

Direct blast initiation of spherical gaseous detonations in highly argon diluted mixtures

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by

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Abstract

In this study, direct initiation of spherical detonations in highly argon diluted mixtures is investigated. Direct initiation is achieved via a high voltage capacitor spark discharge and the critical energy is estimated from the analysis of the current output. Stoichiometric acetylene-oxygen mixtures highly diluted with 70% argon is used in the experiment. Previous investigations have suggested that detonations in mixtures that are highly diluted with argon have been shown to be “stable” in that the reaction zone is at least piecewise laminar described by the ZND model and cellular instabilities play a minor role on the detonation propagation. For the acetylene-oxygen mixture that is highly diluted with argon, the experimental results show that the critical energy where the detonation is “stable” is in good agreement with the Zel’dovich criterion of the cubic dependence on the ZND reaction length, which can be readily determined using the chemical kinetic data of the reaction. The experimental results are also compared with those estimated using Lee’s surface energy model where empirical data on detonation cell sizes are required. Good agreement is found between the experimental measurement and theoretical model prediction, where the breakdown of the 13λ relationship for critical tube diameter – and hence a different propagation and initiation mechanism – is elucidated in highly argon diluted mixtures and this appears to indicate that cellular instabilities do not have a prominent effect on the initiation process of a stable detonation.

Keywords: Direct initiation; spherical detonation; critical energy; stable mixture; spark ignition

1. Introduction

Since the pioneering work by Zel'dovich et al. [1], the problem of direct blast initiation of spherical detonation has been studied extensively for the past four decades. Direct initiation requires the explosive release of a relatively large quantity of energy by the igniter. The magnitude of the critical source energy for successful direct initiation in a given explosive mixture has long been considered as perhaps the most direct experimental means of assessing the explosive sensitivity of the mixture [2].

The direct blast initiation problem lends itself well to both theoretical and experimental investigation of the detonation formation phenomenon [3]. Theoretically, the transient flow field associated with the decaying strong blast is well described by the classical similarity solution of Taylor, von Neumann and Sedov [4]. The influence of chemical reactions in modifying the blast flow field leading to the onset of detonation can be studied readily by a number of analytical techniques. The problem can also be readily described numerically. Experimentally, condensed explosive charges or sparks from a rapid low inductance capacitor discharge can closely approximate the ideal instantaneous point energy source of the classical similarity solution [5]. Thus, blast initiation had been studied extensively in the past five decades. Nevertheless, despite significant advances in the understanding of the direct initiation phenomenon, a quantitative theory that can predict the critical energy required for direct initiation from first principle (i.e. basic thermo-chemical and kinetic rate data of the explosive mixture) is still lacking [3].

A major advance was made by Zel'dovich, Semenov and Kogarko [1], who postulated the correct form of dependence of the critical initiation energy on the chemical length scale. They established that the critical energy for direct initiation should be proportional to the cube of the chemical length scale of the detonation. This criterion (generally referred to as Zel'dovich's criterion) follows from the fact that

the blast is characterized by the explosion length scale (i.e. $R_o = (E_o/p_o)^{1/3}$). It follows from dimensional considerations that the explosion length must correlate with the chemical length scale that describes the detonation structure. If the ZND induction length of a stable one-dimensional CJ detonation is used, it is shown to under-estimate the critical energy by three orders of magnitude [6]. Numerical simulations on direct initiation also indicate that for unstable detonations where instability plays a more prominent role in controlling the initiation process, the energy correlation deviates from the Zel'dovich criterion [7].

Later investigations re-defined the characteristic chemical length and used the detonation cell size as the characteristic length scale for the detonation structure. The formation and propagation of a cellular detonation rely on the strong nonlinear coupling between the details of the transient gasdynamics and chemical reactions, with the end results of the interaction manifested by the detonation cell size [8]. The use of the cell size usually results in a much better correlation with the critical energy. Unfortunately, the cell size is a difficult dynamic parameter to determine from smoked foils and at least it cannot be measured with a degree of certainty better than a factor of two. Since the critical energy is proportional to the cube of the chemical length scale (e.g. cell size), an uncertainty of 100% result in a possible error in the energy by an order of magnitude. Thus it has not been possible to advance the theory of direct blast initiation due to the lack of an unambiguous length scale to characterize the detonation structure.

