*Dedicated to Professor Ioan Bâldea on the
Occasion of His 80th Anniversary*

THE DEVELOPMENT OF A NEW OPTICAL METHOD TO MEASURE THE DELAY TIME OF SPARK IGNITION

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ABSTRACT. The development of a new optical method to measure the delay time following the spark ignition in gaseous explosive mixtures, based on the measurement of the emitted radiation intensity detected with a photo diode active in visible and near infrared range is presented. Several CH_4-air-inert mixtures were used to test the validity of the method. Our results are comparable with the available literature data. The influence of the different operational factors was investigated to establish the range of the optimum conditions.

Keywords: spark ignition, ignition delay, emitted radiation intensity, CH_4-air-inert mixtures

INTRODUCTION

The homogeneous combustion reactions in gas phase are complex reactions involving hundreds of molecular species and coupled elementary reactions. They are generally exothermic reactions and many imply also the participation of chain-branching mechanism. A simplified picture of temperature increase in time was suggestively illustrated in literature [1]. Using a (log T) against time diagram for thermal and chain branching explosions, a straight-line passing through origin or a straight-line intersecting X axis after an induction period, respectively, were found. During a chain-branching explosion, an induction period is necessary for accumulation of a critical concentration of

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active species. When such a system is suddenly brought into a critical state, able to react in an explosive regime, a self-ignition delay time is necessary. The ignition of the explosive regime occurs either as self-ignition, when the whole system reacts simultaneously, or as a forced ignition, starting from a small region where the ignition energy is delivered and propagates through the whole system. The self-ignition is a frequently encountered process with many applications. Consequently, many experimental techniques were dedicated and extensively used. Among these, rapid compression machines (RCM) [2-5], shock tubes (ST) [6-10] or both [11-15] are the most widespread. A qualitative description of the self-ignition delay is given in Figure 1 as a plot pressure P or radiation intensity RI in time:
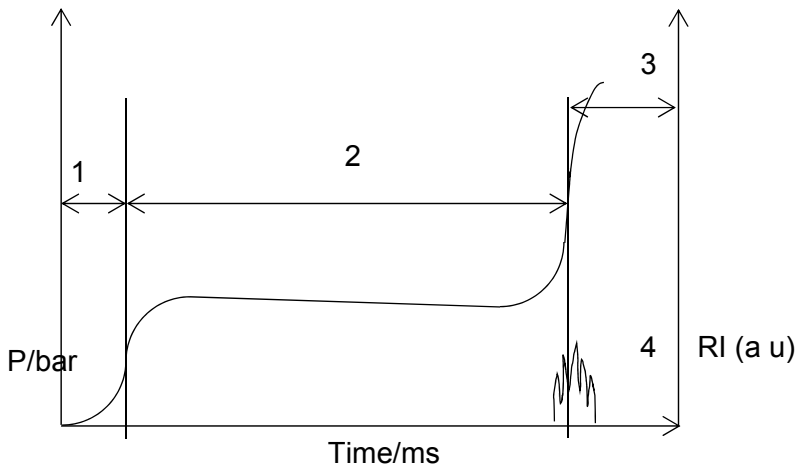
Figure 1. 1-Preignition period; 2-Ignition delay; 3-explosion;
4-Emitted radiation intensity in arbitrary units

The forced ignition, particularly spark ignition, sometimes supplemented with plasma or laser [15-20], was frequently studied using constant volume combustion bombs (CVB). The method presented in this paper relies on the analysis of the radiation intensity variation during the early stages of spark ignition in a CVB.

Due to the numerous applications of delay time in optimization of internal combustion operation, diminution of engine knocking, control of pollutant emission or validation of detailed kinetic models, the literature on these subjects is extremely rich and therefore only selected references were given.

RESULTS AND DISCUSSION

The present paper originates in our previous work regarding the laminar flame propagation in flammable gaseous mixtures using the pressure history and intensity of emitted radiation in closed vessels. It relies on the detailed analysis of the early stages of flame propagation. The use of the third power law of pressure increase during these stages, ΔP, combined with an improved statistical analysis of experimental data, resulted in a new method to evaluate the normal burning velocity and suggested the existence of an ignition delay time, τ, defined as a time interval between the ignition energy deposition and the beginning of steady flame propagation [21-27]:

$$\Delta P = \delta + k_2 * (t - \tau)$$

where δ is the base line correction and t is the time.

The method was validated either by comparing the resulted parameters with those reported in literature [21-28], or by analysing also the time evolution of emitted radiation within the same time interval [29].

