Plasma Chem Plasma Process (2018) 38:1115–1132
https://doi.org/10.1007/s11090-018-9903-5

ORIGINAL PAPER

A New Spark Plug to Improve the Performances of Combustion Engines: Study and Analysis of Unburned Exhaust Gases

Dragos Astanei[1,2] · François Faubert[2] · Stéphane Pellerin[2] · Bogdan Hnatiuc[3] · Maxime Wartel[2]

Received: 27 October 2017 / Accepted: 30 April 2018 / Published online: 8 May 2018

Abstract The ignition sparks provided by the conventional spark plug do not always ensure a fast and complete combustion of the hydrocarbon-air mixture. For this reason, we offer a new type of plug with a double spark using two simultaneous discharges generated by a pulsed high voltage-power supply. This work presents the comparison of two spark plugs, a classical one and a double spark plug, by analyzing the unburned hydrocarbon gases from the exhaust pipe of the engine. For a first gasoline engine, we measure the oxygen concentration in the exhaust gases with a lambda probe and the unburned hydrocarbon by the use of GC–MS coupled with SPME extraction technique. We can observe a clear decrease of total unburned hydrocarbon (THC). For a second motor test bench, powered with propane, we complete measures in function of air/fuel ratio of the THC, NO_x, CO_2 and CO. These results confirm that we obtained a better combustion especially for leaner mixtures.

Keywords GC–MS · SPME · Unburned exhaust gases · THC, CO, CO_2, NO_x · Ignition improvement · Air/fuel ratio

✉ Dragos Astanei
dastanei@tuiasi.ro

✉ Stéphane Pellerin
stephane.pellerin@univ-orleans.fr

[1] Faculty of Electrical Engineering, “Gheorghe Asachi” Technical University of Iasi, Iasi, Romania

[2] GREMI, UMR 7344, Université d’Orléans/CNRS, 63 Avenue de Lattre de Tassigny, 18020 Bourges Cedex, France

[3] Laboratory Engineering Sciences in Electrical Field, Constanta Maritime University, Constanta, Romania

Introduction

For several years, some studies have shown that the capacity to deliver fossil energy could be limited due to both limited production capacity and lack of infrastructure, but also because fossil fuel resources could decline, while consumption increases [1]. This leads to regular price increases while the associated combustion produces a large amount of carbon dioxide responsible for global warming and other pollutants in a small quantity due to the use of catalytic exhaust. The awareness of these two major problems has determined the authorities to impose rules limiting toxic emissions related to the combustion and to promote the decrease of fuel consumption for all types of vehicles. The generalization of catalytic converters in the automotive sector has made progress in terms of pollutant emissions, but several problems remain: limited efficiency as long as sufficient temperatures are not reached, major pollutants such as N_2O and CO_2 that are not treated, new pollution by heavy metals… And moreover, there are many areas in which the engines remain uncatalysed: this is the case for example gardening, power boating etc. It is therefore important to work also upstream of the catalytic converter, and researchers need to find new techniques to directly improve combustion with low pollutants emission and a reduction in fuel consumption. For the automotive sector, solutions are related to the utilization of lean air/fuel mixtures or performing the combustion at higher pressure. In all these cases, the igniting conditions of the air/fuel mixture are more difficult. The possibility of replacing the rich air/fuel mixture with a lean one can ensure a significant reduction of the quantity of harmful emissions such as unburned hydrocarbons (HC), carbon monoxide (CO), nitrogen oxides (NO_x), associated with lower fuel consumption [2]. However, a significant amount of energy, higher than the one furnished by the conventional (spark) discharge is required [3, 4], in order to ensure a better quality of combustion. The discharge should have the largest possible volume to transfer a maximum of energy to the mixture. The best-known device used for this purpose is the spark plug, which shows its limits in the operating conditions of lean air/fuel mixtures.

In this context, we designed and manufactured a plasma based ignition system for Internal Combustion Engine (ICE) that increases the volume of plasma and the discharge electrical power, so as to encourage the speed and the quality of the combustion initiation. The new spark plug consists of a double spark system, with three electrodes, which produces two quasi-simultaneous sparks generated by a pulsed high voltage-power supply. For this purpose, we used this modified double spark plug [5] to ignite a four-stroke engine. The power supply is a micro-system based on AT89S52 microcontroller, which provides pulses with adjustable duration (from 0.5 to 3.5 ms) [6].

The purpose of this study is to compare two spark plugs, a conventional one and the double spark plug, by analyzing the unburned hydrocarbon gases from the exhaust pipe of the engine. We have used a teaching bench (Deltalab) engine equipped with a lambda probe to measure the oxygen concentration in the exhaust gases. Solid-phase microextraction (SPME) withdrew the gases and the analysis being made by gas chromatography coupled with mass spectrometry (GC–MS). This technique [7–10] is increasingly used in key areas of chemistry but is also successfully used to determine the composition and the quantity of the unburned hydrocarbons from the ICE exhaust gases. When the chromatogram is well resolved (good separation between two consecutive peaks) the mass spectra obtained for a peak allows identifying the molecule by means of a database. Generally, the SPME does not allow a total extraction of molecules but it can be reasonably used in the cases of a semi quantitative analysis and to compare the performances of two devices. Thus, a semi quantitative analysis of some target analytes is implemented to compare the efficiency of the two spark plugs for different ignition conditions. To complete this study, we determine the total unburned hydrocarbons by analysis with a flame ionization detector (FID) as well as the NO_x, CO_2 and CO on a realistic motor test bench equipped with ICE used in passenger cars, powered with air/propane mixtures, also by studying the influence of the equivalence ratio.

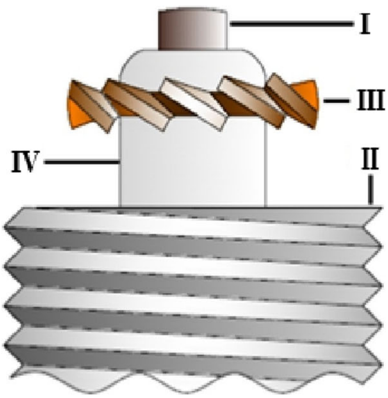
Experimental Set-Up

Double Spark Plug

We have thought about and studied a new ignition system which can speed-up and improve the combustion which can be adapted to all types of ICE without requiring any other modifications [11]. It is composed of a pulsed high-voltage-power supply and, as a main element, a modified spark plug containing three electrodes, named "double spark plug" (Fig. 1). The system can produce two quasi-simultaneous sparks increasing the amount of plasma produced in order to facilitate the ignition of the air/gasoline mixture. The delay between the breakdown in air of the first and the second discharge is under 240 ns at a 10 bars pressure, while at atmospheric pressure this delay between the two discharges is close to zero [5].

The double spark plug was built from a classical spark plug (Fig. 2) by slicing a part from the ground electrode *II* (the thread), cutting a small channel into the ceramic insulator, *IV*, and placing into it a washer shaped electrode, *III*, which is not powered. The geometry thus obtained ensures the existence of two quasi-simultaneous electrical sparks having a cumulated volume of plasma of up to over 2 mm^3 (more than double than the volume of plasma produced by a classical spark-plug), increasing the surface interaction with the mixture, using the same amount of electrical energy that used to supply a discharge produced with a conventional ignition system [11].