Recent studies have indicated that for highly argon diluted mixtures, the detonation is relatively stable in that the reaction zone is at least piecewise laminar described by the ZND model [9, 10]. It is suggested that for such “stable” mixtures, cellular instabilities play minor roles on the self-sustained propagation of the detonation [11] as well as the detonability limit [12]. Thus it is of interest to investigate if the effect of cellular instabilities is relatively insignificant in the direct initiation

phenomenon and if the critical energy for direct initiation correlates well with the ZND reaction length which can be accurately determined using the chemical kinetic data of the reaction.

The present paper describes the result of an experimental study of direct blast initiation in stoichiometric mixture of acetylene and oxygen with 70% of argon dilution at different initial pressures. Experiments are carried out in a spherical bomb and initiation energy comes from a high voltage low inductance capacitor spark discharge. By comparing the chemical induction length obtained by solving the steady ZND equations with detailed chemical kinetics, the validity of the Zel'dovich criterion of dependence of the critical initiation energy on the chemical length scale can be verified. The experimental results are also compared with the predicted values using Lee's surface energy theoretical model where empirical cell size data are used as characteristic length scale. This semi-empirical model makes use of the critical tube diameter, a phenomenon which is shown to be different between stable and unstable detonations. From the results comparison, the breakdown of the 13λ relationship for critical tube diameter of stable detonation in explosive mixtures highly diluted with argon is elucidated and from that, it can be shown that cellular instabilities also play minor roles in the initiation mechanism.

2. Experimental Details

Experiment is carried out in a 8" diameter and 2" wall thickness high pressure spherical bomb. A schematic of the apparatus is given in Fig. 1. For each direct initiation experiment, the sphere is initially evacuated to at least 80 Pa and then incrementally filled through the ball valve with mixtures at various test pressures. A PCB piezoelectric pressure transducer is mounted in the wall of the chamber to measure the time of arrival of the detonation front. It can be determined from the arrival time of pressure signal whether there is a successful detonation or not.

The ignition system as shown in Fig. 2a consists of a high voltage power supply, capacitor bank, a gap-switch, a trigger module (TM-11A) and a slender coaxial electrode mounted on top of the explosion sphere. At the end of this slender electrode there is a 3.5 mm spark gap through which the energy is delivered inside the chamber through the ignition circuit. The ignition circuit can be simplified as an equivalent R-L-C circuit, as be shown in Fig. 2b. Although the total energy stored in the capacitors ($E_T = CV^2 / 2$) can be used to evaluate the energy deposited inside the mixtures, owing to energy loss and ohmic dissipation this kind of estimate cannot represent the actual energy responsible for the initiation. It only gives an order of magnitude approximation to the actual energy deposited in the mixtures. Therefore, 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): $E_s = \int_0^\infty i^2 R_s dt$. For a damped oscillatory discharge the current can be described as: $i(t) = Ae^{-\alpha t} \sin(\omega t)$. Attenuation factor is equal to $\alpha = R_{total}/2L_{total}$ and frequency of the discharge is equal to $\omega = \sqrt{1/(LC)_{total} - (R/2L)_{total}^2} = \omega_n \sqrt{1 - \zeta^2}$, 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 procedure laid out in [13] is adopted and described as follows:

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. 3. By determining the exponential decay in amplitude $Ae^{-\alpha t}$ of the current trace, the attenuation factor α can be estimated and hence the total circuit resistance R_{total} for each case.