This analysis relies on the deconvolution of the composite recorded curve of radiant intensity in time. It is composed of the emitted radiation due to spark emission and to flame emission. The first one is given in Figure 2.
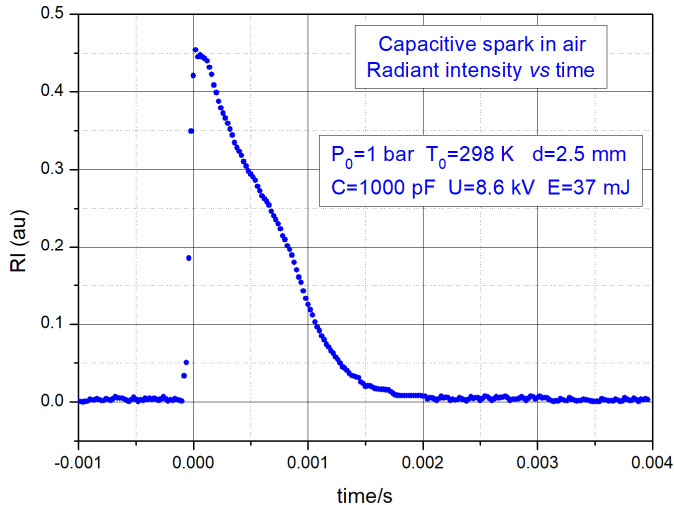
Figure 2. Radiant intensity - time due to spark emission (RI in arbitrary units) for spark gap length d=2.5mm and ignition energy E=37mJ

Even if capacitive sparks with discharging time of ~ 1 μ-second, or inductive sparks with discharging time of ~ 1 m-second were used, the general pattern and characteristics of the discharging emission are similar.

The composite curves, with both spark and flame emission, allow the measurement of the delay times, τ_1, τ_2, and τ_3, defined in Figure 3.
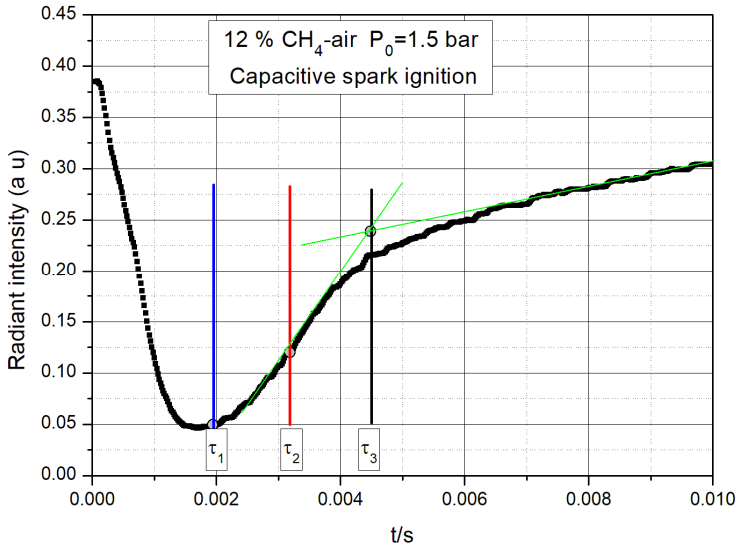

Figure 3. Experimental composite curve RI – time

There are three representative points on this curve: the first one, τ_1, at the end of spark emission and beginning of flame emission, the second one, τ_2, at the first inflection point of the ascending curve and the third one, τ_3.as seen in figure.

These are dependent on the operational conditions and initial mixture composition and can be used to describe the characteristics of the ignition process. Among the most relevant operational parameters are the spark characteristics and the initial mixture pressure and temperature. The mixture composition allows also to take into account the effect of different additives, frequently used in practical applications. As will be seen, the present experimental approach allows the measurements only at normal temperature. However, if mixtures with different fuel/air ratios or explosive mixtures with inert additives are investigated, some temperature dependent properties can be investigated and compared with the data obtained from variation of the same properties by changing the initial mixture temperature.