The double-discharge is thus produced in a vertical direction, as a spark generated by a conventional spark plug, but the total length of the discharges is greater in the case of the double spark plug.



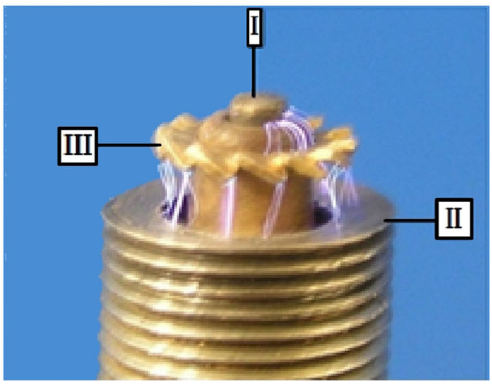


Fig. 1 Schematic diagram and photo of the double spark plug

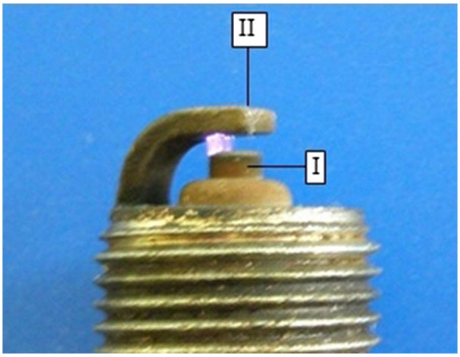


Fig. 2 Photo of a classic spark plug

Deltalab EX1000 Engine Bench Tests

The DELTALAB EX1000 engine bench is a multi-disciplinary teaching bench that allows a complete study of a single cylinder gasoline engine with fuel injection and electronic ignition. The main components of the bench are single cylinder ICE (HONDA GX31 type), microprocessor controlled direct fuel injection, DC electric motor/generator and dynamic load adjustment system. A central computer using the characteristics, loaded by the user, controls the electronic ignition and injection systems. The timing signal for the two systems (ignition and injection) is supplied by an inductive sensor, which is mounted in the engine's cylinder head and indicates the moment when the piston reaches the top dead center (TDC) and the intake valve is opened. The signal given by the sensor is analyzed by the software, which then provides the command to the ignition coil and to the injector.

The gas temperature at the exhaust output is measured using a thermocouple positioned in the holder's place and is between 50 and 70 °C (see Fig. 3b). In order to measure the equivalence ratio and obtain a first combustion evaluation starting from the oxygen concentration in the exhaust gases, a ROTRONICS CMR101 air/fuel acquisition system has been used. The system is connected to a wide band oxygen sensor (Lambda probe) which measures the oxygen percentage in the exhaust gases of the ICE. Based on the measured oxygen percentage the device can compute the following parameters: the equivalence ratio, the air/fuel ratio depending on the used fuel, and lambda (λ) parameter, according to statistical accuracy of a few percents.

Lambda (λ) is a dimensionless parameter that indicates the excess or the lack of oxygen from an air/fuel mixture related to the stoichiometric value. For a stoichiometric air/fuel mixture (the equivalence ratio $\Phi = 1$), $\lambda = 1$. For $\lambda = 1.05$ then the mixture is 5% lean ($\Phi = 0.95$) while for $\lambda = 0.95$ the mixture is 5% rich ($\Phi = 1.05$). There is a close relation between the lambda parameter and the equivalence ratio of the air/fuel mixture given by the equation:

$$\lambda = \frac{1}{\phi} = \frac{m_{ae}}{m_{fe} \cdot r} \tag{1}$$

where m_{fe} and m_{ae} are respectively the masses of fuel and air effectively used and r is the stoichiometric ratio. Otherwise, considering that the quantities of fuel injected as well as the air absorbed into the combustion chamber are constant during a set of experiments, a lower value of λ means a more complete combustion of the hydrocarbons. More oxygen

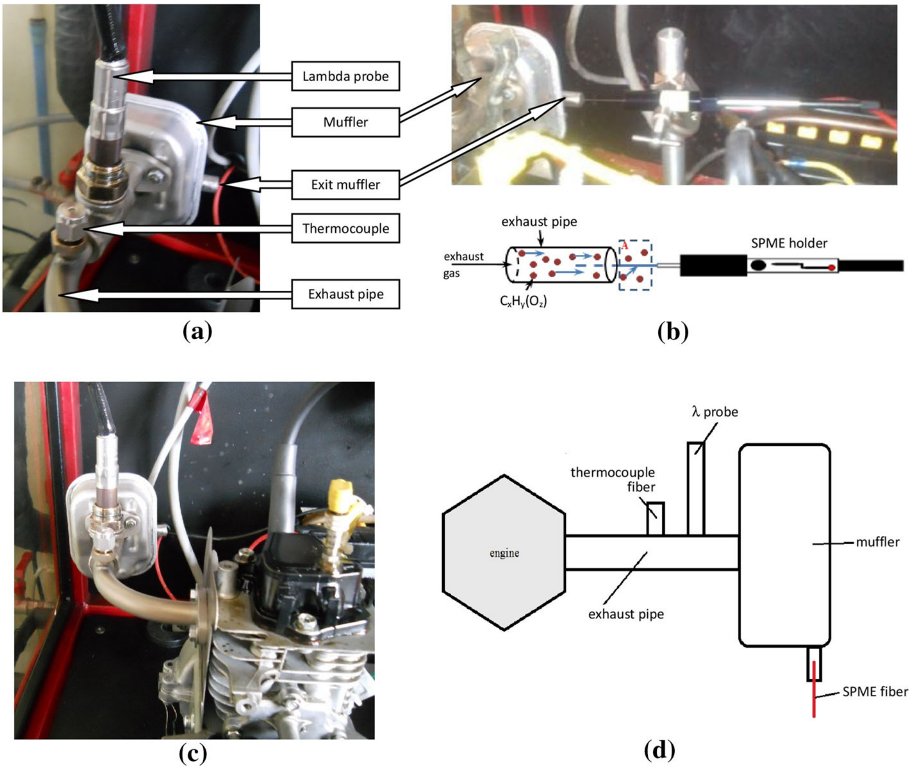


Fig. 3 Positioning of the exhaust gases analyzers. **a** Lambda probe and thermocouple, **b** SPME fiber, **c** general view of the engine stand and **d** global positioning of the sensors

molecules are reacting with the hydrocarbons during the combustion, which means there is a smaller quantity of oxygen which can be retrieved in the exhaust gases.

