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## Effects of Biomass Powder Direct Injection in Controlling Lean Homogeneous combustion

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#### ABSTRACT

Homogeneous Charge Compression Ignition (HCCI) is an innovative strategy to reduce pollutant emissions at the source. In order to control lean combustion ignition, various approaches can be applied. In this work, we propose to control the ignition of reactive biogas mixtures by direct injection of biomass powder. Thus, a particle injector was integrated into an RCM (Rapid Compression Machine) chamber, allowing the study of diphasic gas/solid combustion.

The preliminary tests were applied to different biogas mixtures of  $CH_4$ ,  $CO_2$ , and  $H_2$  having particularly interesting properties. The study highlights the advantages of using biomass mixtures with lean homogeneous combustion. The results were satisfyingly reproducible, highlighting the benefits of injected particles in controlling the ignition timing of HCCI engines.

KEYWORDS: HCCI, Biomass, Biogas, Diphasic Combustion, Ignition Control

#### **1** INTRODUCTION

Lean homogeneous combustion (referred to as the acronym HCCI) is a clean technology that reduces soot and NOx emissions at the source while maintaining appreciable efficiency due to its high combustion rate (Sharma, Rao and Murthy, 2016), (Kraft *et al.*, 2000). In addition, emissions can be even more reduced by replacing conventional fuels with biomass-derived fuels such as biogas or syngas. Biogas is considered as a promising candidate to meet the regulation emission targets due to its flexibility as an energy source and its wide range of applications. When purified, it is a good fuel for application in internal combustion engines due to multiple benefits (Mustafi, Raine and Bansal, 2006):

- It is considered environmentally friendly, as its combustion does not increase the net amount of CO<sub>2</sub> in the atmosphere.
- It ensures maximum availability as it is produced from waste biomass materials.
- It can replace a substantial amount of fossil fuels throughout the world, and thus the emissions.

However, due to its considerable amount of  $CO_2$ , biogas has a low calorific value that the addition of hydrogen can enhance. Therefore, combining biogas/H<sub>2</sub> mixtures with HCCI technology is the best compromise between a low environmental impact and engine efficiency. Nonetheless, the control of HCCI combustion timing remains a crucial problem to be solved. Multiple strategies aiming to control the ignition start are studied in the literature. Whereas, in our research, we aim to prove that the direct injection of solid biomass into the reactive gas mixture is an efficient strategy to control HCCI timing and reduce pollutant emissions. Using coal particles in internal combustion engine was studied by several authors in the literature. Nevertheless, only a few existing programs in the literature consider using dry biomass powder in internal combustion engines (Stover *et al.*, 2019).

The injection of solid particles carried by gaseous vectors will introduce turbulence during the initiation and development of combustion, improving gas homogenization. In addition, the suspended particles may control and stabilize the lean homogeneous combustion of the reactive mixture and ignite it simultaneously in different combustion chamber zones, which can offset the thermal heterogeneities. Based on a gaseous reactive mixture, the quasi-homogeneous species concentration will provide the optimal conditions for the non-production of soot.

In this paper we aim to study the biomass particle's combustion in a hot gaseous mixture, as well as the interaction of particles with the reactive gas mixture. The experimental tests are conducted on the rapid compression machine of Sorbonne University (SU RCM). A particle injector is implemented into the center of the combustion chamber allowing direct injection of powder with a gas vector (nitrogen in our case). The effects of particle injection were studied qualitatively on an inert mixture and two reactive mixtures of biogas enriched with hydrogen. The choice of these mixtures as well as the lean homogeneous conditions are discussed in this work.

#### 2 MATERIALS AND METHODS

#### 2.1 Experimental setup

The experimental setup used to study the effects of particle injection on the lean homogeneous combustion of biogas is the SU rapid compression machine equipped with a particle injector. The schematic diagram of the experimental setup is presented in Figure 1.

