Detonation Initiation by Gradient Mechanism in Propane–Oxygen and Propane–Air Mixtures

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An experimental study of detonation initiation by high–voltage nanosecond gas discharge has been performed in smooth detonation tubes. A gradient mechanism was used to initiate detonations in stoichiometric propane–oxygen mixtures with different nitrogen dilution and in propane–air mixtures. Initial pressures from 0.2 to 1bar have been tested. Detonation was formed within 4 transverse tube sizes at initial pressures higher than 0.2 bar for the propane–oxygen mixture and higher than 0.8 bar for the diluted mixture with 40% of nitrogen. The discharge energy inputs were 0.2—0.3 J. The gradient mechanism of detonation formation similar to the one suggested by Zeldovich has been shown to be the governing process. For the mixture with air, a detonation tube with an annular discharge chamber has been designed and tested.

Introduction

Studies of different mechanisms of detonation initiation in combustible gaseous mixtures are related to the necessity of development an ignition device for application in pulsed detonation engines (PDE). The application requires a robust energy–efficient method of initiation Chapman–Jouguet (CJ) detonations in tubes within distances of several calibers and times of < 1 ms. The most common approach is deflagration to detonation transition (DDT) facilitation by the application of various types of obstacles that turbulize the flow and increase the flame velocity, which was initially suggested by Shchelkin [1].

Alternatively, the concept of detonation initiation through a gradient mechanism was theoretically introduced by Zeldovich [2]. According to this work, an ignition delay time (or induction time) gradient formed due to a corresponding temperature gradient leads to the onset of a spontaneous combustion wave. The mixture first ignites at the point with the lowest delay and then propagates by spontaneous ignition with a velocity D_{sp} determined by the shape of the gradient. Depending on the value of D_{sp}, different modes of flame propagation can be realized. When D_{sp} exceeds the Chapman–Jouguet velocity D_{CJ}, a detonation wave can emerge from a constant–volume explosion as the spontaneous wave decelerates to the D_{CJ} value. This case was numerically studied in a series of papers reviewed in [3], where the detonation wave always occurred from a hot spot in the region between the shock wave and the turbulent flame. Various scenarios of hot spots formation due to shock–flame interaction and the role of their structure were also discussed in that paper. When D_{sp} falls into the interval between the speed of sound and DCJ , the spontaneous wave propagating over the induction time gradient can become coupled with the compression wave and thus evolve into a detonation wave through shock amplification and acceleration. A similar concept of the shock–wave amplification by coherent energy release (SWACER) was introduced in [4], where this process was investigated in more detail.

This paper focuses on the further study of this mode of detonation initiation with regard to its future application in pulsed detonation engines, continuing the study in [5,6]. In the first part, a four–cell discharge chamber is used to enhance the efficiency of the gradient formation in terms of energy input. Each of the discharge cells is of the same geometry as the ones used in all previous setups. This approach allows to estimate the required shape of the gradient along the channels and, thus, the necessary parameters of the discharge. The second part of the paper is dedicated to the experimental study of this mechanism for the practically important propane–air mixture. For optimal configuration of discharge propagation and its interaction with the gas flow, annular discharge channel has been developed.

DDT initiation by spark discharge

The spark mode of discharge development was realized in the whole pressure range under maximum pulse amplitude of 160 kV, which corresponded to pulse energy of 15 J. The discharge gap in the experiments at an initial pressure of 0.3 bar was 100 mm. This resulted in an energy input into the gas of <10 J. Typical *x–t* diagrams based on the pressure transducers (PT) and the infrared sensors (IR) are presented in Figure 1. The time is counted off from the moment the nanosecond discharge developed. The origin of the *X*-coordinate coincides with the grounded electrode of the discharge gap. The negative *X*-values correspond to the discharge channel and are used to represent the ignition process dynamics captured with ICCD cameras. Each data set in the positive *X*-values region corresponds to a pair of pressure transducers and infrared sensors installed at different distances from the nozzle inlet. It is seen from the data that, by the first measuring section, the flame front was closely coupled with the shock wave and propagated with a supersonic velocity

