered at 310 nm, so OH* is a good indicator to measure the LOL. According to the methodology developed by [22], the LOL was determined as the gradient of the OH* intensity

along the chemiluminescence image recorded by an ICCD Photron Fastcam APX I2 and a 60 mm f/3.5 UV lens and equipped with a 310 nm (FWHM10) band-pass filter (BFP). A long constant gating time of 449 μs was used to allow turbulent fluctuation in the LOL to reach an average as well as to provide an image of the quasi-steady mean LOL after the Start Of Injection (SOI) [22]. LOL was defined as the average distance between the injector tip and the first value above 50 % of the maximum intensity of each profile, following the ECN method [20].

The Schlieren set-up, presented in Figure 1, is composed of a continuous white LED with a 1 mm pinhole, mirrors and collimating mirrors to reflect light through the test section in the NOSE chamber. A 6 mm pinhole was set up to record Schlieren light by the high speed camera (Phantom V1611). A 550 nm low-pass filter was added to limit emissions due to soot radiation.

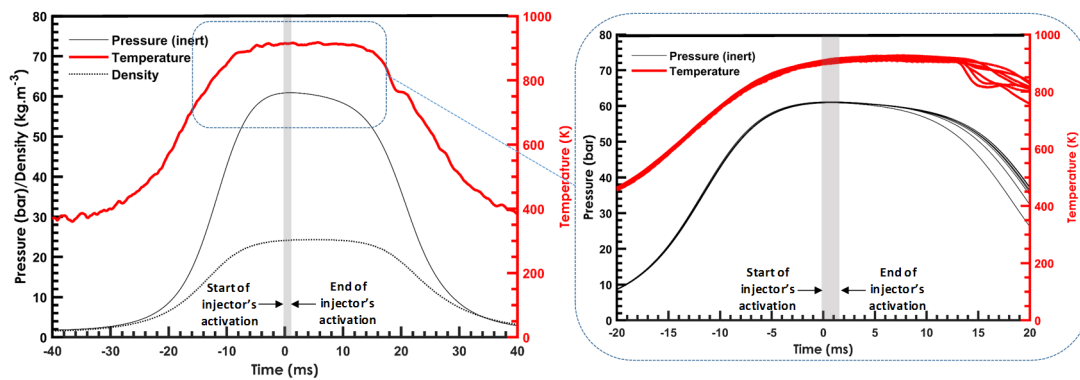


Figure 2: Average thermodynamic measurement results and zoom caption of 10 superimposed pressure and temperature curves.

As can be seen in Figure 2, the measured pressure and temperature reach the requirement of the standard Spray-A condition around TDC at 900 K and 60 bar, so a required density of 22.8 kg/m^3 . The results clearly show that the disparity of the values of temperature and pressure are lower than 1 % during the injection duration and confirm that NOSE can achieve a wide range of standard Spray-A conditions. SOI was defined here as the time between the start of injection current and the first injection image of the spray obtained by Schlieren visualization, usually called “hydraulic delay”. To

use the same criterion to define ID with both techniques, the results shown in this work were corrected by the hydraulic delay. The cool and main flame ignition delays were detected using the Schlieren images. OH* chemiluminescence was also used to detect the main flame ignition delay by using a Photomultiplier tube side-on and a band-pass filter of 307 nm. It was set up close to the window and positioned on the opposite side to the ICCD camera, as shown in Figure 1.

