Measurement of Critical Energy for Direct Initiation of Spherical Detonations in High-Pressure H_2-O_2 Mixtures

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1 Introduction

The critical energy for direct initiation of detonation is an important dynamic parameter to provide a quantitative measurement of the detonation sensitivity of an explosive mixture in an unconfined environment. With the recent interest in hydrogen economy, measurement of this dynamic parameter at high pressure condition is desirable to assess the potential detonation hazards of high pressure storage facilities of hydrogen [1].

The detonation sensitivity behavior of hydrogen mixture at elevated initial pressure can differ from that of other hydrocarbon mixture due to the unique characteristics in hydrogen oxidation kinetics and the (extended) second explosion limit effects. Although simple theoretical analysis had indicated that above some critical pressure value, the required energy for direct initiation in hydrogen-air increases gradually with increasing initial pressure [2], nevertheless, this phenomenon has not been verified experimentally and indeed only very few data on the critical energy of direct initiation at high-pressure conditions are available.

The aim of this paper is to obtain measurements of critical direct initiation energy of spherical hydrogen-oxygen detonation. Experiments are carried out in a spherical bomb and initiation results from a high voltage capacitor discharge. One of the challenges in measuring critical direct initiation energy is to estimate the actual amount discharged into the mixture for initiation. Therefore, various methods (i.e. the calorimeter method and current method) are explored and compared in this study to obtain a good estimate of the correct amount of energy deposited into the mixture used to initiate the detonation. These results will be useful to understand the effect of kinetics on hydrogen detonation sensitivity at high pressure conditions and to develop proper correlation model for safety assessment.

2 Direct detonation initiation experiments

The experiment is carried out in high pressure spherical explosion chamber 8” in diameter and 2” in wall thickness. As seen in the diagram the chamber is connected to the manifold through a ball valve. The sphere is incrementally filled through this valve with a mixture of hydrogen and oxygen at various test pressures. A pressure transducer is mounted in the wall of the chamber to measure the blast pressure and the time of arrival of the detonation front. A slender electrode is mounted on top of the sphere. At the end of this electrode there is a spark gap through which the energy is delivered inside the chamber through an ignition circuit. The spark energy measurement is presently based on a method developed by Lee and Knystautas [3].

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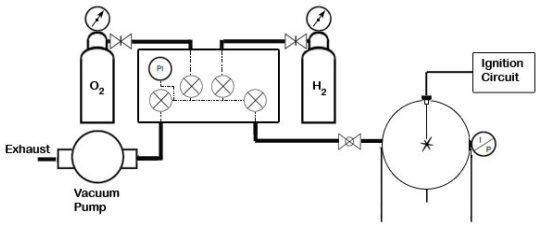

Figure 1. Experimental setup

3 Energy estimation from the high-voltage capacitor discharge

3.1 Current measurement method

As seen in the ignition circuit diagram the energy deposited inside the sphere comes from the discharge of the high voltage capacitor bank. Total energy stored in the capacitors ($E_T = CV^2/2$) could be used to estimate the energy required to detonate the hydrogen-oxygen mixture. However this estimate will only give the order of magnitude approximation to the actual energy deposited in the sphere since it does not take into account energy loss in circuit components and loss via ohmic dissipation. The actual energy deposited inside the mixture is found by integrating the square of current function $i(t)^2$ multiplied by the spark resistance (R_s): $E_s = \int_0^\infty i^2 R_s dt$. The ignition circuit in Fig. 2 can be simplified to an equivalent RLC circuit shown in Fig. 1. Thus for a damped oscillatory discharge the current can be described as: $i(t) = Ae^{-\alpha \cdot t}\sin(\omega t)$. Attenuation factor $\alpha = R_{total}/2L_{total}$, frequency of the discharge $\omega = \sqrt{1/(LC)_{total} - (R/2L)_{total}^{\ 2}} = \omega_n\sqrt{1-\varsigma^2}$, where ω_n is the natural frequency of the undamped discharge and ζ is the damping factor. The total circuit resistance ($R_{total} = R_{circuit} + R_s$) is a combination of the resistance of all the circuit components $R_{circuit}$ and igniter spark gap resistance R_s. To estimate the energy E_S deposited into the mixture from a trace of the current function on the oscilloscope, the following procedure is outlined:

1. Natural frequency ω_n is determined using ω and ζ found from the current trace.
2. Total circuit inductance is found from $L_{total} = 1/\omega_n^2 C_{total}$
3. Total circuit resistance is found as follows $R_{total} = 2L_{total}\alpha$
4. Spark Resistance R_s is determined by subtracting $R_{total} = R_{circuit}$ when the spark gap is shorted out $R_s \approx 0$ from $R_{total} = R_{circuit} + R_s$ when the spark is not shorted. Current traces of the two cases are shown in Fig 4.
5. Total Energy deposited into the mixture is found by numerically integrating the square of the current multiplied by spark resistance $E_s = \int_0^\infty i^2 R_s dt$.