The high-voltage-power supply consists of an ignition coil and a designed control source based on a microcontroller from the ATMEL family, AT89S52 [6, 11]. Two electrical signals given by two analog potentiometers are converted into digital signals through a MCP3202 dual analog–digital convertor, to be used by the microcontroller to form the pulses. Using these two controllers the width or the phase of the control pulses can be modified, and the type of the control signal can be changed by modifying the software loaded into the microcontroller's memory. For this study we have used two signal control *a_1.5* and *a_3.0* with a single pulse during respectively 1.5 and 3.0 ms (Table 1), that we can compare to the conventional ignition system, called *stand*, Doing so, we fitted the engine stand used for testing and that can provide a single control pulse for each combustion cycle having a width of about 1.5 ms [11, 12]. The energy corresponding to *a_1.5* signal is around 35 mJ and for *a_3.0* signal is close to 100 mJ for both tested spark plugs.

As it can be observed in the table above and taking into account the statistical measurement errors on more than 50 cycles (less than 10%), there is almost no difference concerning the electrical energy for the discharges produced using the classical and the double spark plug. In fact, the electrical energy is strongly dependent on the energy accumulation period thus on the control pulses width [6, 13].

Table 1 The energy and the lifetime of a spark produced inside an ICE cylinder in an air/gasoline mixture, in the case of the two control signal used

	Pulse width (ms)	Discharge energy (mJ)		Discharge lifetime (ms)	
		Classical spark plug	Double spark plug	Classical spark plug	Double spark plug
Stand	1.5	$36^{\pm 5}$	$35^{\pm 5}$	$0.30^{\pm 0.05}$	$0.27^{\pm 0.04}$
a_1.5	1.5	$40^{\pm 4}$	$37^{\pm 3}$	$0.38^{\pm 0.04}$	$0.25^{\pm 0.02}$
a_3.0	3.0	$96^{\pm 10}$	$108^{\pm 8}$	$1.65^{\pm 0.17}$	$0.75^{\pm 0.05}$

During the exhaust gases analysis process (SPME sampling, only done on this engine—see below), the four engine's parameters that can influence the amount of hydrocarbons from the exhaust gases have been maintained constant: air/fuel equivalent ratio, engine temperature, engine speed and load. The air regulator (throttle) was placed in fixed position and the fuel consumption was fixed to 2.2 ml/min in order to have an equivalence ratio in the combustion chamber as close as possible to the stoichiometry. The engine load was fixed in a position for an engine speed of 2000 rpm.

EP6 Engine Bench Tests

The proposed ignition system based on the double spark plug was also equipped on an ICE used in passenger cars—EP6 (PSA BMW) type. The results obtained when using the M10 type double spark have been compared with the ones corresponding to a new generation of conventional spark plugs—NGK PLZKBR 7 A-G type—a type which has platinum electrodes. The complete description of the engine bench can be found in [12, 14–16]. We describe here its main characteristics only.

The engine used for the tests is a four-cylinder turbocharged spark ignition ICE with direct injection characterized by four valves (two for intake and two for exhaust) for each cylinder with a total displaced volume of 1598 cm^3 and a compression ratio of 10.5. For the experiments the engine was transformed into a one-cylinder engine by stopping the fuel alimentation for the other three cylinders (V_d = 399.5 cm^3). The engine bore and stroke are respectively 77 and 85.8 mm. When all the four cylinders operate the engine can provide a maximum rated power of 110 kW at 5800 rpm and a torque of 240 Nm at 1400 rpm. The fuel used was the propane C_3H_8 (stoichiometry ratio—15.67 kg air for 1 kg of propane) which has a similar behavior in the combustion chamber as the gasoline but it does not contain impurities that could deteriorate the measurement equipment.

The engine was driven by an electric motor with a constant speed of 2000 rpm with a variation of less than 0.5%, the speed being measured with an optical encoder mounted on the crankshaft giving a resolution of 0.1 crank angle degrees. The quantities of both air and propane injected into the engine were measured and controlled with Bronkhorst Coriolis high accuracy mass flow meters: 300 NL/min ± 0.7% for air and 100 NL/min ± 1% for propane. In order to obtain a homogeneous air/fuel mixture the gases were premixed into an intake plenum. The air fuel mixture was kept at a constant temperature of 30 °C using a heater. The engine cooling water and oil have a temperature of 80 °C during the tests.

The conducted tests have already allowed the determination of engine mechanical parameters (the indicated mean effective pressure *IMEP*, the coefficient of variation of the indicated mean effective pressure COV_{IMEP}, the in-cylinder pressure peak position *PPP*, the coefficient of variance of the maximum pressure peak position COV_{PPP}, the flame development angle for 10 and 50% of fuel mass burned) [12], and we now consider analyzing the exhaust-gas composition (NO_x, HC, CO, CO_2, O_2, total unburned hydrocarbons—THC). For these tests, exhaust samples were drawn through a heated sample line (temperature set to 190 °C), and emission measurements were carried out with a classical Horiba emission analyzer bay referred as Mexa-7100HEGR, that was calibrated regularly before conducting the experiments [17, 18]. It has an accuracy of less than 2% of the instantaneous measured values and 1% for the full range. In this multi-channel analyzer, the CO and CO_2 concentrations have been determined by using non-dispersive infrared method, NO_x by chemiluminescent method, O_2 by polarization mode dispersion measuring technique and HC by heated FID method.

During the tests, the *IMEP* was set to 4.5 bar and the control pulse used to command the ignition coil was of type *a* (one pulse for each cycle). All the considered parameters have been measured for varying the equivalence ratio from stoichiometry values ($\Phi = 1.0$) to very lean mixtures $\Phi = 0.6$. The equivalence ratio has been adjusted by modifying the quantities of air and propane from the mixture, while the engine was alternatively equipped with the double spark plug and with the conventional spark plug without performing any changes to the ignition parameters.

The diluted mixtures (leaner mixtures) are characterized by a reduced combustion speed increasing the time between the spark ignition and peak pressure point. Also highly diluted mixtures are difficult to be ignited. That is why the spark angle has been adjusted for different mixture equivalence ratios, in order to maximize the brake torque [14, 19]. For this specific engine the maximum brake torque is obtained for a *PPP* in the range of 15° after TDC. The spark angle was considered the same for the two ignition systems (double spark plug and conventional spark plug) in order to evaluate for which system the combustion speed is higher [11].

Solid-Phase Microextraction

For recovering the hydrocarbons molecules from the exhaust gases an extraction technique called solid-phase microextraction (SPME) has been used, in which the organic molecules (unburned hydrocarbons) are adsorbed onto a fused-silica fiber covered with a polymer coating. The SPME holder has been precisely positioned in front of the ICE exhaust pipe in axial direction, with the exposed fiber placed in the middle of the exit muffler (Fig. 3b). The SPME fiber is exposed completely outside the needle for grab time-weighted average sampling. Then a replaceable Teflon© cap is used to seal the needle to preserve sample integrity between the test and the analysis. Teflon© is a good material for sealing the fiber needle, has little memory effect, and can be used repeatedly [20].

Such SPME [21–23] sampling method was chosen for the tests due to its capability to generate chromatograms easier to integrate than the ones produced by classical methods such as charcoal tubes method.