of over 700 m/s. The temporal delay between the waves was <10 μs, which yielded <7 mm for the gap length. Assuming that the flame front and the shock wave traveled at that velocity between the nozzle inlet and the first sensor would yield their propagation with nearly zero delay after the discharge. Such a pattern resulted in successful DDT between the third and the fourth sensor, 230 mm from the nozzle inlet, which is 11–12 transverse tube sizes. The average velocity in this region—2100 m/s—was slightly lower than the calculated CJ value. Also, a retonation wave was clearly seen propagating backward with an average velocity of 1700 m/s. The DDT time did not exceed 300 μs.

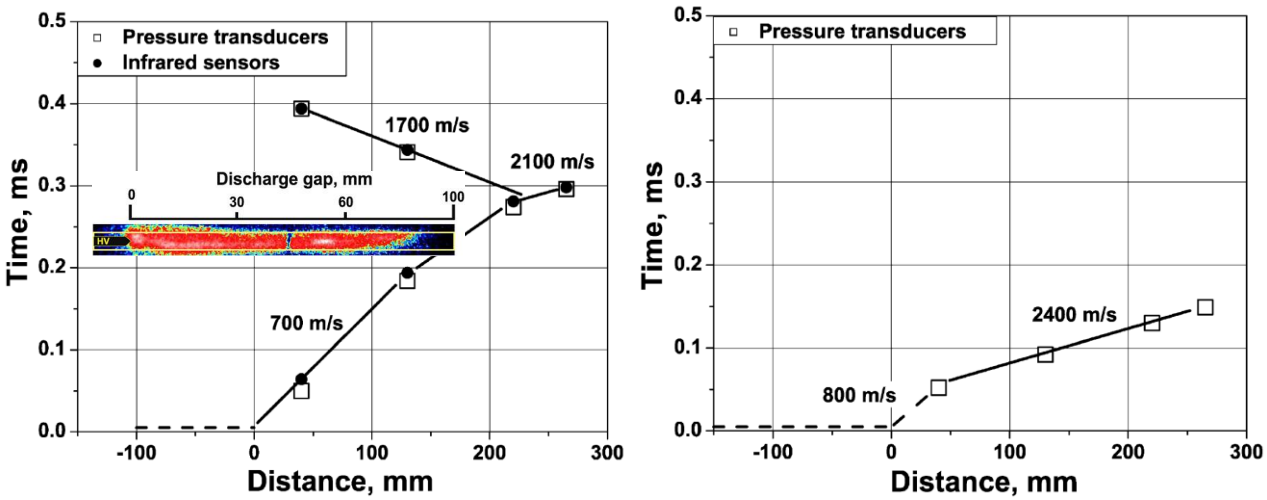

Figure 1. *x–t* diagram of DDT, spark mode. a) initial pressure of 0.3 bar; b) initial pressure of 1 bar. Insert in figure a) represents the ICCD image of ignition development in the discharge gap. Stoichiometric propane–oxygen mixture.

The ignition process inside the discharge channel was studied with a LaVision Picostar HR 12 ICCD camera. One image with a 1-ns gate was taken during each experiment through an interference filter with the maximum transmittance at 431.1 nm and a full width at half maximum of 2.6 nm. The ICCD image taken 5 μs after the discharge is shown in insert of Figure 1. The image intensity corresponds to flame emission inside the discharge channel. The light line in the image designates the channel, with its high-voltage electrode being on the left and the detonation tube being on the right. It is seen that 5 μs after the discharge the mixture was already ignited over almost all the cell volume, which is represented by the horizontal dashed line in Fig. 1,a. This implies simultaneous ignition of the mixture in terms of the typical temporal scale of gas-dynamic processes and confirms the assumption that the propagation of both the flame wave and the shock wave occurred without a significant delay. After simultaneous ignition inside the cell, a DDT occurred in the detonation tube. It is seen that, in the early stages (0–200 μs), a deflagration wave propagated with a velocity of approximately 700 m/s, which agrees well with the sensor data. A detonation wave was formed less than 300 μs after the discharge. The spark mode of DDT initiation at an initial pressure of 1 bar resulted in a significantly shorter DDT time and run-up distance, though under the same energy input of 10 J from the high-voltage pulse (Figure 1,b). The corresponding *x–t* diagram based on pressure transducer data is presented. The average velocity became equal to the CJ value between sensors 1 and 2, which implied that the detonation wave was formed before or shortly after sensor 1. Thus, the DDT time amounted to 50 μs and the run-up distance was 50 mm or less. Thus it was shown that the ignition in the distributed spark mode leads to fast DDT even in a smooth tube.