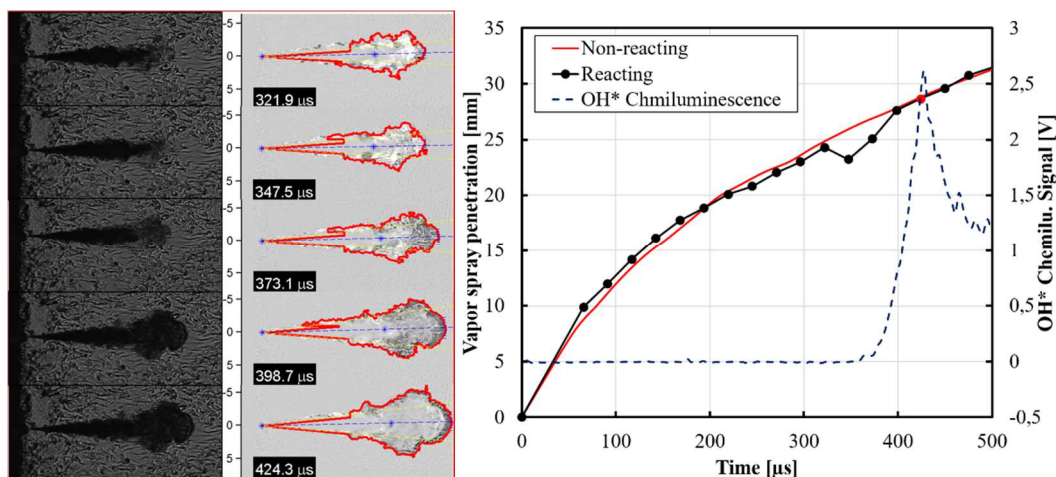


Figure 3: The series of Schlieren images and post-processed images during cool flame and hot flame events (left) and validation of the ignition delay criterion on Schlieren images by the OH* chemiluminescence collected by a Photomultiplier at 900 K on n-dodecane (right).

3. Results and discussion

3.1. Ignition Delay

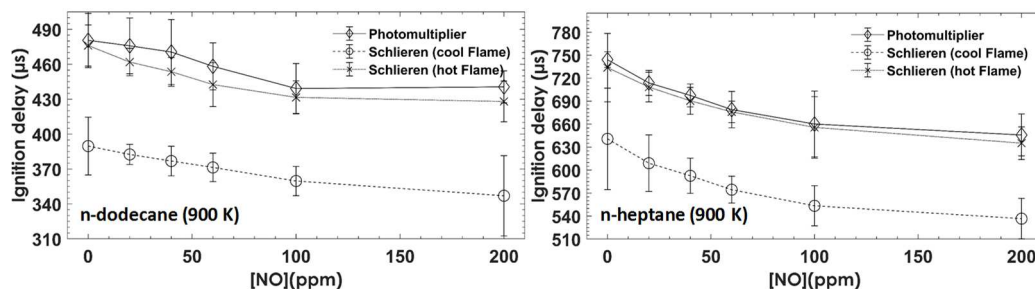


Figure 4: Comparison of ignition delay obtained from the cool and hot flame to those obtained from the photomultiplier as a function of percent of NO in the new one shot engine for n-dodecane (left) and n-heptane (right) at 900 K with 15% O₂.

The ignition delays of the cool and the main flames are plotted as a function of NO concentration at 900 K (Figure 4, Figure 5) for n-heptane and n-dodecane fuels. The ID measured with both Schlieren and photomultiplier techniques are in good agreement, with a disparity below 5 %. When no NO was added, the ignition delays were found to be close to those determined by [17] in the same spray conditions. The ID followed the expected trend with respect to the fuel cetane number, decreasing with increasing cetane number and with increasing ambient temperature. The high temperature ID (hot flame) is preceded by the appearance of the cool flame in all cases (Figure 4). The ID of n-heptane and n-dodecane decreased monotonically as the NO concentration was increased, in agreement with the conclusion by Chen et al. [24] where n-heptane ignition was promoted by NO up to 100 ppm in a rapid compression machine.