To adsorb/absorb the organic molecules from the exhaust gases two different SPME fibers have been used [23, 24]:

- The first one is coated with a 75 μm layer of Carboxen/Polydimethylsiloxane; it has the capability to adsorb molecules with lower molecular weight, in the range from 30 to 200 g/mol. The maximum fiber working temperature is 250 °C.

- The second SPME fiber used is coated with a 100 μm layer of Polydimethylsiloxane (PDMS). It can absorb organic compounds with a molecular weight in the range from 75 to 320 g/mol. The maximum working fiber temperature is 280 °C.

Preliminary tests done by increasing by step of 1 min the exposure sorption times before acquiring chromatogram and comparison of consecutive global amplitudes of similar peaks, revealed that PDMS fiber reaches its equilibrium/saturation limit in 6 min of absorption, a much faster saturation for Carboxen fiber, in around 3 min of adsorption. As a consequence, the chosen sampling times were respectively 4 min for the PDMS fiber and only 2 min for the Carboxen fiber.

All experiments were done after a 5-min of engine running to ensure a warm operation, while verifying that in each case, the temperature of the gases at the cylinder exit, as measured by an external thermocouple, becomes almost constant. This temperature reaches 300–400 °C according to working conditions: there is no important difference between the two spark plugs, but the gas temperature tends to increase with the width of the control pulses [11].

The SPME fiber is located at the exit of silencer exhaust (Fig. 3): it is the easiest location to avoid modifying the experimental setup. In this condition, the fiber exposure temperature, which is checked using external thermocouple between 50 and 70 °C, is low enough to allow a good equilibrium in the adsorption process, and is always far lower than the maximum fiber working temperature.

To make sure SPME sampling reproduces well, we have maintained constant extraction and desorption times, parameters that can also influence the amount of hydrocarbons from the exhaust gases. The same applies to the period between two consecutive experiments.

Gas Phase Chromatography/Mass Spectrometry Analysis

The chromatography analysis was carried out using an Agilent Technologies Gas Chromatograph–Mass Spectrometer (GC–MS) system (6850 Network GC system coupled with 5973 Network Mass Selective Detector), with a column HP5 MS, 30 m × 0.25 μm and an internal diameter of 530 μm. The carrier gas used is helium at an initial flow rate of 1 ml/min. After the adsorption process, the fiber was manually placed into the GC split/splitless injector for 5 min at 250 °C in splitless mode to allow the complete desorption. With the aim of blocking molecules at the head of column, the GC oven is kept for 4 min at the initial temperature of 40 °C and is further gradually heated up to 300 °C with a gradient of 10 °C/min. After they are separated in the column, the molecules reach the mass spectrometer where they can be identified their mass spectra. The GC–MS system used in this study can detect molecules between the limits of their molecular weight from 10 to 250 a.m.u. (atomic mass unit). Although a too weak MS window opening shows unnecessary elements such as moisture, oxygen, nitrogen, the resulting interferences observed in the first minutes of the chromatograms (see Fig. 4) do not mask the molecules studied.

At least four tests have been carried out for each type of spark plug, control signal and SPME fiber used. The values obtained represent an average of the results for each test, this way reducing the experimental errors. The standard deviations determined for six consecutives chromatograms corresponding to the use of the classical spark plug for a discharge energy of 35 mJ for each combustion cycle—corresponding to an 1.5 ms control signal—has values within reasonable limits: less than 12% if the ICE working parameters are kept steady (cf Fig. 4). The period of 5 min chosen for molecules desorption during which the fibers are placed in the GC injector proved to be sufficient so that all the

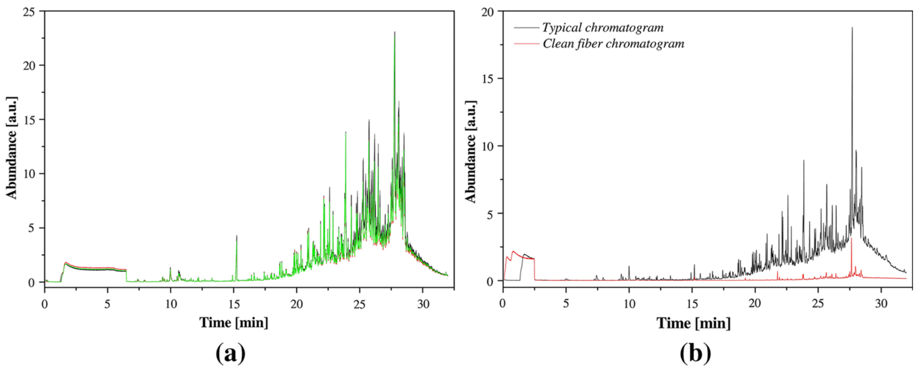


Fig. 4 Typical chromatograms obtained for using the PDMS fiber. **a** Four chromatograms obtained for the same engine operation conditions; **b** highlighting the residual molecules from the fiber after a 5 min desorption time

molecules are almost completely released from the fiber. For this reason, the fiber can be successfully reused for following tests without influencing the results.

Results and Discussion

Lambda Probe

A first assessment of double spark plug performances regarding the combustion is carried out by comparing the values of the exhaust gas parameters (see Fig. 5) with the ones obtained when using a conventional spark plug. The λ parameter is considered as a function of the engine load for the control signal obtain when the ignition coil is triggered with control pulses which have a width of 1.5 ms and an amplitude of 5 V. During the tests

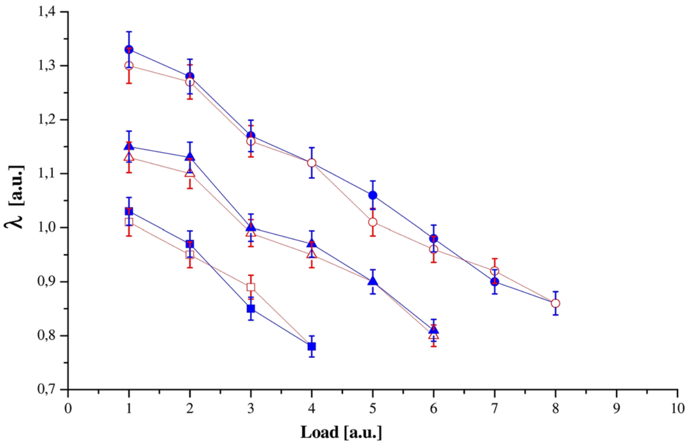


Fig. 5 Variation of the λ parameter versus engine load. *a_1.5* control signal—throttle positions (see text): filled circle, open circle 4; filled triangle, open triangle 5; filled square, open square 6. The full symbols filled circle, filled triangle, filled square are for the double spark plug; the empty ones open circle, open triangle, open square for the conventional spark plug

the gasoline consumption is 2.2 ml/min and the engine speed around 2000 rpm, with a variation of less than 5%. Considering the fact, the quantity of injected fuel is fixed according to the characteristics loaded into the software, the equivalence ratio of the feeding gas can only be adjusted by modifying the throttle position. The λ parameter is measured for three different positions of the air regulator (throttle), denoted *throttle* 4, 5, and 6 (from richer 4, to leaner mixture 6), using both the double and the conventional spark plugs and also controlling the combustion ignition by using the bench system. The results presented in Fig. 5, show that the spark plug type does not influence the combustion in an obvious manner when the ignition is triggered using a conventional ignition system through which the control pulse width determines a spark energy around 35 mJ.