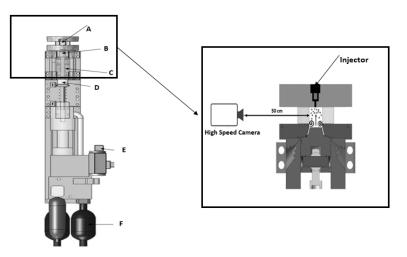


Figure 1: Schematic diagram of the SU rapid compression machine with the particle injector. A. Combustion chamber, B. Piston, C. Pre-compression chamber, D. Hydraulic cylinder, E. Three stage servo-valve, F. Pressure accumulator

The major components of the RCM are the hydraulic system, the single-piston, the combustion chamber, and the system of control and acquisition. The hydraulic system consists of a hydraulic power unit, oil pressure accumulators, a three-stage servo valve, and a hydraulic cylinder. It is responsible for controlling the piston's movement, allowing it to compress the gas mixture in the pre-compression chamber and to be locked at the top dead center (TDC), keeping the compressed gas in the combustion chamber to maintain high pressure and temperature for a longer time.

The studied gas mixtures were prepared monometrically inside a large vacuum tank of 30 L allowing dozens of shots with the same load. It is equipped with a propeller to make the mixture homogeneous and a capacitance diaphragm manometer for measuring partial pressures. The experimental procedure is kept the same for all studied tests to ensure the reproducibility of the process.

Experiments were carried out with a compression ratio of 15 and a relatively short compression time of 30 ms to avoid chemical reactions in the compression phase. The technical features of the experimental setup are described in detail in Ref. (Guibert, Keromnes and Legros, 2007). A particle injector was integrated into the combustion chamber center to allow the direct injection of solid particles.

The combustion chamber is a quartz cylinder (40x40 mm), enabling optical access.

In the present work, a high-speed intensified camera is used to record the ignition events. No filter is used, and therefore, the visualized signal corresponds to the visible light intensity. It is important to remark that the captured signal includes black body emission and chemiluminescence emission. The exposure time is set to 99  $\mu$ sec and the frame rate to 10 kHz, yielding a resolution of 640×480 pixels, which is proved to be sufficient for proper visualization of the particles.

#### 2.2 Choice of the biogas mixtures

Our research aims to find an ideal composition of biomass-derived fuel with minimum emissions and high combustion efficiency. Only a small part of this research is presented in this paper.

Biogas is a clean energy produced by anaerobic digestion of solid biomass, also known as methanisation. Its composition differs according to the sources, technology, and operational conditions of its production. Typically, it consists of 30-75% methane, with the remainder being mainly carbon dioxide, hydrogen, and water vapor (Occitanie, 2018). The use of biogas in engine applications is a good alternative, significantly when enhanced by hydrogen. In addition, its low calorific value can be recovered by using hydrogen. The presence of  $CO_2$  present in biogas leads to a decrease in NOx emissions (similar to the dilution effects of EGR).

In this study, we focus in particular on biogas enriched with hydrogen mixtures. Thus, the duality biofuels/ HCCI combustion engine combines the advantage of alternative fuels as well as alternative combustion strategies. In general, lean homogeneous combustion has the benefit of the dilution rate in reducing local NOx and soot emissions. According to an investigation conducted by Mittal et al. (Mittal, Raju and Sung, 2012), higher end of compression pressure and temperature resulted in higher temperature field uniformity. Thus, an end of compression pressure of 8 to 11 bars was chosen, with a low to an intermediate temperature of 940 K, mainly considered to reduce NOx emissions.

The end of compression temperature can be varied by changing the proportions of diluted gases for the same fuel composition (Di *et al.*, 2014), (Würmel *et al.*, 2007). However, a significant amount of charge dilution is required in order to achieve lean homogeneous combustion (Izadi Najafabadi and Abdul Aziz, 2013). Thus, a dilution rate of 70% is considered in this study.

In order to choose the biogas/H2 mixtures used in this study, numerical simulations were performed on Chemkin-Pro using a closed batch reactor. Numerous iterations were proposed to the adiabatic core HRM for simulating RCM experiments. For example, Curran et al. (Curran *et al.*, 1998) demonstrated that under some conditions, chemical reactivity during piston compression can significantly alter the auto-ignition process and thus induction times, which motivates the account for gas reactivity during compression within the simulations. As a result, Mittal (Donohoe *et al.*, 2014) proposed considering heat loss during and after the compression phase. This can be done by using the volume profile generated from the non-reactive pressure trace issued from the RCM in Chemkin simulations. Based on these guidelines, the combustion process of biogas mixtures was simulated, taking into consideration compression and heat transfer.