Spark characteristics effects on ignition delay times

The different ignition delay times can be significantly influenced by spark characteristics. To ensure similar conditions for spark discharge when pressure or composition are changed, an analysis of several operational factors is necessary. For capacitive sparks, these effects are illustrated in Figures 4 and 5.
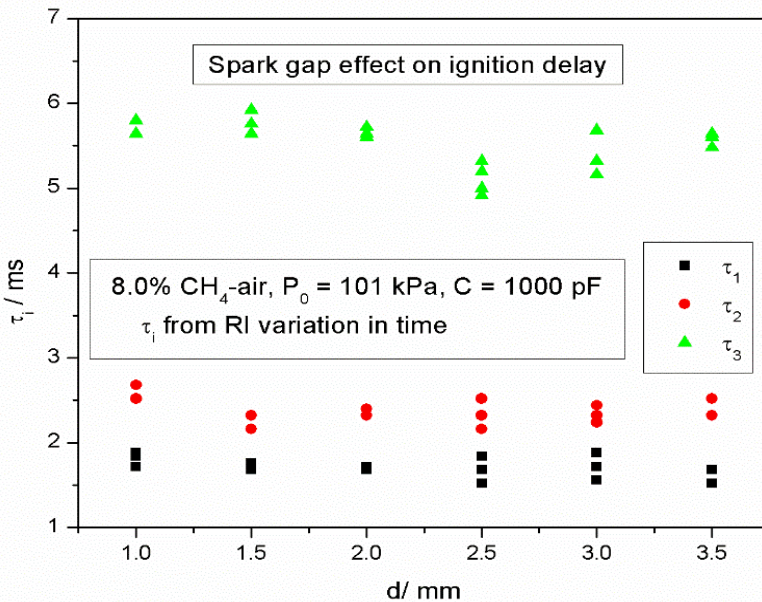
Figure 4. The effect of spark gap length (*d*) on the ignition delay for capacitive sparks with *C*=1000 *p*F

It can be observed that only τ_1 exhibits the best stability within a wide interval of spark gap. The observed differences originate, presumably, from the different stages of the flame development. It can be concluded that τ_1, when the radiant emission from the spark becomes nonsignificant and that from the minimal flame formation starts, is the best choice to be used when other operational parameters are varied. A similar trend was also observed for inductive-capacitive sparks as a function of the spark gap length.
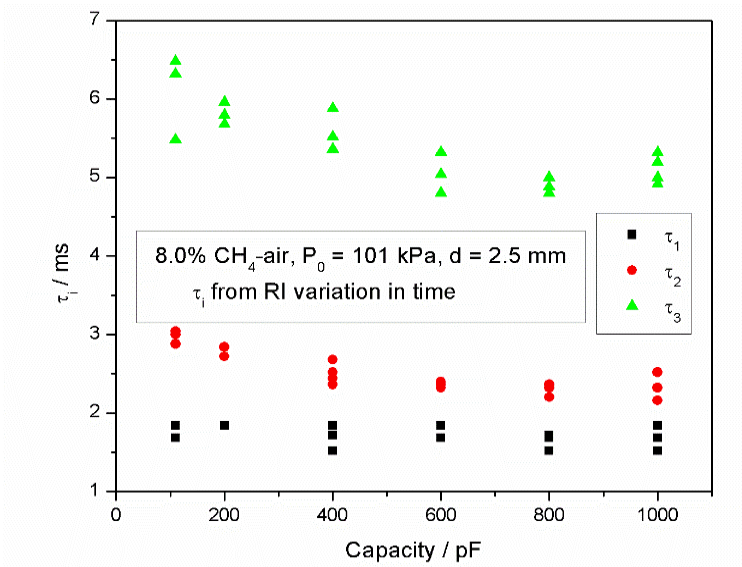
Figure 5. Effect of condenser capacity on the ignition delay
for a spark gap length d=2.5 mm

Again, only τ_1 has the best stability within a large interval of capacity.

Effect of initial pressure and temperature on the delay time

The dependence of the ignition delay on the initial pressure and temperature of the flammable mixtures was thoroughly studied in different experimental configurations for both self-ignition and spark ignition. The most frequently, the analytical form of this dependence is of Arrhenius type: exponential with respect to temperature and product of powers with respect to other parameters [13].

$$\tau = a_0 \cdot \prod P_j^{n_j} \cdot P_0^b \cdot \exp(E_a/RT) \tag{1}$$

where a_0, n_j, and *b* are empirical parameters, P_j or P_0 is the partial or total pressure, respectively, *T* is the temperature (K) and *R* = 8.314 J mol^{-1} K^{-1} is the universal gas constant [30, 31]. According to Equation (1), the baric coefficient *b* has the significance of an overall reaction order.