A comparison between two control signals having a width of respectively 1.5 and 3.0 ms, is performed by keeping the same ignition angle. The results we obtained, are shown in Fig. 6.

A significant improvement of the combustion quality occurs when a higher amount of energy is delivered to the discharge obtained by using a wider control pulse. The difference is even more visible for no load regime and for very loaded regime—engine regimes which involve stability in operation problems. The combustion improvement is more noticeable when using the double spark plug [25].

Evolution of the Unburned Gases Studied by SPME/GC–MS

Figure 7a, b show typical examples of chromatograms obtained both for the Carboxen and the PDMS fibers. It has been obtained when using the double spark plug and a control signal that provide pulses with a width of 1.5 ms. On the chromatograms, each peak corresponds to an unburned molecule adsorbed from the exhaust gases and the peak area is proportional to the quantity of matter. As it can be observed in the figure below, the Carboxen fiber is more appropriate to adsorb compounds with a lower molecular weight. For this reason, it is used to compare the quantity of the molecules that appear in the first half of the chromatograms (smaller molecular weight molecules) while the PDMS fiber is

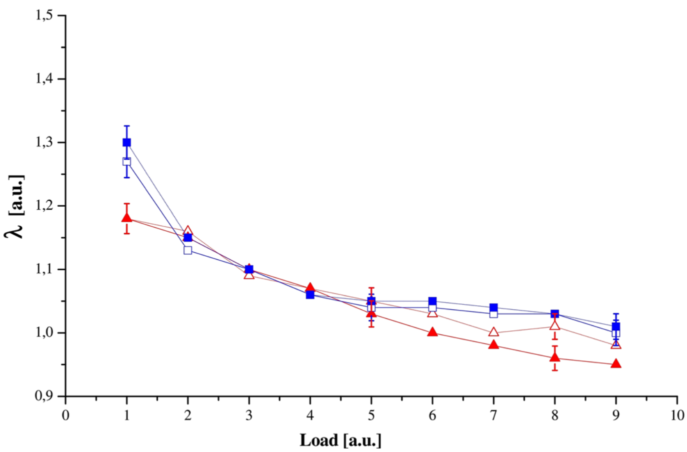


Fig. 6 Influence of the energy and in-cylinder pressure on the combustion for *a_1.5* control signal (filled square, open square), and *a_3.0* control signal (filled triangle, open triangle). The full symbols filled square, filled triangle are for the double spark plug, and the empty ones open square, open triangle for the conventional spark plug. Typical error bars are shown only on a few points to avoid overloading the graph

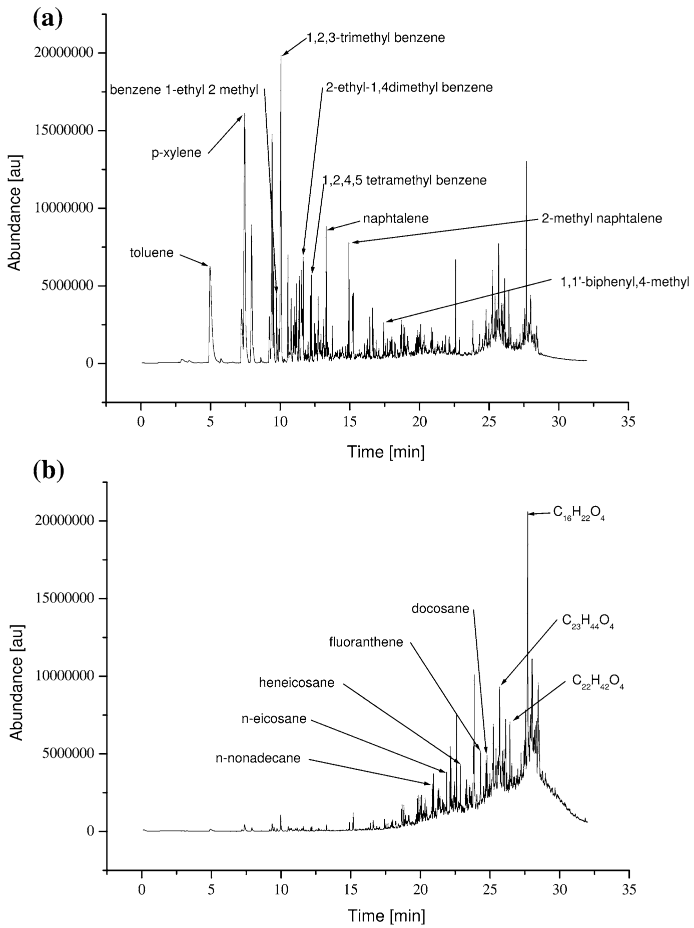


Fig. 7 **a** Typical chromatograms obtained with the use of Carboxen fibers (case of double spark plug, with *a_1.5* control signal). **b** Typical chromatograms obtained with the use of PDMS fibers (case of double spark plug, with *a_1.5* control signal)

considered for the molecules that appear in the second half of the chromatograms. In the middle part of each chromatogram, the same molecules are found for each extraction at the same retention time:

- For Carboxen fiber: 20.85 min—nonadecane; 20.95 min—9H-fluorene, 9 methylene; 22.60 min—dibutylphtalate.
- For PDMS fiber: 20.85 min—nonadecane; 20.96 min—9H-fluorene, 9 methylene; 22.62 min—dibutylphtalate.

Based on the resulting chromatograms more than 55 molecules have been identified from over 200 present. The identification is made taking the spectral match with NIST database [26] into account. Moreover, their match factor and reverse match factor are determined for each molecule (see Table 2). In most cases, the values of these factors are relatively close to 1000, which suggests that identification is very likely. They are on the