DDT initiation by transient plasma

At a pressure of 0.3 bar, the combined transient initiation mode was realized with a pulse amplitude of 80 kV and under a longer discharge gap of 150 mm. The energy of the pulse amounted to 4–5 J, whereas the energy input in the initial streamer discharge was less than 1.5 J. The DDT pattern for this kind of initiation in terms of an *x–t* diagram is shown in Fig. 2,b. The initial velocity of the shock wave is equal to 600 m/s, which is slightly lower than that for the spark initiation at the same initial pressure. During the initial stages, the flame wave was following the shock wave with the same velocity, but with a notable delay of 30–40 μs. The flame wave then accelerated and caught up with the shock wave shortly after sensor 3. The acceleration took more time and distance to occur in comparison with the spark mode: the average velocity between sensors 3 and 4 amounted to only 1150 m/s. However, the DDT did occur between these sensors, since a retonation wave is clearly seen propagating backward from some point in that region. The DDT length and time amounted to approximately 250 mm and 400 μs, respectively.

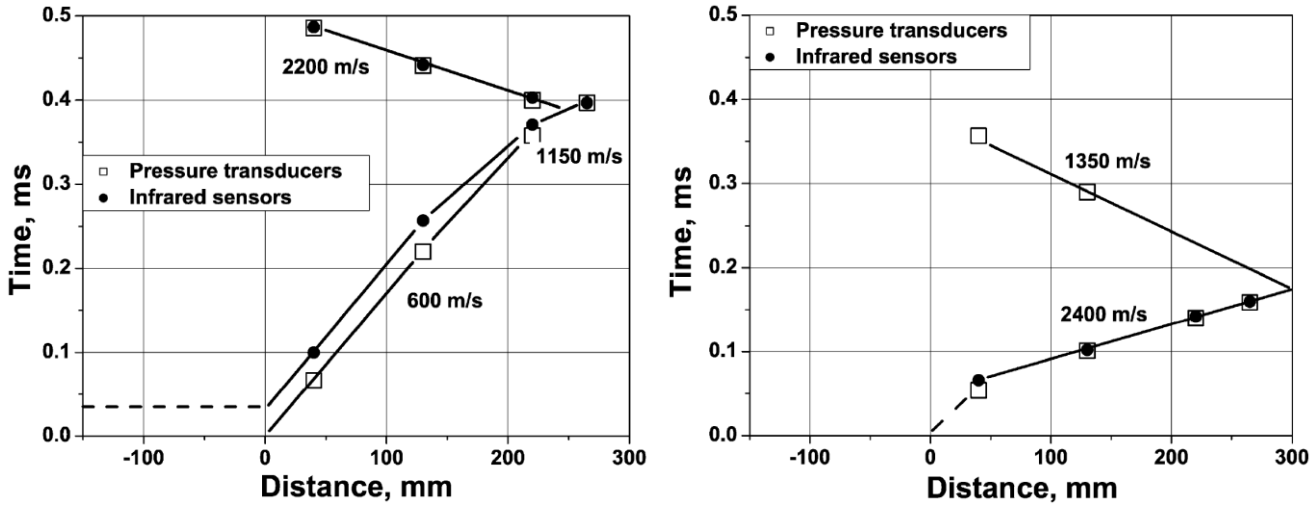

Figure 2. a) *x–t* diagram of DDT at an initial pressure of 0.3 bar, transient initiation mode. b) *x–t* diagram of DDT at an initial pressure of 1 bar, transient initiation mode. Stoichiometric propane–oxygen mixture.