For systems with constant composition and temperature, Equation (1) takes a simpler form:

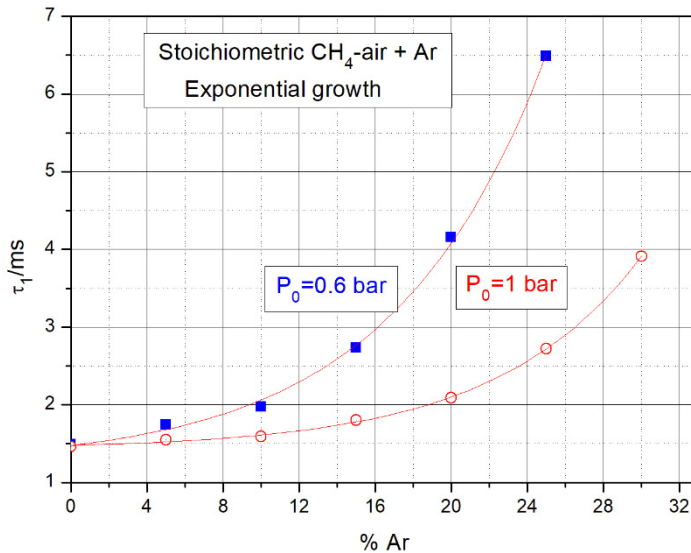$$\tau = a_0 \cdot \prod P_j^{n_j} \cdot P_0^b \tag{2}$$

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or:

$$\tau = a_1 \cdot P_0^b \tag{3}$$

where a_1 includes the mixture composition.

Effect of initial mixture composition on the delay time

Addition of different inert gases (like Ar, N_2, CO_2, H_2O or exhaust products) allows to modulate the mixture properties, including also the delay time. [32, 33] An illustrative example is given in Figure 6.


Figure 6. Effect of inert addition on the ignition delay, τ_1

Another possibility to change the mixture properties and implicitly both the ignition delay τ_1 and baric coefficient b is to use different fuel/air ratios as given in Figure 7 for different methane-air mixtures.

Sometimes, in many practical conditions, the mixture composition can be changed using both different fuel/air ratios and inert gases addition, keeping the initial temperature constant or changing it. In these cases, the minimal flame temperature, characteristic for the critical ignition properties including the ignition delay, can be obtained only by using numerical calculations. The adiabatic flame temperatures can be calculated with ECHIMAD, the 0D COSILAB package version 3.0.3 and the kinetic modelling with CHEMKIN or 1D COSILAB

package developed by Rogg and Peters [34-39]. Due to an increased interest in this field, numerous other numerical calculations can be found in the pertinent literature [34-40].
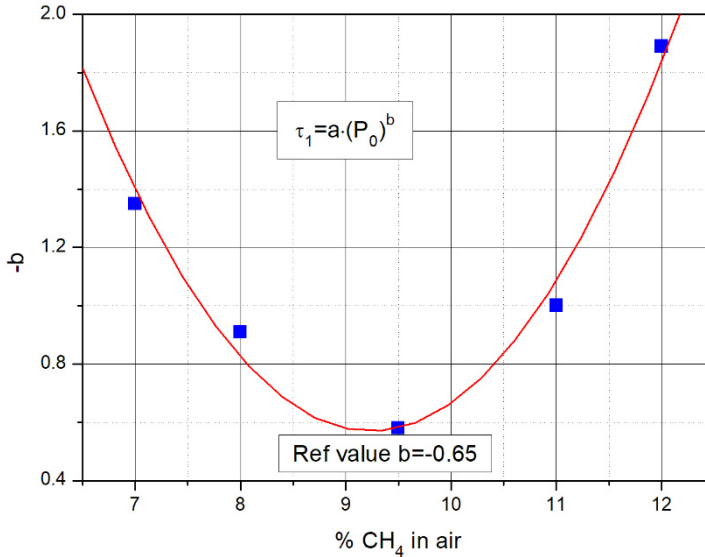
Figure 7. Variation of the baric coefficient b from the law (2) with CH_4 content in mixtures with air (ref. [28])

Additionally, taking into account the availability of these numerical calculations, the evaluation of temperature dependent properties becomes possible. In Table 1 the ignition delay τ_1 can be compared to experimental or calculated maximum of pressure rise ΔP_{max} and to calculated or available from other measurements of normal burning velocity S_u.