Table 2 The selected molecules

Family	Molecule	Chemical formula	CAS Number	Retention time (min)	Match factor	Reverse match factor	Type of fiber
Aromatics	Toluene	C_7H_8	108-88-3	4.93	930	930	Carboxen
	p-Xylene	C_8H_{10}	106-42-3	7.45	940	940	Carboxen
	Benzene 1-ethyl 2-methyl	C_9H_{14}	611-14-3	9.73	950	950	Carboxen
	1,2,3-trimethyl benzene	C_9H_{12}	526-73-8	10.03	900	920	Carboxen
	2-ethyl-1,4-dimethyl benzene	$C_{10}H_{14}$	1758-88-9	11.99	770	850	Carboxen
	1,2,4,5-tetramethyl benzene	$C_{10}H_{14}$	95-93-2	12.22	930	930	Carboxen
	Naphthalene	$C_{10}H_8$	91-20-3	13.29	890	950	Carboxen
	2-methyl naphthalene	$C_{11}H_{10}$	91-57-6	14.93	920	930	Carboxen
	1,1′-biphenyl, 4-methyl	$C_{13}H_{12}$	644-08-6	17.41	890	910	Carboxen
	Fluoranthene	$C_{16}H_{10}$	206-44-0	24.33	810	930	PDMS
Alkanes	n-nonadecane	$C_{19}H_{40}$	629-92-5	20.86	840	920	PDMS
	n-eicosane	$C_{20}H_{42}$	112-95-8	21.89	700	900	PDMS
	Heneicosane	$C_{21}H_{44}$	629-94-7	22.88	740	900	PDMS
	Docosane	$C_{22}H_{46}$	629-97-0	24.74	720	910	PDMS
Carboxylic acids	Oxalic acid, 2-ethylhexyl ester tridecyl	$C_{23}H_{44}O_4$	N/A	26.18	730	770	PDMS
	Hexanedioic acid, bis (2-ethylhexyl) ester	$C_{22}H_{42}O_4$	103-23-1	26.46	840	890	PDMS
	1,2-Benzene-dicarboxylic acid, mono (2-ethylhexyl) ester	$C_{16}H_{22}O_4$	4376-20-9	27.78	900	900	PDMS

other hand close to each other, which indicates the good estimate of the background subtraction.

The identified hydrocarbons and oxidized derivatives are commonly found into the ICE exhaust gases [6, 27–30]. The most common volatile organic compounds identified are:

1. Straight-chain, ramified and cycled alkanes (near quantification limit: nonane, decane, undecane, and in a quantifiable amount: nonadecane, docosane).
2. Hydrocarbons aromatics compounds (toluene, p-xylene, ethylbenzene, 1,2,3-trimethyl-benzene, naphthalene and fluoranthene).

3. Oxidized derivatives (carboxylic acids).

The purpose of this study is the assessment of the double spark plug performances by comparing it with a conventional spark plug for different control signals. What is not required is a quantitative analysis of the exhaust gases, with standard compounds representing the selected molecules. That is why only the molecule identification and the analysis of the difference in the peak area between different chromatograms (for different ignition parameters) are considered. The results obtained for the control signal *a_1.5* and *a_3.0* for the double spark plug are compared to the ones obtained when using the conventional ignition system equipped on the engine bench (classical spark plug, 1.5 ms, 35 mJ).

In order to compare the two ignition systems' efficiency, nine different molecules have been taken into consideration for the Carboxen fiber (low molecular weight), and eight ones for the PDMS fiber, see Table 2. The molecules have been chosen by taking into account their position on the chromatograms and especially a well-defined corresponding peak, important aspect in the integration process and identification. If the peak corresponding to a certain molecule overlaps another peak then huge errors could interfere in its surface determination. The molecules were especially chosen because they represent the major classes of molecules stemming from the engine combustion.

In order to evaluate the quantity of unburned hydrocarbons from the exhaust gases for the case of different control signals and the two spark plugs, based on the obtained chromatograms (considering the areas under peaks corresponding to the selected molecules, which is proportional with the quantity of a given molecule), a reduction factor, σ, has been considered:

$$\sigma = \left(1 - \frac{A_{s\tan d}}{A_{comp}}\right) \cdot 100\ [\%] \tag{2}$$

where A_{stand} is the peak area corresponding to the use of the classical spark plug (1.5 ms, 35 mJ) and A_{comp} is the peak area corresponding to the chromatogram to be compared (spark plug type, control signal *a_1.5* or *a_3.0*).

The results have shown that the amount of unburned hydrocarbons from the exhaust gases is considerably reduced when the ignition is triggered using the double spark plug (higher volume of plasma) and also when the energy supplied to plasma is more important (wider control signals), even when using the conventional spark plug. In Fig. 8, the most representative results are presented, obtained by comparing the double spark plug supplied with around 35 and 100 mJ (control signals *a_1.5* and *a_3.0*) with the conventional spark plug supplied with an energy of 35 mJ.

As can be seen in the figure, by replacing the classical spark plug with a modified one and using the same amount of electrical energy (35 mJ) we can easily obtain a quantitative decrease of some unburned hydrocarbon molecules. This effect can generally be enhanced by delivering a higher quantity of energy to the discharge (higher values of the plasma temperatures): taking the indicated statistical errors bars into account, this is particularly visible for the representative alkanes and carboxylic acids, but also for few aromatics (Naphthalene, 2-methyl naphthalene, 1,1′-biphenyl, 4-methyl and Fluoranthene). Almost no effect is observed on p-Xylene, Benzene 1-ethyl 2-methyl and 1,2,3-trimethyl benzene, whereas, the toluene quantity in the exhaust gases seems to increase when a higher amount of energy is delivered to the plasma.

It should be noted that the decrease of the unburned hydrocarbons quantity in the exhaust can also be achieved by supplying the conventional spark plug with more energy

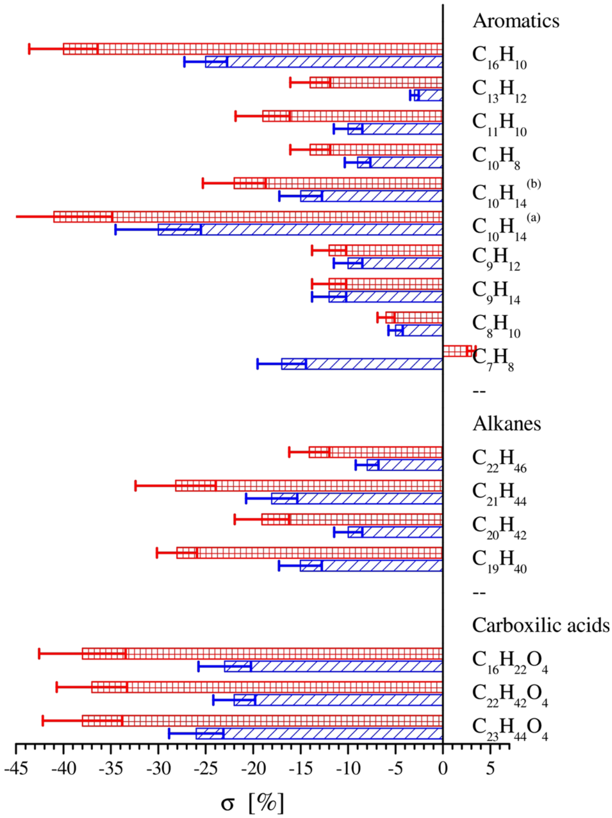


Fig. 8 The reduction factor, σ, for using the double spark plug supplied for three representatives class of molecules, with *a_1.5* (▨) and *a_3.0* (▦) control signals (resp. 35 and 100 mJ)

but in this case the differences are not as significant as in the case of the double spark plug [11].