5. Total energy deposited into the mixture is found by numerically integrating the square of the current multiplied by spark resistance: $E_s = \int_0^{\infty} i^2 R_s dt$

It has been indicated by Lee et al. [14] that only the energy deposited in the first $\frac{1}{4}$ of the discharge cycle is responsible for detonating the mixture; the energy subsequent to this time was found to have no noticeable influence on the initiation processes since initiation is already completed. This has also been verified in the recently work by Kamenskihs, Ng and Lee [13] and it is shown that the current method gives a good estimate to the actual energy amount deposited into the mixture responsible for direct initiation.

3. Results and Discussion

Typical pressure traces for a successful direct initiation of a self-sustained spherical detonation and for an unsuccessful initiation case in the stoichiometric $\text{C}_2\text{H}_2\text{-O}_2\text{-70\%Ar}$ mixture at an initial pressure of $p_o = 100$ kPa are shown in Figs 4. Figure 4a corresponds to a successful “go” and the initial energy is estimated to be 0.89 J from the method described in previous section. The onset of detonation, as indicated by the large pressure rise in the pressure trace, is achieved in about 85 μs . From the time of ignition and time-of-arrival of the wave given in the pressure trace, the wave velocity can also be determined. The velocity of the wave is 1705.8 m/s, which is of 93.1% of CJ detonation velocity. The experimental results indicated that the final detonation velocity were slightly less than the CJ value, which could be an effect of the curvature of the spherical detonation. It is worth noting here that an accurate measurement of the wave velocity is indeed not essential to determine the successful/unsuccessful initiation cases, which are mainly recognized from the rises of the pressure

traces. The exact value of the wave velocity is also not considered for the subsequent theoretical analysis. Figure 4b is the pressure trace of an unsuccessful “no go” case with the source energy of 0.80 J, an amount less than the critical energy. No significant pressure rise happens for a long period of time until $\sim 3160 \mu\text{sec}$ at which point the mixture may be initiated by auto-ignition due to the shock reflection from the chamber wall. In such case, no direct initiation occurs. Table 1 summarizes the details of the experimental results and parameters for successful direct initiation and unsuccessful initiation.

The present experimental results of all measured critical energies are summarized in Fig. 5, plotted as a function of ZND induction length. All the ZND chemical length scales are computed using the Chemkin package [15] with the Konnov chemical kinetic mechanism [16]. The Konnov mechanism had been validated and proved to be suitable for detonation simulation [17]. The variation of induction zone length as a function of initial pressure is plotted in Fig. 6 and the power law fit is found to be:

$$\Delta[\text{mm}] = a \cdot (p_o[\text{kPa}])^b \quad \text{where } a = 2.3215, b = -1.127 \quad (1)$$

These experimental results are used to verify the Zel’dovich criterion of blast initiation. According to Zel’dovich et al. [1], the parametric connection between the critical initiation energy E_s of the point source and the chemical length scale should be:

$$E_s \sim \Delta^3 \quad (2)$$

Zel’dovich criterion is based on the argument that the blast energy must be such that, when the blast has decayed to the CJ state, the blast radius must be at least of the order of the induction zone length. This cubic scaling form of dependence of critical energy on the ZND chemical length scale is also shown in comparison with the experimental data in Fig. 5. We can see that for the range of initial pressure considered in the present experiment, where the induction zone length is small and the detonation is stable, the agreement with Zel’dovich’s criterion is quite good.

Since stable detonations in heavily argon diluted mixtures propagate via the classical ZND model, hence the ZND reaction zone length which can be readily computed from chemical kinetics calculation, appears well to be an appropriate length scale that characterizes the structure of the detonation front and to scale with the critical energy for direct blast initiation of stable detonation. These results seem to indicate that detonations in highly argon diluted mixtures are piecewise ZND laminar in that cellular instabilities play minor roles on the initiation and subsequent self-sustained propagation of the detonation.