Table 1. The efficiency of several diluting components on inerting the stoichiometric CH_4-air mixtures containing 10% inert

| Inert	τ_{1}/ms	$\Delta P_{max}/bar$	$S_u/(cm/s)$
-	1.46	5.94	37.40
Ar	1.59	5.60	31.08
N_2	1.79	5.36	28.35
CO_2	2.45	4.68	19.14

Dilution with an inerting component results in a decrease of the ignition delay and of minimal flame temperature. A corresponding activation energy can be evaluated and compared with other reference values [40].
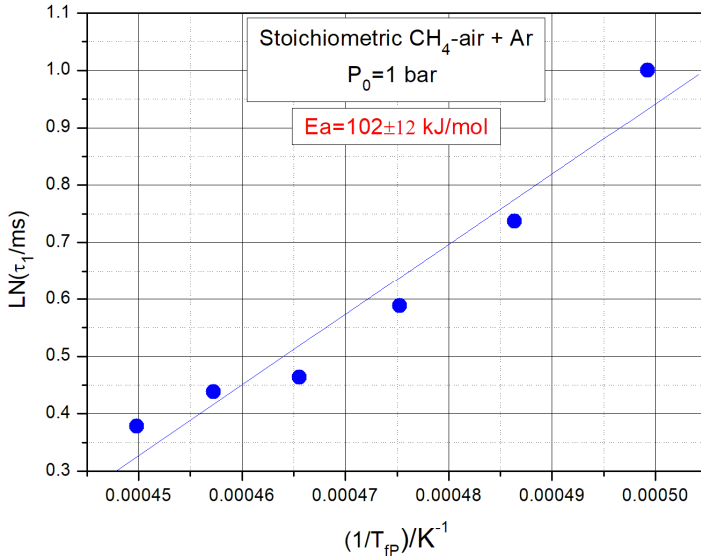
**Figure 8.** Variation of the ignition delay with flame temperature T_{fP}
(calculated here at constant pressure)

To underline the significance of τ_1, its relationships with other properties were examined. Some results are given in Figures 9 and 10:
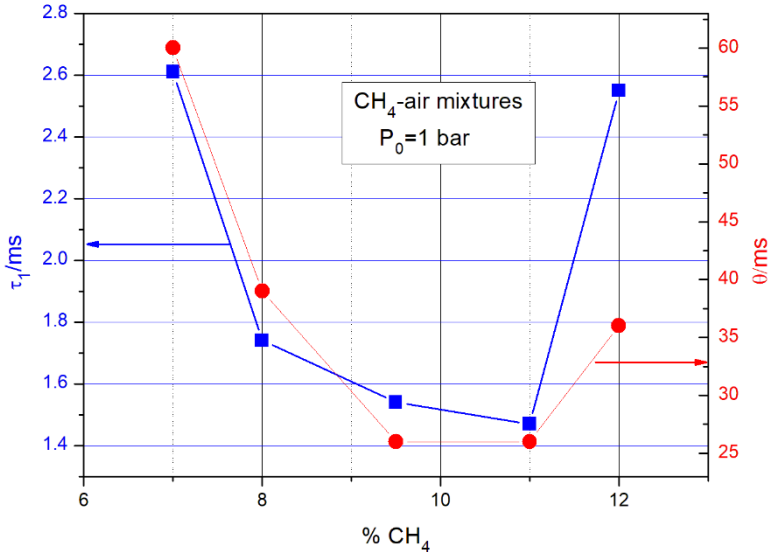
Figure 9. Comparison between the ignition delay τ_1 and the time θ
necessary to reach the maximum of the pressure rise ΔP_{max}

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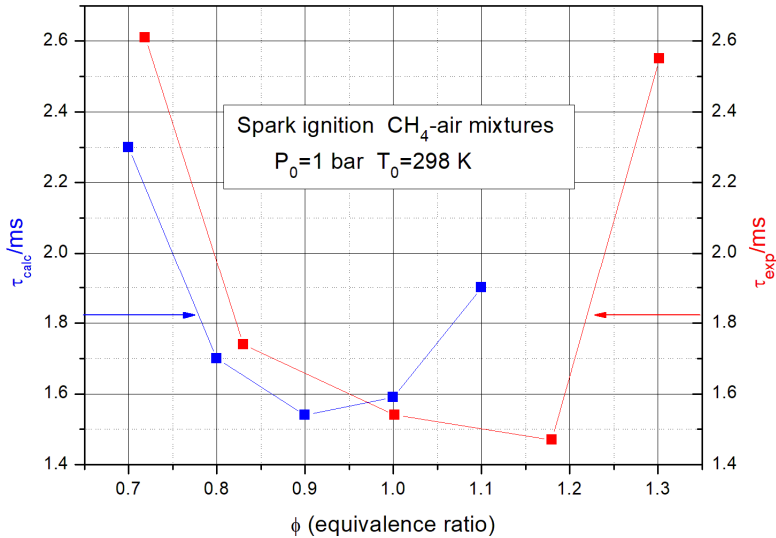
Figure 10. Comparison between experimental and calculated ignition delay.