In order to study the influence of hydrogen and biogas composition on the auto-ignition of a reactive gas mixture, several simulations were carried out on Chemkin with different volume proportion of hydrogen: 0.1, 0.2, 0.3 and 0.5, and a temperature of 1020 K.

The evolution of the auto-ignition delay times of the three biogas lean mixtures ( $\phi$ =0.6) at T<sub>c</sub>=1020 K with hydrogen proportion is presented in Figure 2.

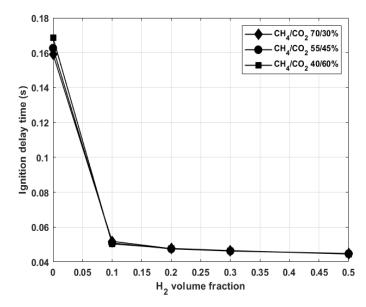


Figure 2 : Effects of hydrogen on the ignition delay times of three biogas mixtures

It is shown that the auto-ignition delay time decreases when the hydrogen percentage in  $CH_4/CO_2$  mixtures increases. The influence is the highest for a hydrogen percentage lower than 10%; then, the auto-ignition delay time decreases slightly. In other words, the addition of a small amount of hydrogen is sufficient to take control of the reactivity of pure methane mixture.

This promoting effect of hydrogen addition on methane combustion has been studied experimentally and numerically by several authors (Cheng and Oppenheim, 1984) (Porpatham, Ramesh and Nagalingam, 2007), (Ma *et al.*, 2014). It was explained by Gersen et al. (Gersen *et al.*, 2008) by the fact that the activation energy of hydrogen is two times higher than methane. ( $E_a(H_2) = 2*E_a$  (CH<sub>4</sub>)).

It appears that the promoting effect of hydrogen on self-ignition is mainly due to its chemical kinetics. However, its kinetic effect is difficult to determine since its oxidation results from the formation of radicals, H, O, OH, and HO<sub>2</sub>, and its behavior varies, whether at low or high temperatures.

For high temperatures, Huang et al. (Huang *et al.*, 2006) studied the auto-ignition times of stoichiometric  $CH_4/H_2/air$  mixtures in shock tubes for end pressures and temperatures of 16 to 40 bar and 1000 to 1300K, respectively. They explained the hydrogen promoting effect by two main causes:

- The decomposition of hydrogen into H radicals at high temperatures and their participation in the H+O<sub>2</sub>→O+OH reaction
- The rapid oxidation of hydrogen following the reaction  $OH + H_2 \rightarrow O + H_2O$

Huang et al. (Huang *et al.*, 2006) also explained that the slow  $CH_3O_2 + H_2$  reaction is the reason why the effect of hydrogen is less important at low than at high temperatures. More recently, Liu et al. (Liu *et al.*, 2018) conducted experiments on Tsinghua's MCR to determine the effects of hydrogen and carbon monoxide on the self-ignition delay and flame spread of methane for stoichiometric and rich mixtures at temperatures of 860-1080 K and a pressure of 20 bar. They found that the addition of H<sub>2</sub> has a small influence on the self-ignition delay for temperatures lower than 950 K and a large influence for temperatures above this value. Carbone dioxide is shown to affect less the ignition delay times in the presence of at least 10% hydrogen. However, its presence reduces the formation of soot and nitric emissions(Bhaduri, Jeanmart and Contino, 2017). Hence, biogas mixtures containing 45 and 60% of  $CO_2$ , enriched with 10% hydrogen and diluted with 70% Ar /CO<sub>2</sub>. are considered for this study. The end of compression temperature is nearly 990 K, mainly chosen to reduce NOx emissions. The particles are injected into the reactive mixtures with an injection pressure of ~20 bar.

The experimental conditions of these two reactive mixtures used to study the effects of solid particles injection are presented in Table 1.