For further comparison, the critical energies for direct initiation are also estimated using the Lee et al. surface energy model [5, 18]. This semi-empirical phenomenological model relates the point blast initiation mode with the planar wave initiation mode and the critical energy is correlated with the cell size as the characteristic length scale. The link is established based on the minimum surface energy of the critical tube to the surface area of the critical size of the minimum detonation kernel, in other words, the correlation is through equating the surface energy contained in the wave in both cases at criticality. Thus the surface energy contained in the point blast initiated spherical detonation wave at the time when the wave has decayed to the CJ state from its overdriven state is equivalent to the energy in the planar detonation wave in the critical tube diameter situation. Hence equating the minimum surface areas of both waves at criticality:

$$4\pi R^{*2} = \left(\frac{\pi}{4}\right) d_c^2 \quad (3)$$

thus yields a kernel radius $R^* = \frac{1}{4} d_c$. From strong blast wave theory, the blast wave energy is given by:

$$E_s = 4\pi I \gamma p_o M_s^2 R_s^3 \quad (4)$$

According to Zel'dovich's criterion, when the blast wave decays to $M_s = M_{CJ}$, it should engulf a kernel size of radius $R_s = R^*$.

The effect of argon dilution on the initiation energy can be elucidated in Eq. (4). To complete the initiation model, the value of critical tube diameter, d_c , is required. Critical tube diameter of common fuels-air mixtures generally follows well the empirical correlation $d_c = 13\lambda$ where d_c is the critical tube diameter and λ is the characteristic detonation cell size [19]. Although this empirical correlation appears to be quite adequate for most gaseous hydrocarbon explosives, but it is shown to be invalid for mixtures with regular cell patterns such as mixtures that are highly diluted with argon [20-22]. For such mixtures, it is found that the correlations ranged from $d_c = 25-30\lambda$. This breakdown indicated a distinct difference in the detonation critical tube diameter and propagation phenomena for the undiluted and highly diluted mixtures. It has been suggested that the detonation stability or cell regularity plays an important role in the detonation undergoing a sudden expansion into open space. Lee proposed two mechanisms, one local and one global for the diffracted wave to explain the failure and the breakdown of the 13λ rule [23]. The 13λ rule is generally applicable for mixtures with high degree of instability and the phenomenon is characterized by local explosion centers re-initiation mechanism. The breakdown of the 13λ correlation is ascribed to the regularity of the detonation front, typical for highly argon diluted mixtures. For stable detonations where the ZND structure is valid, the failure occurs due to the global mechanism when the curvature of the attenuated detonation exceeds some critical value required for self-sustained propagation. From these arguments, one may equivalently expect that these two distinct mechanisms will lead to a difference in the initiation process between an unstable detonation and a stable detonation in highly argon diluted mixture.

Therefore, for the stable C₂H₂-O₂-70% mixture considered in this work, the empirical correlation of $d_c = 25-30\lambda$ is used and the energy equation immediately yields:

$$E_s = 4\pi I \gamma p_o M_{CJ}^2 \left(\frac{d_c}{4} \right)^3 = \frac{3375\pi \rho_o V_{CJ}^2 I \lambda^3}{2} \quad (5)$$

where ρ_o is the initial density of the mixture, V_{CJ} the theoretical CJ detonation velocity, λ the cell size and I is a numerical constant (for $\gamma = 1.4$, $I = 0.423$, where V_{CJ} is computed from a thermodynamic equilibrium program named CEA [24]). As in many semi-empirical initiation models, the detonation cell size is required as a length scale for the energy estimation. For the mixtures and initial conditions considered in this work, the cell size data are available in literature [25] to complete Lee's surface energy model. In his Ph.D. dissertation, Radulescu [25] obtained a correlation between his experimentally measured cell size data λ for the acetylene-oxygen-70% argon mixtures and initial pressures, and the expression is given as:

$$\lambda [\text{mm}] = a \cdot (p_o [\text{kPa}])^b \quad \text{where } a = 113.8, b = -1.20 \quad (6)$$

The theoretical critical energy is thus a combination of Lee's surface energy model and the cell size correlation, which are Eq. (5) and Eq. (6), respectively. In fact, using the previous correlation of induction zone length with initial pressure given by Eq. (1), the cell size can be essentially scaled linearly with induction length by:

$$\lambda \sim 49 \cdot \Delta \quad (7)$$

where the proportionality constant in the linear relationship is essential a constant (i.e. with a weak dependence on initial pressure p_o). Hence, this once again concludes that the ZND induction length should be a sufficiently appropriate length scale which can be accurately determined from the chemical kinetics computation. The measurement of cell size can be avoided to correlate the critical energy for the stable mixture with high argon dilution.