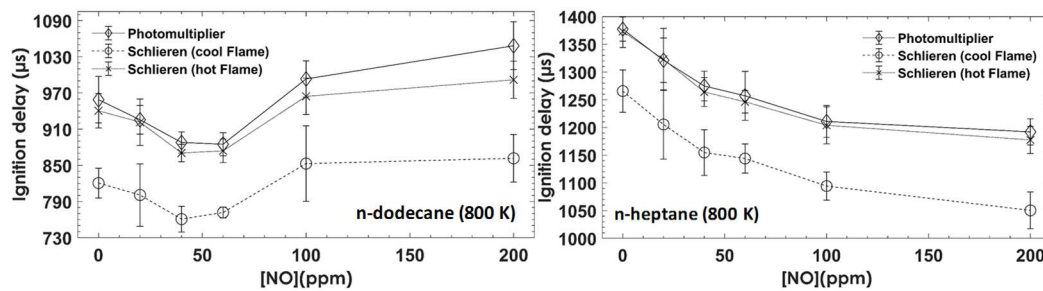


Figure 5: Comparison of ignition delay obtained from the cool and hot flame to those obtained from the photomultiplier as a function of percent of NO in the new one shot engine for n-dodecane (left) and n-heptane (right) at 800 K with 15% O₂.

NO had a direct impact on the detection of the cool flame. The difference observed in this work between cool and hot flame was constant (around 100 µs) with increasing NO. This constant value can be explained by the dynamism of turbulent cool flame propagation described by Dahms et al. [25] which is not impacted by NO. At 800 K, the same tendencies were found for n-heptane (Figure 5). However, the cool and hot flame delays of the n-dodecane spray were not monotonic. From a concentration of 0 to 40 ppm of NO, the delays decreased then increased to reach a delay greater than without NO when the concentration of NO was greater than 100 ppm. Thus, at this operating

condition, the effect of NO on the autoignition delay process of n-dodecane exhibited two different aspects: NO can both promote and inhibit ID, depending on the added concentration.

Figure 6 shows the normalized ignition delays of n-dodecane and n-heptane for two temperatures, 800 K and 900 K, as a function of percent of NO. For n-heptane, ID was monotonically shortened as more NO was added at the two temperatures. Thus, NO concentration had the same promoting effect and at 200 ppm the ID was reduced by 15 %. However, for n-dodecane at low temperature (800 K), NO slightly promoted total ignition at lower levels of addition. This promoting effect peaked at around 40 ppm of NO, after which the ID increased with further NO addition, thus the impact of NO was less significant ± 10 %. NO tends to increase the extent of oxidation for high-octane fuel components. However, for the low-octane component n-heptane and n-dodecane, NO has an inhibiting effect on hydrocarbon oxidation, particularly at low temperatures. In this work, this effect has been observed for n-dodecane. The different behaviors seen for n-dodecane and n-heptane could be explained by the dominant inhibiting impact of NO on n-dodecane at low temperatures, which is less significant for n-heptane. This behavior is further explained in terms of reaction kinetics in the NO chemistry.

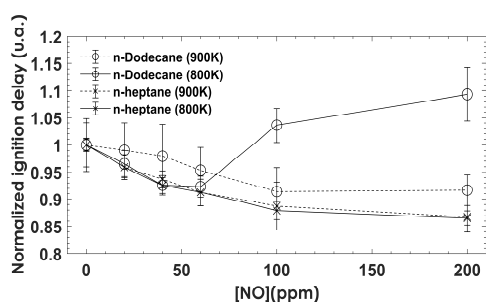


Figure 6: Normalized ignition delay with respect to ignition delay without NO as a function of percent of NO in the new one shot engine for n-dodecane (circles) and n-heptane (crosses) at 900 K (dashed line) and 800 K (solid line). NO additions from 0 ppm to 200 ppm.

3.2. Lift-off length

While the ignition delay characterizes the first ignition area of the spray, the LOL can be associated to a stationary regime where the flame is stabilized behind the liquid spray. For both temperatures, the LOL were plotted as a function of the NO concentration. The LOL for the n-heptane fuel was affected by NO and decreased linearly, though the effect on the LOL was found to be slightly less than for the cool and the main flame. However, whereas the NO concentration decreased the LOL for the n-dodecane spray at 900 K, a decrease then an increase of the LOL was found at 800 K. By considering a different combustion parameter and using another diagnostic, this result confirms the promoting and inhibiting effect of ID observed in the previous section. The promoting or the inhibiting effect of the NO is therefore similar to that of the cool and the main flame but with a lesser impact on the LOL.