In order to realize this initiation mode at an initial pressure of 1 bar, the pulse amplitude was raised to 120 kV under the same interelectrode gap of 150 mm. The pulse energy was 12 J, 1.2 J of which was deposited in the plasma by the first streamer discharge. A significant portion of the pulse energy—over 3 J—was deposited during the second discharge. The *x–t* diagram under these conditions is presented in Fig. 2,b. At the higher pressure, the flame wave caught up with the shock wave sooner: the delay was only 10 μs at sensor 1, completely diminishing later on. The average velocity value corresponded to the CJ value already between sensors 1 and 2. The DDT time was slightly over 60 μs and the run-up distance was ~50 mm, which is quite close to the values observed for the spark initiation under the same conditions.

DDT initiation by streamer discharge: gradient mechanism

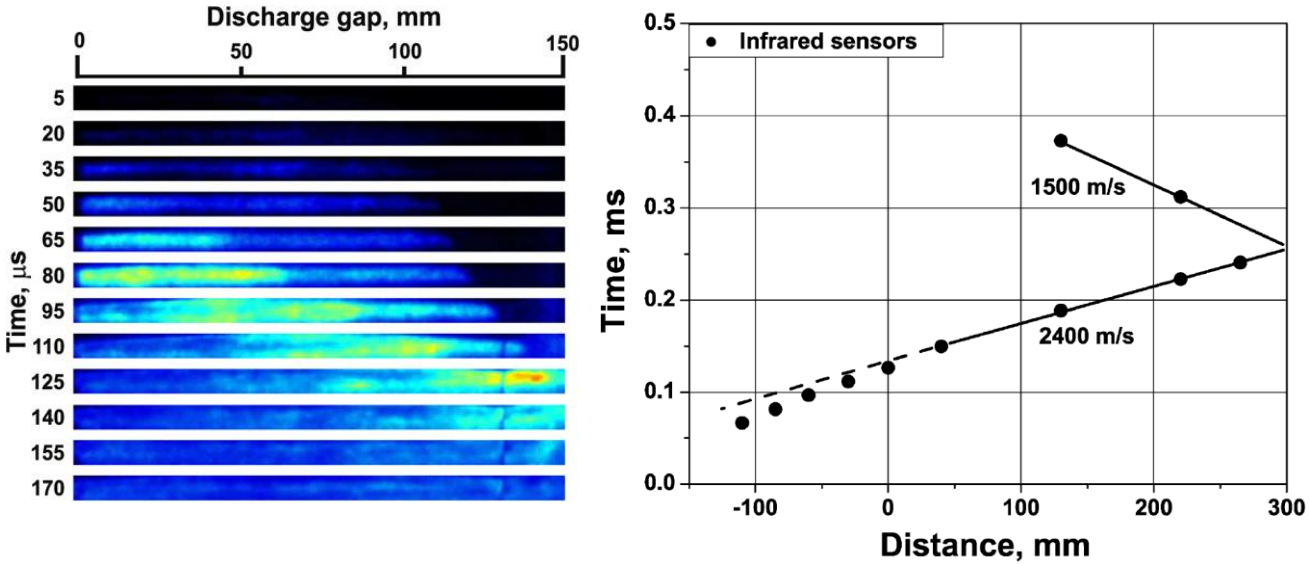

Figure 3. a) *x–t* diagram of DDT at an initial pressure of 1 bar, streamer mode; b) Time-resolved ICCD imaging of fuel mixture ignition inside the discharge chamber. DDT at an initial pressure of 1 bar, streamer mode. Discharge energy W = 1 J. Stoichiometric propane–oxygen mixture.