The comparison between the experimental results and the critical energies predicted by the theoretical model is shown in Fig. 7. The maximum percentage difference between the theoretical prediction and the experimental measurement is about 40%. Taking into account the common ambiguity in the use of cell size data λ – variation ranging up to a factor of 2 between different foil measurements – and the cubic dependency of the theory on λ , the comparison still shows a fairly good agreement between the theoretical prediction and the experimental result. This good agreement between both the experimental and theoretical results thus elucidated the difference in the initiation mechanism for stable detonation where cellular instability play a minor role in the initiation process and evidences stems from the breakdown of 13λ for the critical tube diameter used in the surface energy model.

4. Concluding Remarks

In the present study, we investigated the direct initiation of spherical detonation and obtained the critical initiation energy at different initiation pressures in the stable mixture of $C_2H_2-2.5O_2$ with 70% of argon dilution where the ZND model structure is valid. By investigating detonations in these mixtures highly diluted with argon (where it has been indicated that the effect of cellular instabilities is not prominent in the propagation mechanism), the Zel'dovich criterion of the cubic dependence of critical initiation energy on the ZND chemical length scale is verified. For such mixtures in which the detonation is stable, the ZND induction length is sufficiently appropriate for correlating different dynamic parameters (e.g. critical energy for direct initiation) and can be readily scaled using a linear relationship with a constant proportionality factor over a wide range of initial pressure in obtaining the characteristic cell size of a detonation. Furthermore, good agreement between the critical energies obtained from the experimental result and theoretical prediction using Lee et al's surface energy model [18], where the breakdown of 13λ relationship is taken into account for the problem of critical tube diameter in stable

mixtures with high argon dilution, appear to confirm that cellular instabilities play only minor roles – if any – in the formation of a self-sustained propagating “stable” detonation.

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Table

Table 1. Results for successful direct initiation and unsuccessful initiation in $C_2H_2-2.5O_2$ with 70% of argon at 100 kPa

$E(J)$ $0.5CV^2$	Voltage V_o (kV)	$C_{circuit}$ (μF)	$R_{circuit}$ (Ω)	R_{spark} (Ω)	$E_{1/4 Cycle}$ (J)	E_{total} (J)	V (m/s)	$\% V_{CJ}$	Direct initiation
57.8	17	0.4	0.139	0.0268	0.89	6.71	1705.8	93.1	Yes
51.2	16	0.4	0.153	0.0242	0.80	5.79	45.9	2.5	No

Table 1

Figure Captions

Fig.1 Sketch of spherical bomb.

Fig.2 Schematics showing a) the ignition Circuit components and b) its equivalent RLC circuit diagram.

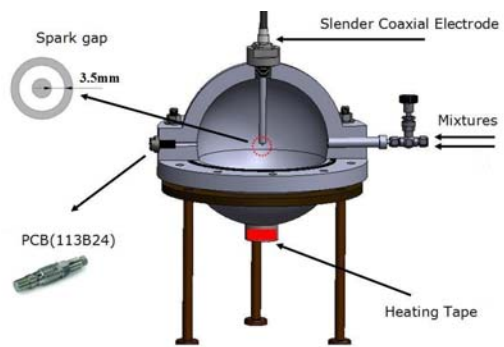
Fig.3 Discharge current through shorted and open spark gap in $C_2H_2-2.5O_2$ with 70% of argon. ($P_o= 60$ kPa $V_o= 18$ kV $C_{total}= 2 \mu F$).