Mixtures	CH <sub>4</sub> /CO <sub>2</sub> (Volume %)	H <sub>2</sub> (Volume %)	Dilution rate Ar/ N <sub>2</sub> (%)	ф (-)	Pressure (bar)	Temperature (K)
1	55/45	10	70	0.6	8	990
2	40/60	10	70	0.6	11	990

Table 1: Biogas/H<sub>2</sub> mixtures studied with solid biomass particles

#### 2.3 Choice of the biomass samples

In this study, the main objective is to find an eco-friendly/efficient technology to reduce pollutant emissions at the source. Thus, the injected particles are of biomass origin, particularly lignocellulosic biomass. The latter primarily comprises macromolecular substances (cellulose, hemicellulose, and lignin) with smaller amounts of extractives and inorganics (Vassilev *et al.*, 2010). Therefore, in order to understand the diphasic combustion gas/solid, it is essential to better understand first the combustion of solid biomass. The process of biomass combustion involves several physico-chemical aspects of high complexity and can be achieved in four steps: heating and drying (water vaporization), devolatilization (pyrolysis), combustion of gases, and char burning (Vassilev *et al.*, 2010).

According to the literature, several parameters can influence the ignition process of powder, including the temperature, oxygen concentrations, and the type and size of these particles (Khatami, Stivers and Levendis, 2012), (Simões *et al.*, 2017), (Li, Paul and Czajka, 2016).

According to the timing of the ignition onset, three modes of ignition can be identified (Fatehi, 2019): Homogeneous, heterogeneous, or hetero-homogeneous ignition. The ignition temperature and delay time are key parameters in studying solid combustion. Among the multiple methods presented in the literature to determine the ignition time of powder combustion, those based on optical diagnostic techniques are the most widely used. Molina and Shaddix (Molina and Shaddix, 2007) studied the combustion of bituminous coal diluted particle jets in an entrained flow reactor. They defined the start of the ignition based on the CH\* chemiluminescence emissions. In contrast, Khatami et al. (Khatami, Stivers and Levendis, 2012) and Riaza et al. (Riaza *et al.*, 2017) studied the ignition delay time of coal particles in a drop tube reactor under different conditions. They defined the ignition delay time as the time between the initial entrance of a particle in the reactor and the first appearance of luminous combustion.

The following study is based on this definition to determine the ignition delay times of powder. The effects of solid biomass on gas mixtures are studied qualitatively using cornstarch particles whose chemical formula is similar to cellulose. However, prior to studying the impact of these particles on mixtures 1 and 2, preliminary tests are conducted on the combustion of particles in an inert mixture. Hence, cornstarch particles were injected into an inert gas mixture of  $Ar/O_2$  at the end of compression temperature of 955 k and 1050 K. The mixture's temperature in the chamber was controlled by changing the proportion of Argon to oxygen.

The conditions of both tests are presented in Table 2.

Table 2: Experimental conditions of inert mixtures

End of compression temperature Tc (K)	Ar (Volume %)	O <sub>2</sub> (Volume %)	Injection pressure (bar)
Tc=955 K	43	57	20
T <sub>c</sub> =1050 K	57	43	20

#### 3 **RESULTS**

#### 3.1 Solid combustion in inert mixture

The combustion of particles was first studied by injecting cornstarch into inert mixtures of 955 and 1050 K. The injection tests were applied tens of times to validate the reproducibility of the combustion process. Figure 3 represents the results of only two tests for each temperature. Images in the first columns correspond to the start of injection, and those in the second columns to the appearance of the first luminosity (particle ignition).