Exhaust Gases (THC, CO_2, CO, NO_x) Studied with the PSA Engine

Figure 9 shows the results of the analysis obtained with the use of the PSA engine, and their statistical uncertainties evaluated from the accuracy of the Horiba emission analyzer and from the accumulation of measurements. They indicate that the THC emissions are lower for equivalence ratios close to stoichiometry and they increase for leaner mixtures, even if the quantity of injected fuel is smaller. Otherwise, the exhaust emissions analysis confirms the results obtained when applying the gas chromatography methods for the Honda GX31 engine: the measurements of the total unburned hydrocarbons (THC) proved that the double spark plug could ensure a better combustion (less THC) especially under difficult ignition conditions (lean mixture) as shown in Fig. 9. This decrease using double spark plug, is particularly beneficial for a better use and preservation of the catalytic converters conventional vehicles are equipped with. These catalytic converters, allow chemical reactions which tend to transform the most toxic constituents of the exhaust gases

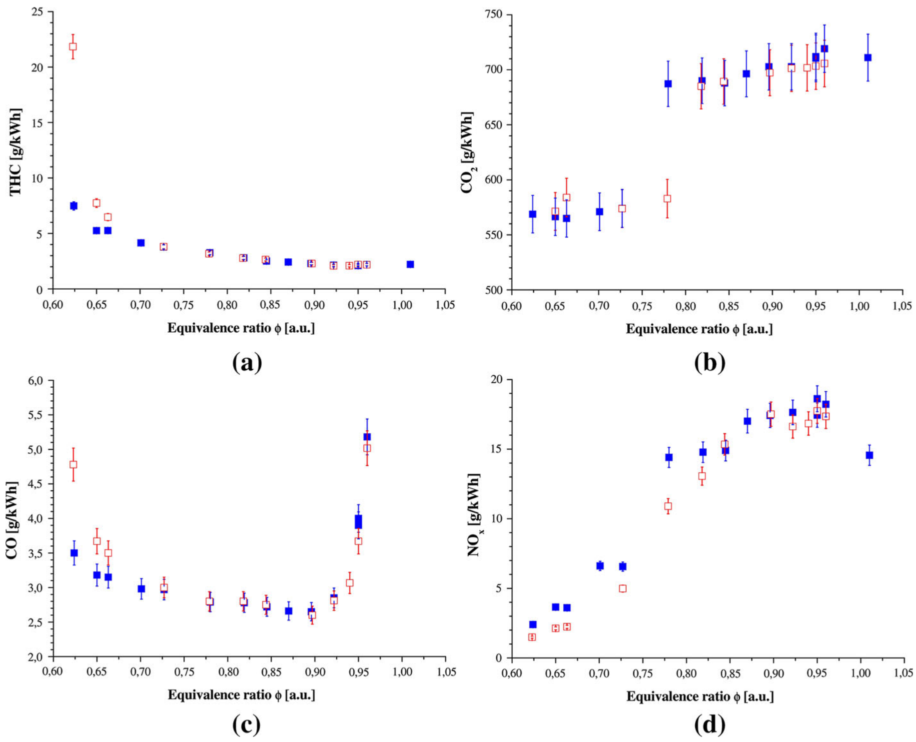


Fig. 9 Specific exhaust gas emissions versus equivalence ratio for double (filled square) and conventional (open square) spark plugs. **a** THC, **b** CO_2, **c** CO, **d** NO_x

(carbon monoxide, unburned hydrocarbons, nitrogen oxides) into less toxic elements (water, carbon dioxide and dinitrogen).

The CO_2 emissions decrease from around 720 g/kWh with the equivalence ratio up to around 560 g/kWh—leaner mixtures involve lower quantities of fuel. In CO_2 emissions evolution there is a step drop in the range of 0.77 equivalence ratio, decrease which can also be observed in NO_x evolution. There is a higher quantity of CO_2 obtained when the double spark plug is used for higher equivalence ratios while for lean mixture the CO_2 quantity seems to be more important for the classical spark plug.

The CO emission analysis have shown that the CO quantity from the exhaust can be reduced by operating with an equivalence ratio in the range of 0.9, values for which there is an emission of around 2.6 g/kWh. The CO quantity has the highest values for equivalence ratio close to 1 and very close to the engine operating limit: 0.6. As for the THC, the CO emissions are less important when using the double spark plug and when the mixture is very lean.

The NO_x emissions (plotted in Fig. 9) show a ten times decrease of the NO_x quantity (from 18 to less than 2 g/kWh) in the exhaust gases for the case of operating with lean mixtures as compared with mixtures close to stoichiometry. Comparing the two ignition systems shows that there is a slight increase in NO_x emissions for the double spark plug. This increase is most probably due to higher in-cylinder pressure obtained for the double spark plug which also involves higher flame temperatures, probably via the Zel'dovich mechanism (thermal NO formation) [31].

The differences obtained in this case (using the EP6 engine) are lower than in the case of the other test engine (Honda GX31) due to the engine construction and performances. The EP6 engine is designed to be used on passenger cars having low pollutant emissions while the GX31 is used for applications which do not require its use for long periods of time and for which standards regarding the emissions do not exist.

Conclusion

The tests on engines have proved that the double spark plug can be successfully used on ICEs without modifying the engine structure or the ignition system. The exhaust gas analysis using the lambda sensor revealed that there is less oxygen in the exhaust gases which means a more complete combustion (more oxygen is consumed reacting with hydrocarbons molecules) when the double spark plug is used, especially if the supplying energy is more important. The engine temperature evolution in time indicates that the combustion flame of the double spark reaches higher temperatures which probably involve higher NO_x emissions.

The chromatography tests have shown that the increased volume of plasma given by the double spark plug can ensure a better combustion—less target compounds representing unburned hydrocarbons in the exhaust gases. An increased effect of the modified ignition system could be achieved by providing more energy to the discharge. The SPME method provided chromatograms having repeatability within reasonable limits (less than 12% if the ICE working parameters are kept steady).

The conducted tests on the GX31 engine have also proved that using the double spark plug a faster combustion can be achieved; also the engine operation stability is upgraded. The tests conducted with the EP6 engine have confirmed the previous results obtained with the GX31 engine. The double spark plug ensures a better stability in operation as compared with a conventional spark plug especially for lean mixtures when the ignition conditions are more severe. It also guarantees a faster flame development.

The quantity of unburned hydrocarbons (THC) from the exhaust gases is reduced when the double spark plug is used as well as the carbon monoxide (CO), especially when using lean mixtures. However, the NO_x quantity is slightly increased, most probably due to higher in-cylinder pressure obtained for the double spark plug which also involves higher flame temperatures.

Generally speaking, the advantages offered by the proposed ignition system could be greatly improved, for example by using a higher supplying energy (wider control pulses). But it also allows to define control signal with two consecutive discharges for each combustion cycle [11]. Then, the misfires can be avoided: when the discharge corresponding to the first control pulse does not ignite (misfire) and the air/fuel mixture is ignited by the second discharge (commanded by the second control pulse). Even if in this case there are important losses due to late ignition the energy of the combustion cycle is not completely lost and fuel vapors are not evacuated into the exhaust gases where they can damage the lambda sensor/sensors or the catalytic converter.