The streamer mode of discharge development could be realized at a discharge gap of 150 mm, an initial pressure of 1 bar, and a pulse voltage of 80 kV. The pulse energy and the energy input amounted to 4.5 and 1 J under these conditions, respectively. The secondary re-reflected pulse was too weak to produce any significant effect on the preionized gas under these conditions. The streamer mode of discharge propagation without the formation of a hot channel resulted in a different DDT pattern. The results are presented in terms of an *x–t* diagram and in terms of a time-resolved frame

sequence captured with the UltraSpeedStar16 camera in Fig. 3. From the IR sensor data, it is seen that the flame front already propagated at the CJ velocity between sensors 1 and 2, which is quite similar to the pattern observed at 1 bar under other initiation modes. However, the DDT time was significantly longer and amounted to 150 μs. At the same time, ICCD imaging showed that the ignition did not occur simultaneously over the channel. Instead, a flame was seen originating in the region closest to the high-voltage electrode after a delay of over 50 μs and then propagating and accelerating along the channel. The initial flame front velocity exceeded 1500 m/s, and accelerated to 2000 m/s at the channel outlet. The corresponding data points were plotted in the *x–t* diagram in the negative *X*-values region. It is seen that the *x–t* trajectory of the flame wave inside the discharge channel agrees well with the one inside the detonation tube. The dashed line in the diagram is plotted to better illustrate the acceleration of the flame front inside the channel.

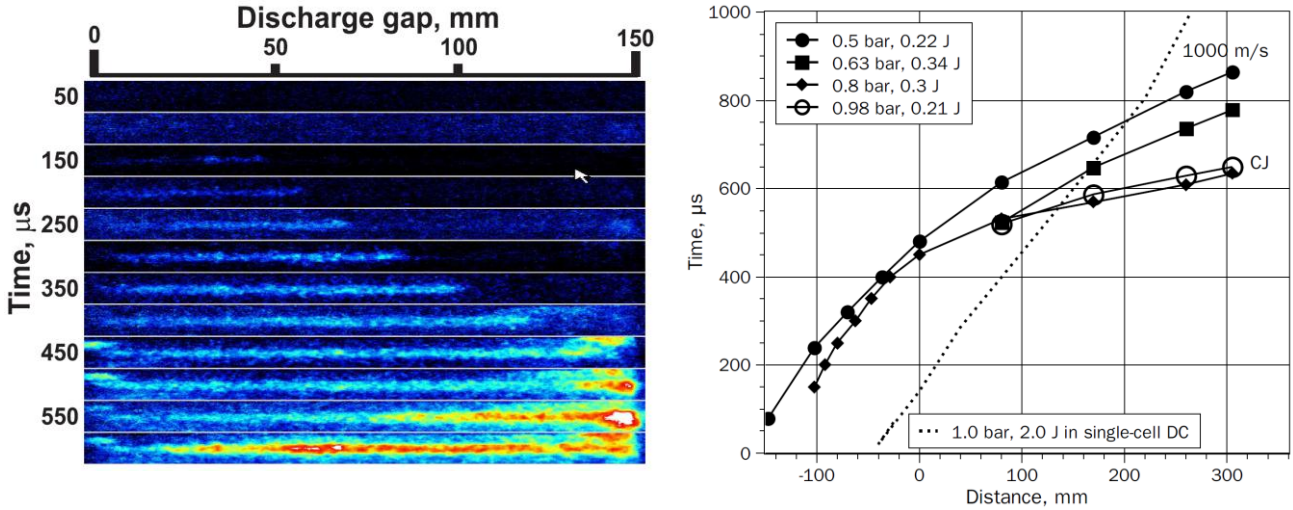

Figure 4. a) Time-resolved ICCD imaging of mixture ignition inside the 4-channel discharge chamber. DDT at an initial pressure of 0.8 bar, propane-oxygen with 40% of nitrogen, streamer mode; b) x-t diagrams of DDT at initial pressures of 0.5-1 bar, propane-oxygen with 40% of nitrogen, streamer mode.

An experimental study of detonation initiation by high-voltage nanosecond gas discharge has been performed in a smooth detonation tube with a four-cell discharge chamber designed to enable a gradient initiation mechanism. Stoichiometric propane-oxygen mixtures diluted by nitrogen were used at initial pressures from 0.2 to 1 bar. Detonation was formed within 4 transverse tube sizes at initial pressures higher than 0.2 bar for the propane-oxygen mixture and higher than 0.8 bar for the diluted mixture with 40% of nitrogen (Figure 4). The discharge energy inputs were 0.2-0.3 J. The time of detonation formation was below 0.5 ms for all conditions. The value of the ignition delay time gradient, formed by non-uniform radical production during streamer discharge and following rapid plasma thermalization, has been estimated.