The calculated ignition delays were obtained by using numerical calculations [34-39].

The adiabatic maximum rate of pressure rise, ΔP_{max}/bar, either calculated or experimental [41], measured explosion violence $(dP/dt)_{max}$/(bar s^{-1}), calculated volumetric rate of heat release, R_{max}/(J s^{-1} m^{-3}) are also properties related to the measured ignition delay times. Some of these correlations will be analysed in a future paper where the effects of other operational parameters will be taken into consideration. The interest for new alternative fuels like methanol and ethanol [42-44] suggest also another direction of research based on the method discussed here.

CONCLUSIONS

Taking into account the increasing interest for the ignition delay measurement when electrical sparks are used, the development of new and simpler techniques is quite beneficial. While the pertinent literature is based preponderantly on the measurement of flame size increase using high-speed time-resolved schlieren recording, our simpler method relies on the deconvolution of the composite recorded curve of radiant intensity in time, measured with a photodiode, in a constant volume combustion bomb. It is

composed from the emitted radiation due to spark emission superposed over of the flame emission. A procedure based on our previous paper allowed the development of this new optical method. The influence of the spark characteristics, initial mixture composition, pressure and temperature was studied and reported. Some relationships with other mixture properties were also discussed. All reported values are highly reproducible and in agreement with both experimental and calculated literature data.

EXPERIMENTAL SECTION

The experimental measurements were carried out in the Laboratory of Chemical Kinetics from the Department of Physical Chemistry, University of Bucharest, using the equipment given schematically in Figure 11.

The gaseous mixtures containing CH_4, air and sometimes inert were prepared in stainless steel cylinders at 4 bar total pressure by partial pressure method using methane 99.99% purity from SIAD and used 24 h after mixing. The experiments were carried out in a stainless-steel cylindrical explosion cell with diameter equal to height: $\Phi = h = 6$ cm ($V_0 = 1.70 \cdot 10^{-4}$ m^3 and with a radius of the equivalent spherical volume $R^*_{cell} = 0.03434$ m). The upper lid has a transparent window made from synthetic glass allowing the observation of the radiation intensity variation with a Si PIN S 123 photodiode (Hamamatsu). The lower lid was fitted with a pressure transducer. The ignition was initiated by high voltage sparks between 1.5 mm diameter stainless steel electrodes with rounded tips within a spark gap of 2.5 mm. The high voltage sparks were either capacitive or inductive. The capacitive sparks were obtained using an adjustable vacuum condenser fed from an adjustable high voltage source (Advanced Energy) interrupted at the end of discharge by an UltraVolt Opto Coupler OC250 (Voltage Multiplier INC.).

The ignition energy was higher than the minimum ignition energy (0.29 mJ) ensuring a safe ignition without important induced turbulence. The inductive sparks were obtained from an automotive ignition coil having the primary winding with an inductance L = 9.5 mH, fed from a dc stabilized power supply. The pressure variation during the explosion process was monitored with a Kistler piezoelectric pressure transducer type 601A coupled with a charge amplifier type 5011B and recorded using a Tektronix TDS 210 oscilloscope.
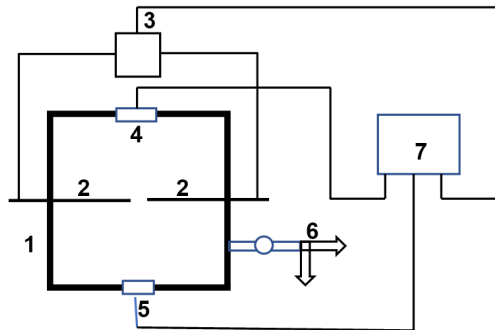
Figure 11. A simplified scheme of the experimental setup: 1. Explosion cell; 2. Spark electrodes; 3. Adjustable high voltage generator; 4. Photodiode; 5. Piezoelectric pressure transducer and charge amplifier. 6. Connections to vacuum and mixing systems; 7. Oscilloscope connected to spark generator (3), photodiode (4) and pressure transducer (5)

ACKNOWLEDGEMENTS

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