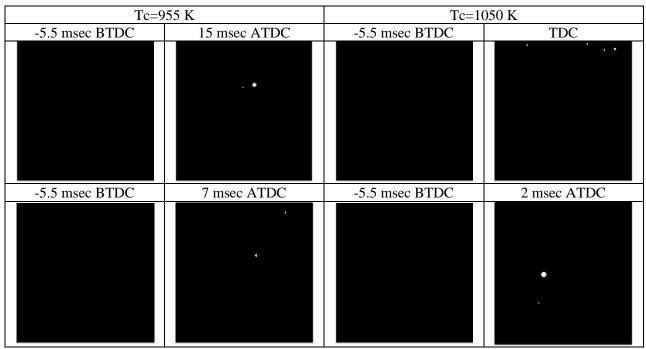


Figure 3: Combustion images of particles in hot inert mixtures (955 and 1050 K); the luminosity corresponds to the ignition of cornstarch

For all these tests, particles were injected -5,5 msec BTDC. The location of the first burnt particles is changeable from one test to another due to the continuous change in the internal aerodynamics caused by the flow movement. The particles can also be dependent on the temperature distribution inside the combustion chamber.

For inert mixtures compressed to 955 K, the first luminosity appears between 17 msec ATDC to 29 msec ATDC. This corresponds to an ignition delay time ranging from 22.5 msec to 34.5 msec. On the other hand, for inert mixtures compressed to 1055 K, the combustion process of solid cornstarch is more reproducible with ignition delay times ranging from 5 to 8 msec.

The lower reproducibility at lower temperatures is majorly due to heat loss at the enclosure wall due to higher residence time.

The results presented in this section prove that injected particles ignite at temperatures 955 K and 1050 K. Therefore, they will ignite in the experimental conditions of reactive mixtures 1 and 2. They also highlight the significant effect of higher temperatures improving the reproducibility of the combustion process and decreasing the ignition delay times (Fatehi, 2019).

#### 3.2 Solid combustion in reactive biogas enriched with hydrogen mixtures

The effects of direct injection of cornstarch on the combustion of mixtures 1 and 2 were performed more than ten times to verify the reproducibility of the combustion process. The evolution of the pressure curve for these tests is presented in Figure 4 A and B.

The shaded region represents tests with particle injection, and the solid line represents the one without injection.

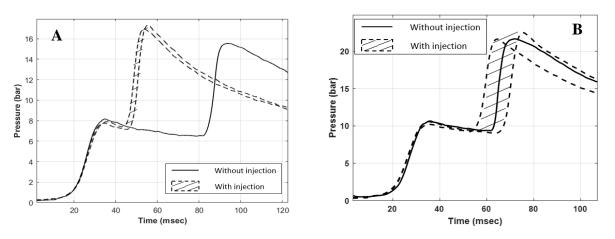


Figure 4 : Pressure curve of the combustion of A) mixture 1, B) mixture 2. Shaded region represents tests with particle injection and the solid line represents the one without particle injection

It is shown that for mixture 1, the particle injection reduces the ignition delay times significantly from 51 ms in gas combustion to less than 15 ms in gas/solid combustion. However, the particle injection in mixture 2 seems not to affect the ignition delay times of the initial gas mixture. Whereas, it is shown that the ignition delay times of solid/gas mixture and gas mixture are in the same range. However, despite being compressed to the same temperature (990 K), the particles ignite in mixture 1, about 8-19 ms earlier than in mixture 2. This is mainly due to the higher heat capacity of mixture 2 caused by a higher amount of carbon dioxide. This will cause a decrease in the thermal diffusivity of the mixture; the ignition delay of solid particles decreases. Several authors have elucidated this effect of heat capacity of the ignition of the solid particle. Zou et al. (Zou *et al.*, 2015) studied the ignition of a single particle in a drop tube reactor and stated that the higher thermal capacity could reduce the rate of temperature increase of the particle, resulting in a delay of the ignition.

This result is highly important as it highlights the dependency of solid ignition on the thermodynamic and transport properties of the reactive gas mixture.