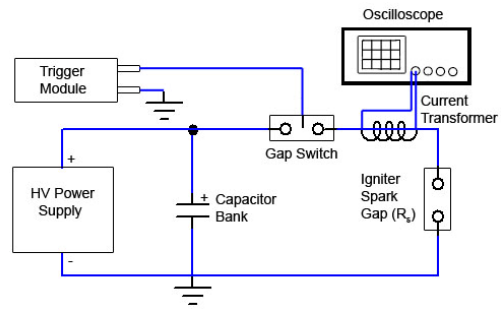
Fig.4 Pressure trace for a) the successful direct initiation and b) the unsuccessful direct initiation of a spherical detonation in $C_2H_2-2.5O_2$ with 70% of argon at 100 kPa.

Fig.5 Critical initiation energy for direct initiation of a spherical detonation obtained from the experiments in $C_2H_2-2.5O_2$ with 70% of argon as a function of ZND induction zone length.

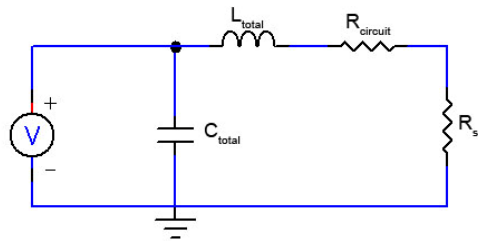
Fig.6 Variation of ZND induction zone length as a function of initial pressure.

Fig.7 Critical initiation energy for direct initiation of a spherical detonation obtained from the theoretical prediction (solid line) and experiments in $C_2H_2-2.5O_2$ with 70% of argon as a function of initial pressure.

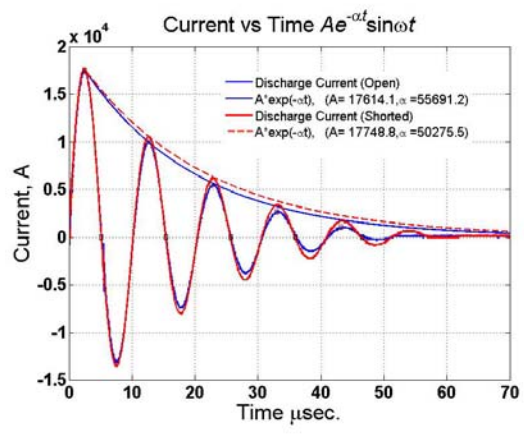


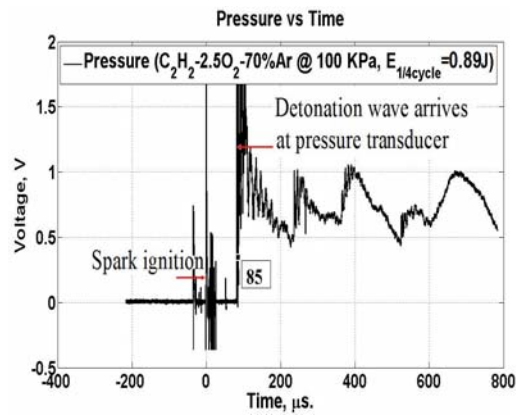


(a)

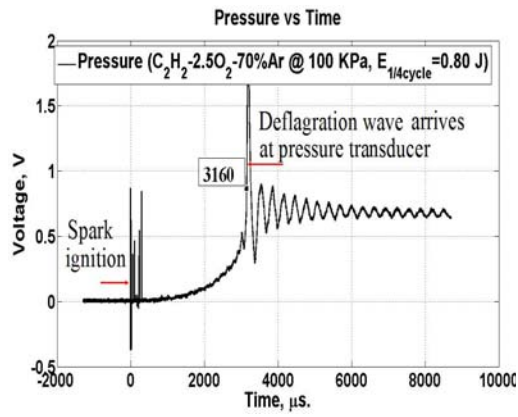


(b)





(a)



(b)