Role of gradient mechanism

According to gradient initiation mechanism theory the condition for the gradient mechanism to lead to detonation wave formation is that spontaneous combustion velocity D_{sp} is greater than sound speed V_s and less than Chapman-Jouguet velocity D_{CJ}. Spontaneous combustion velocity D_{sp}, or the ignition pattern inside the discharge channels in general, is determined by the temperature gradient and by the distribution of radicals produced by the discharge. Knowing the temperature and estimating the radical concentration at different points along the channel, it is possible to evaluate the corresponding ignition delay time distribution using zero-dimensional chemical kinetics equations. Taking into account that $D_{sp} = (d\tau/dz)^{-1}$, where z is the axial coordinate along the channel, this will allow to estimate whether the Zeldovich's gradient condition can be fulfilled. To estimate the absolute values of these parameters, energy branching of the 200 mJ put in into the gas by the discharge was considered. According to model of energy exchange in nonequilibrium plasma, described above, 20-30% of energy input is branched into direct heating of the gas by rapid thermalization mechanisms. As for the radical production, it has been shown in [7] that O atoms are produced at an overall energy price of ~8 eV.

A peak temperature increase was estimated as $\Delta T \sim 700$ K and to the peak atomic oxygen production as 3.5%. The chemical kinetics calculation performed under these conditions using the reaction mechanism described in [5] yields an ignition delay time of 33 ms, which agrees with the experimentally observed result.

The results are shown with red line in Fig. 5 together with the experimental distribution. The condition on the gradient holds if the slopes of the two curves coincide in some region but in general Zeldovich's criteria requires much weaker gradient than experimentally observed one.

It should be noted, however, that the Zeldovich's criterion was introduced using the conditions of "weak" excitation with rather slow energy release in chemical reactions. Under the conditions of high-voltage, high-current pulsed discharge the excitation is rather strong and chemical energy release time becomes comparable or even less than the gasdynamic time. Under these conditions we should take into account the ignition delay time profile variation due to propagation of strong compression waves from the reaction zone. It changes significantly the Zeldovich conditions assuming constant ignition delay time gradient during all DDT process. The basic difference between proposed criterion modification and original Zeldovich's model is consideration of compression waves influence on the ignition delay time distribution. It is assumed that ignition occurs near the tip of the high-voltage electrode. Fast energy release (faster than the gasdynamic expansion time) will lead to temperature, pressure and sound speed increase in this gas volume. Parameters of this high-pressure, high temperature gas are: pressure - P_4, temperature - T_4, sound speed - a_4, specific heat ratio γ_4. Initial parameters of the gas in the vicinity of the ignition point are: pressure - P_1, temperature - T_1, sound speed - a_1, specific heat ratio γ_1. Thus we have a common task of the arbitrary discontinuity which has a well-known solution:

$$\alpha = \frac{p_4}{p_1} = \frac{2\gamma_1 M_s^2 - (\gamma_1 - 1)}{\gamma_1 + 1}\left[1 - \frac{\gamma_4 - 1}{\gamma_1 + 1}\frac{a_1}{a_4}\left(M_s - \frac{1}{M_s}\right)\right]^{-2\gamma_4/(\gamma_4 - 1)}$$

with the formation of a shock wave with propagation velocity M_S. From this equation one can derive $M_s = \vartheta(\gamma, \alpha)$. Shock wave generated by energy release at initiation point will propagate through the gas, increasing its temperature, pressure and density:

$$\frac{T_2}{T_1} = 1 + \frac{2(\gamma - 1)}{(\gamma + 1)^2}\frac{\gamma M_s^2 + 1}{M_s^2}(M_s^2 - 1) = \Psi(\gamma, \alpha)$$