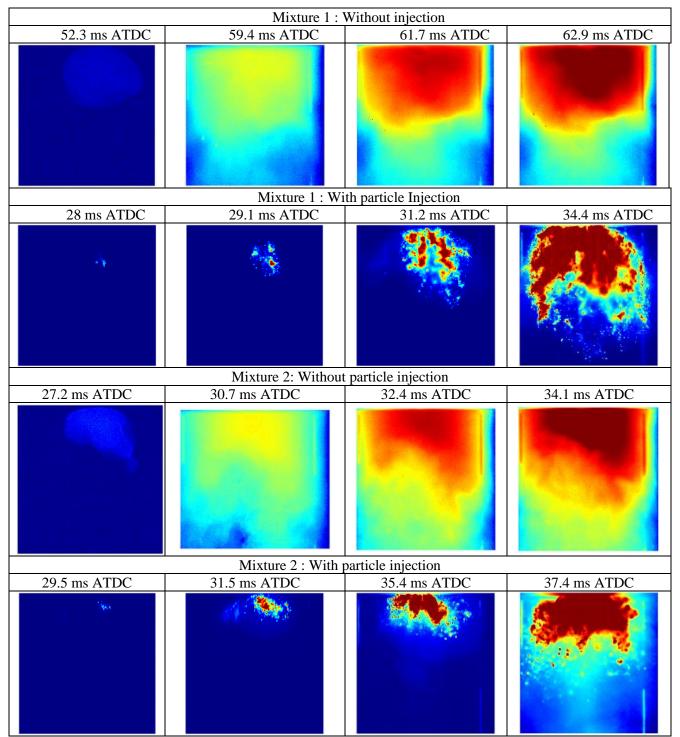


Figure 5: Combustion images of mixtures 1 and 2 with and without particle injection

In order to better understand the pressure curves analysis and to check whether the particles or the reactive gas initiate the ignition, the combustion behavior of the studied tests is presented in Figure 5.

Regarding the reproducibility of the combustion tests, only one injection test is presented for each mixture, along with the test without particle injection. For mixtures without injection, the combustion appears to be consistent with the instantaneous auto-ignition of a specific load that spreads quickly to cover only the upper part of the chamber. Following particle injection -5.5 msec BTDC, the first ignition site is initiated by a particle combustion a few milliseconds after the start of injection. The produced energy and the thermodynamic conditions in the chamber were sufficient to trigger the combustion of particles at the exit of the nozzle. The heat released from the combustion of the concentrated particles in the upper part of the chamber accelerates the chemical reactions in the gas combustion. Several sites appear around the initiation nucleus, and the multi-sites combustion propagates mainly in the upper part of the combust.

In other words, it seems that the solid particles initiate the ignition process inducing a change in the combustion behavior for all tests. Without injection, the combustion was shown to ignite and propagate in the upper part of the combustion chamber, whereas, with particle injection, multi-site combustion is favored for all the injection tests. The main difference occurs in the particle distribution in the combustion chamber, so the way the combustion propagates.

Two configurations are possible to explain this promoting effect. The first supposition, the combustion is initiated by the burn of either the volatiles (main composition  $CO_2$ ,  $CH_4$ , and CO) or the char with oxygen. Both of which are produced by the heating, drying, and devolatilization of the injected particles. The combustion, being exothermic, will supply the gas mixture with sufficient activation energy for the chemical reactions to occur. The second supposition, upon production of the volatiles, the gas mixtures' composition change, creating locally rich mixtures. these regions will activate the chemical reactions and provoke the combustion of the mixture.

#### 4 CONCLUSION

This study highlights the effects of direct injection of biomass powder into the lean homogeneous combustion of a biogas/H<sub>2</sub> mixtures. It proves that the direct injection of solid particles into reactive gas mixtures is a promising technology that can control the ignition timing of lean homogeneous combustion. The results were satisfyingly reproducible, which shed light on particles' capability in controlling the ignition timing. However, an optimization of the characteristics of the biomass powder and the biogas/syngas mixtures is essential for improving the combustion process. Developing a multi-zones model that characterizes biomass powder's combustion can be used to detect the ignition delay time of particles for different initial conditions. It would also be interesting to enhance further the theoretical model by including gas-phase reaction effects on the particle boundary layer. The application of other visualization techniques, such as Schlieren or Shadowgraphy, to observe the volatile release near the particle surface and its influence on the surrounding flow of hot gases would be a very interesting study, particularly for high volatile biomass. Furthermore, a parametrical study on the effects of the injection pressure, particle diameter, and fuel composition can be performed to understand under which conditions the two combustion modes appear.

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