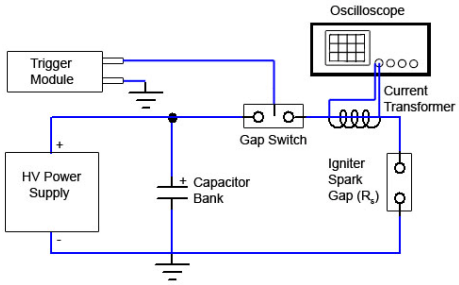

Figure 2. Ignition Circuit Components

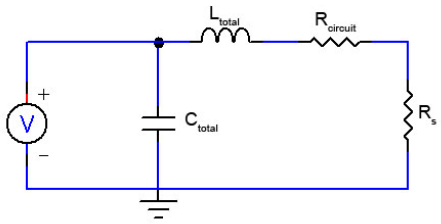

Figure 3. Equivalent RLC Circuit Diagram

It should be noted that according to Lee et al. only the energy deposited in the first ¼ of the discharge cycle is responsible for detonating the mixture. Thus the results will include both the total energy and energy deposited in the first ¼ cycle.

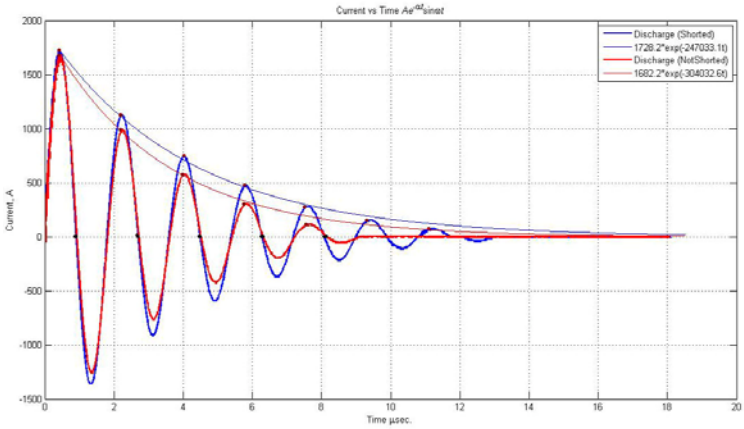

Figure 4. Comparison of the discharge current through shorted and open spark gap.

3.2 Calorimeter Method

Another technique that is being currently considered is the Calorimeter method. It will be used as an alternative energy measurement method in order to verify results obtained from the current measurement method. The idea is that the energy deposited into the mixture is estimated by measuring the change in static pressure of air in a fixed volume container. Since the density in a fixed volume container remains constant the energy can be calculated using ideal gas law:

$$E_s = \Delta Q = m_{air} \cdot c_v \cdot (T - T_{initial}) = m_{air} \cdot c_v \cdot T_{initial} \cdot (\frac{(P_{initial} + \Delta P)}{P_{initial}} - 1)$$

4 Results and discussions

To obtain some estimates, the simple relationship proposed by Lee et al. [4], linking the initiation energy and the cell size, is used. The model is obtained based on the minimum surface energy of the critical tube to the surface area of the critical size of the kernel. Together with the blast theory, the critical initiation energy can be obtained as:

$$E_c = 4\pi\gamma p_o M_{CJ}^2 I\left(\frac{13\lambda}{4}\right)^3 = \frac{2197}{16}\pi\rho_o V_{CJ}^2 I\lambda^3$$

where ρ_o is the initial density of the mixture, V_{CJ} the CJ detonation velocity, λ the cell size and I is a numerical constant (for γ = 1.4, I = 0.423). This expression thus provides a means of correlating the initiation energy with the cell size λ. The characteristic cell size λ for the hydrogen-oxygen mixtures at different initial pressures are estimated using a recently developed correlation model by Ng et al. [1-2] and is shown in figure 5. This model takes form of $\lambda = A(\chi)\cdot\Delta$ where Δ is the characteristic induction zone length which can be determined from chemical kinetics calculation. $A(\chi)$ represents a proportionality factor and is a function of a stability parameter χ [5]. This correlation model is validated against different cell size data and is shown to provide a good cell size estimates for hydrogen mixtures. The above correlation provides a simple estimate of the critical energy for direct initiation. Other detailed models will also be investigated in the course of this work.