Acknowledgements This work was a part of an international cotutelle agreement between "Gheorghe Asachi" Technical University of Iasi, Romania and Orléans University, France and financially supported by AUF (Agence Universitaire de la Francophonie). The authors would like thank Prof. Fabrice Foucher and Dr. Antonio Mariani, for giving the opportunity to test the proposed ignition system on engine bench in the PRISME Laboratory – Polytech Orleans. They also thank Mrs. Valérie Vasseur, native English speaker, for the proofreading of the manuscript.

References

1. Raj Singh B, Singh O (2012) Global trends of fossil fuel reserves and climate change in the 21st century. In: Shahriar Khan (ed) Fossil fuel and the environment. InTech. ISBN: 978-953-51-0277-9. https://www.intechopen.com/books/fossil-fuel-and-the-environment/global-trends-of-fossil-fuel-reserves-and-climate-change-in-the-21st-century
2. Docquier N, Cande S (2002) Combustion control and sensor: a review. Prog Energy Combust Sci 28:107–150
3. Glassman I (1996) Combustion, 3rd edn. Academic Press, New York
4. Dale JD, Checkel MD, Smy PR (1997) Application of high energy ignition systems to engines. Prog Energy Combust Sci 23:379–398
5. Hnatiuc B, Hnatiuc E, Pellerin S, Chapelle J (2006) Experimental analysis of a double spark ignition system. Czech J Phys 56(8):851–867
6. Hnatiuc B, Astanei D, Pellerin S, Cerqueira N, Hnatiuc M (2014) Diagnostic of plasma produced by a spark plug at atmospheric pressure: reduced electric field and rotational—vibrational temperatures. Contrib Plasma Phys J. https://doi.org/10.1002/ctpp.201300059
7. Sheng G, Goldberg J, Harigaya R, Ty J (2008) GS-MS determination of volatile organic compounds in gasoline and diesel emissions. Dartm Undergrad J Sci Chem 2006:47–53
8. Baimatova N, Koziel JA, Kenessov B (2017) Passive sampling and analysis of naphthalene in internal combustion engine exhaust with retracted solid phase microextraction device and GC–MS. Atmosphere 8-7:130. https://doi.org/10.3390/atmos8070130
9. Woolcock PJ, Koziel JA, Johnston PA, Brown RC, Broer KM (2015) Analysis of trace contaminants in hot gas streams using time-weighted average solid-phase microextraction: pilot-scale validation. Fuel 153:552–558
10. Woolcock PJ, Koziel JA, Cai L, Johnston PA, Brown RC (2013) Analysis of trace contaminants in hot gas streams using time-weighted average solid-phase microextraction: proof of concept. J Chromatogr A 1281:1–8
11. Astanei D (2014) Improving the performances of the combustion engines by improving the ignition system. Ph.D. Thesis, Technical University of Iasi (Romania)/University of Orléans (France)
12. Astanei D, Faubert F, Pellerin S, Hnatiuc B (2018) Evaluation of the efficiency of a double spark plug to improve the performances of combustion engines: pressure measurement and plasma investigations. Submitted to Contributions to Plasma Physics
13. Astanei D, Muntaneu F, Nemes N, Pellerin S, Hnatiuc B (2015) Electrical diagnostic of high voltage discharges produced in a new spark-plug. In: Proceedings of 13th IEEE international conference on engineering modern electric systems (EMES 2015), June 11–12, 2015. Oradeav (Romania), pp 1–4. https://doi.org/10.1109/emes.2015.715838800001
14. Mariani A, Foucher F, Moreau B (2013) The effects of a radio frequency ignition system on the efficiency and the exhaust emissions of a spark-ignition engine. SAE International No 2013-24-0053. https://doi.org/10.4271/2013-24-0053
15. Mariani A, Foucher F (2014) Radio frequency spark plug: an ignition system for modern internal combustion engines. Appl Energy 122:151–161
16. Zhou J (2013) Etude de l'effet du taux d'oxygène sur la combustion en moteur à allumage commandé suralimenté. Ph.D. Thesis, Orleans University
17. Masurier J-B (2016) Etude expérimentale de la combustion HCCI par l'ajout d'espèces chimiques oxydantes minoritaires. Ph.D. Thesis, Orléans
18. Sarangi AK (2012) Diesel low temperature combustion: an experimental study. Ph.D. Thesis, Loughborough University. https://dspace.lboro.ac.uk/2134/10192
19. Klimstra J (1985) The optimum combustion phasing angle—a convenient engine tuning criterion. SAE Technical Paper 852090. https://doi.org/10.4271/852090
20. Koziel JA, Pawliszyn J (2001) Air sampling and analysis of VOCs with solid phase microextraction. J Air Waste Manag Assoc 51:173–184
21. Mindrup R, Shirey R (2001) Improved performance of SPME fibers and applications. Sigma-Aldrich Co., note no T401042
22. Baimatova N, Koziel JA, Kenessov B (2015) Quantification of benzene, toluene, ethylbenzene and o-xylene in internal combustion engine exhaust with time-weighted average solid phase microextraction and gas chromatography mass spectrometry. Anal Chim Acta 873:38–50
23. Air sampling of VOCs using SPME for analysis by capillary GC (1998) Sigma-Aldrich Co.,/Supelco catalog, App Note 141–145. https://www.sigmaaldrich.com/content/dam/sigma-aldrich/docs/Supelco/Application_Notes/4680.pdf

24. Tuduri L, Desauziers V, Fanlo J-L (2001) Potential of solid-phase microextraction fibers for the analysis of volatile organic compounds in air. J Chromatogr Sci 39:521–529
25. Astanei D et al (2013) Etude d'une bougie a double étincelle pour la combustion propre. J Natl de la Recherche en IUT-JNRIUT 4:51–63
26. NIST mass spectral library (2017) Identify any spectrum with our NIST library upgrade. http://nistmassspeclibrary.com/2017/05/identify-any-spectrum-with-our-nist-library-upgrade/
27. Elliot M, Nebel G, Rounds F (2012) The composition of exhaust gases from diesel, gasoline and propane powered motor coaches. J Air Pollut Control Assoc 5(2):103–108
28. Caplain I, Cazier F, Nouali H, Mercier A, Déchaux JC, Nollet V, Joumard R, André JM, Vidon R (2006) Emissions of unregulated pollutants from European gasoline and diesel passenger cars. Atmos Environ 40:5954–5966
29. Volckens J, Olson DA, Hays MD (2008) Carbonaceous species emitted from handheld two-stroke engines. Atmos Environ 42:1239–1248
30. Jia LW, Shen MQ, Wang J, Lin M (2005) Influence of ethanol-gasoline blended fuel on emission characteristics from a four stroke motorcycle engine. J Hazard Mater A123:29–34
31. Liñan A, Williams FA (1993) Fundamental aspects of combustion in Oxford engineering science series. Oxford University Press. https://www.cambridge.org/core/journals/journal-of-fluid-mechanics/article/div-classtitlefundamental-aspects-of-combustion-by-a-linan-andamp-f-a-williams-oxford-university-press-1993-167-pp-isbn-019507626-5-25-div/B9510B2EEADDD2EC1C81A2519D3EF0DB