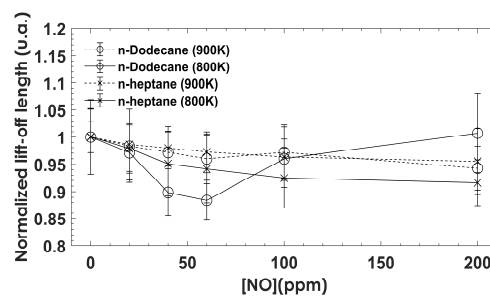


Figure 7: Normalized lift-off length with respect to lift-off length without NO as a function of percent of NO in the new one shot engine for n-dodecane (circles) and n-heptane (crosses) at 900 K (dashed line) and 800 K (solid line). NO additions from 0 ppm to 200 ppm.

3.3. Simulation

To understand the impact of NO on the ignition stages, a Senkin constant volume simulation was carried out using the same conditions as in the experiments ($T=800$ K and 900 K, $O_2=15\%$ and $P=60$ bar) for a large range of equivalence ratios and for a wide range of NO concentrations. The simulation was performed for vaporization kinetics in the premixed case at constant volume. The

PFR mechanism including the NO sub-mechanism from Dubreuil et al. [15] was used to simulate n-heptane oxidation. For n-dodecane, the NO sub-mechanism from [15] was included in the mechanism from Mz -Ahmed et al. [26]. The main flame delays were determined from the temperature histories.

The promoting and inhibiting effects are presented as the percentage of the ID when no NO is added. For n-heptane, NO decreased the ignition delay regardless of the equivalence ratio when the concentration was less than 50 ppm. After 50 ppm at 800 K, a plateau was reached for an equivalence ratio greater than 1 and the promoting effect of NO increased for a lean equivalence ratio. At 900 K, the promoting effect was more constant but always greater for a lean equivalence ratio. From Figure 8, at 200 ppm of NO, the delay was found to be 87% of the delay without NO for both temperatures. That means that, at 800 K, ID occurs when the equivalence ratio is leaner than at 900 K.

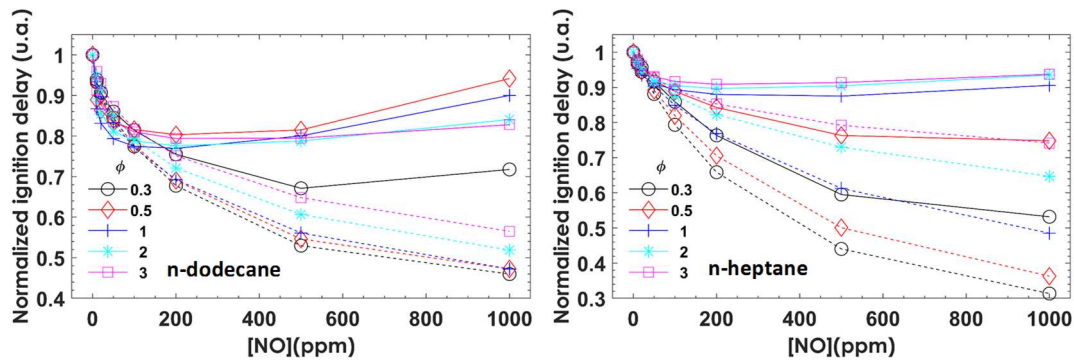


Figure 8: Normalized ignition delay with respect to ignition delay without NO as a function of percent of NO for different equivalence ratios at 900 K (dashed line) and 800 K (solid line) for n-dodecane (left) and n-heptane (right). NO additions from 0 ppm to 1000 ppm.