Figures 6a and 6b show the pressure traces for a successful and unsuccessful direct initiation of a spherical detonation in the stoichiometric H_2-O_2 mixture at an initial pressure of p_o = 1atm. For successful initiation shown in figure 6a, a large pressure rise occurs about 150 µsec indicating a detonation. For much lower initiation energy (i.e. spark energy ¼ cycle), no significant pressure rise happens until 900 µsec at which the initiation is due to the mechanism. (e.g. due to the shock reflection from the chamber wall). In such case, no direct initiation occurs.

A series of shots are being done to measure the critical energy of H_2-O_2 mixtures at p_o = 1.0, 1.8 and 3.5atm (see figure 5b). More experiments will be performed at higher pressure near the critical value of the extended second explosion limit of hydrogen-oxygen mixtures and to narrow down the energy range to determine the critical value for direct initiation of a detonation.

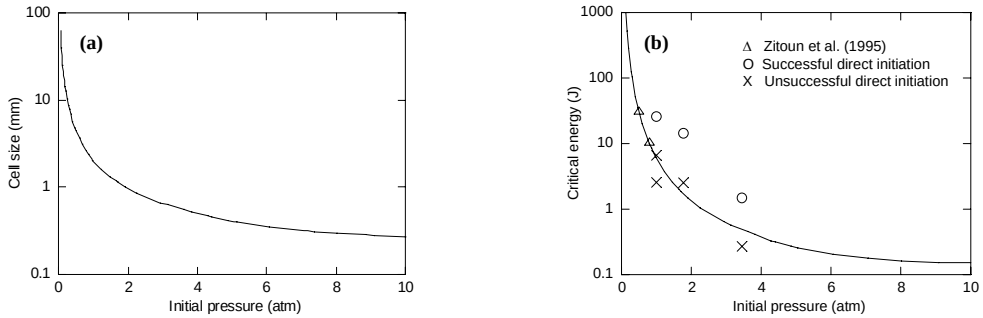

Figure 5. (a) Cell size estimation using the model by Ng et al.; and (b) the critical energy calculated using the simple relationship and measured value from experiments.

(a) Successful direct initiation

Capacitor bank 1uF		**Spark**			
Avg. Energy (J)	**Voltage** [kV]	$R_{circuit}$ [Ohm]	R_{spark} [Ohm]	**Energy (1/4)** [J]	**Energy (total)** [J]
165	18.1659	1.064	0.045	25.7575	61.98

(b) Unsuccessful direct initiation

Capacitor bank 0.21uF		**Spark**			
Avg. Energy (J)	**Voltage** [kV]	$R_{circuit}$ [Ohm]	R_{spark} [Ohm]	**Energy (1/4)** [J]	**Energy (total)** [J]
51.4	22.1252	0.222	0.0502	2.5547	7.4886

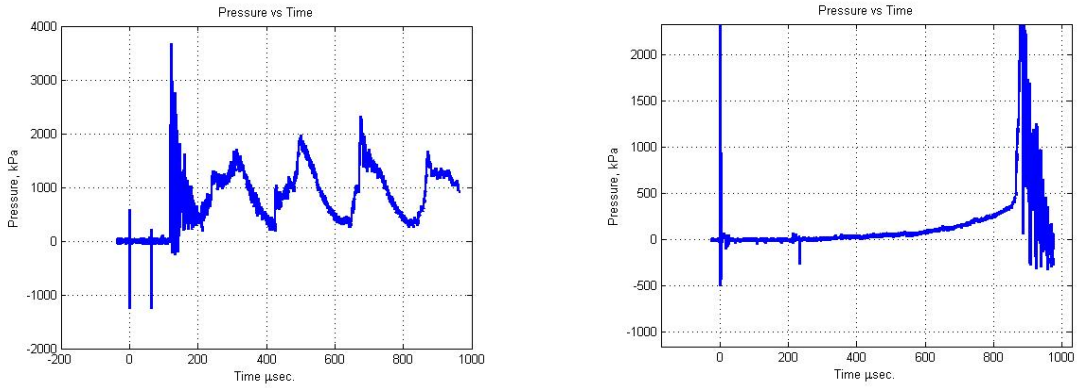

Figure 6. Pressure traces for the (a) successful and (b) unsuccessful direct initiation of a spherical detonation in hydrogen-oxygen mixtures at p_o = 1atm.

Acknowledgment

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