The ignition delay predictions from simulation show that at 800 K, the ignition of n-dodecane was promoted by NO up to 200 ppm, whilst higher NO levels did not further promote ignition. However, at poor region, higher NO levels had an inhibiting effect on n-dodecane ignition. At the higher temperature, NO promoted the ignition delay in all cases. According to the kinetic modeling, the acceleration of the low-temperature reactions resulting in a reduction of the low temperature heat

release delay is due to the production of active species through reactions involving NO, as already demonstrated under plug-flow and jet-stirred reactor conditions [27,28]: $\text{NO} + \text{HO}_2 = \text{NO}_2 + \text{OH}$ and $\text{NO} + \text{RO}_2 = \text{NO}_2 + \text{RO}$. The $\text{NO} + \text{RO}_2$ reactions are less pronounced or influential than the $\text{NO} + \text{HO}_2$ reaction, as demonstrated in the literature [28,29]. At high NO concentrations, the inhibiting process, via: $\text{NO} + \text{OH} + \text{M} = \text{HONO} + \text{M}$ and $\text{ROOH} + \text{NO} = \text{RO} + \text{HONO}$, becomes an increasingly important competitor that could even significantly delay low-temperature ignition, as demonstrated before [30].

4. Summary and conclusions

A new experimental set-up was designed and improved to guarantee high quality results based on Spray A conditions. At the thermodynamic conditions chosen (60 bar over 800– 900 K), the ignition delays of n-heptane and n-dodecane in varying amounts of NO (0–200 ppm) were measured in a rapid cycling machine NOSE. The consistency of the data obtained from ID and LOL indicates a good level of repeatability and outlines a global agreement with the ECN dataset for 0 ppm of NO. Experimental results showed that NO advanced ignition in most cases. NO had a strong effect on ID, with increases in NO tending to reduce the ignition delay at 900 K. The effect of NO on the autoignition delay process of n-dodecane exhibited two different aspects at 800 K: NO can both promote and inhibit the ignition delays, depending on the NO concentration added. Results obtained show that ID and LOL present the same trend as a function of temperature and NO concentration. Fuels with shorter ignition delays generally produce shorter lift-off lengths. In following work, the effect of different species in the ambient gases will be studied to assess their impact on combustion development and soot production/oxidation. Additionally, the influence of the droplet size distribution on ID will be performed by laser diffraction analysis.

Acknowledgements

The authors acknowledge the French National Research Agency (contract ANR-14-CE22-0015-01) for financial support to the ECN-France project and Region Centre Val de Loire (CPER 2007-2013 Energies du Futur) and FEDER for financial support to build the experimental set-up.

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Figure captions:

Figure 1: The NOSE chamber: cross-section (left) and experimental scheme of the optical set-ups (right).

Figure 2: Average thermodynamic measurement results and zoom caption of 10 superimposed pressure and temperature curves.

Figure 3: The series of Schlieren images and post-processed images during cool flame and hot flame events (left) and validation of the ignition delay criterion on Schlieren images by the OH* chemiluminescence collected by a Photomultiplier at 900 K on n-dodecane (right).

Figure 4: Comparison of ignition delay obtained from the cool and hot flame to those obtained from the photomultiplier as a function of percent of NO in the new one shot engine for n-dodecane (left) and n-heptane (right) at 900 K with 15% O₂.

Figure 5: Comparison of ignition delay obtained from the cool and hot flame to those obtained from the photomultiplier as a function of percent of NO in the new one shot engine for n-dodecane (left) and n-heptane (right) at 800 K with 15% O₂.

Figure 6: Normalized ignition delay with respect to ignition delay without NO as a function of percent of NO in the new one shot engine for n-dodecane (circles) and n-heptane (crosses) at 900 K (dashed line) and 800 K (solid line). NO additions from 0 ppm to 200 ppm.

Figure 7: Normalized lift-off length with respect to lift-off length without NO as a function of percent of NO in the new one shot engine for n-dodecane (circles) and n-heptane (crosses) at 900 K (dashed line) and 800 K (solid line). NO additions from 0 ppm to 200 ppm.

Figure 8: Normalized ignition delay with respect to ignition delay without NO as a function of percent of NO for different equivalence ratios at 900 K (dashed line) and 800 K (solid line) for n-dodecane (left) and n-heptane (right). NO additions from 0 ppm to 1000 ppm.