We need to increase the temperature in the new point at distance L from the point of initiation to the initial value in the point of initiation:

$$T(L) = T(0) / \Psi(\gamma, \alpha)$$

Form the other hand, temperature gradient

$$\frac{\Delta T}{\Delta x} = \frac{T(0) - T(L)}{L} = T(0)\frac{1 - 1/\Psi(\gamma, \alpha)}{L}$$

Using for L an obvious estimation $L = \tau_E M_s c = \tau_E \vartheta(\gamma, \alpha) c$, where τ_E is chemical energy release time, M_S is shock wave Mach number, and c is the sound speed in the gas, one can derive a modified Zeldovich's criterion for the case of strong initial perturbations in the reactive system and short energy release time leading to the chemical energy release at almost constant volume conditions:

$$\frac{\Delta T}{\Delta x} = T(0)\frac{1 - 1/\Psi(\gamma, \alpha)}{\tau_E \vartheta(\gamma, \alpha) c} \approx 10\ K/mm$$

For typical mixtures used in current work it is easy to estimate all parameters in equation 3. The estimations give *dT/dx* ~ 10 K/mm. This gradient is shown also in the Figure 5 as the modified model, together with the original Zeldovich's criterion. As was expected, the modified criterion requires much shorter gradient because of the compression wave influence on the mixture to the right of the initiation point. Of course the good agreement between the modified Zeldovich's criteria and the experimentally measured gradient is rather coincidence because of unsteady nature of the interaction. But this correlation shows that this interaction can really modify the optimal ignition delay time distribution and can bring it very close to experimentally observed distributions (Figure 5).

Summary

Nonequilibrium plasma demonstrates great abilities to control ultra-lean, ultra-fast, low-temperature flames and becomes an extremely promising technology for a wide range of applications, including aviation GTEs, piston engines, RAMjets, SCRAMjets and detonation initiation for pulsed detonation engines. To use nonequilibrium plasma for ignition and combustion, it is necessary to understand the mechanisms of plasma assisted ignition and combustion under various conditions and to simulate numerically discharge and combustion processes under various conditions.

Two fast DDT mechanisms have been observed. When initiated by a spark or a transient discharge, the mixture

ignites simultaneously over the volume of the discharge channel, producing a shock wave with a Mach number over 2 and a flame wave. The delay between shock and flame wave formation is governed by the energy transferred into translational degrees of freedom and by radicals formation. The waves then form an accelerating complex resulting in DDT. At an initial pressure of 1 bar, the DDT length and time do not exceed 50 mm and 50 μs, respectively for propane-oxygen-nitrogen mixtures.

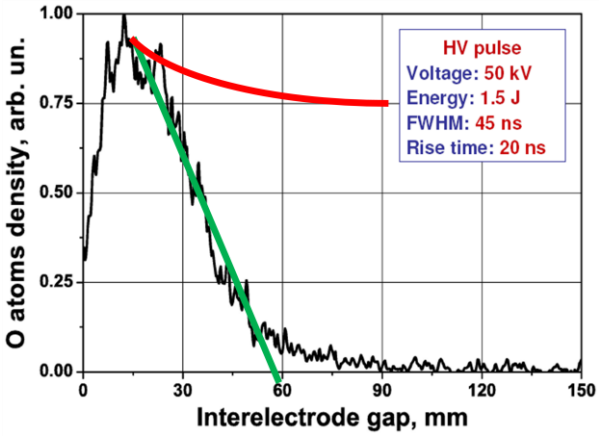

Figure 5. Calculated O atoms concentration distribution over the interelectrode gap, corresponding to Zeldovich's model (red curve) and Modified Zeldovich's criterion (green curve). Propane-oxygen with 40% of nitrogen.

A gradient mechanism of deflagration-to-detonation transition similar to that proposed by Zeldovich has been observed experimentally under streamer initiation. The mixture inside the discharge channel is excited nonuniformly by the streamer, thus forming an excited species concentration gradient. The gradient corresponds to that of ignition delay time. The hottest spot with the shortest ignition delay is at the high-voltage electrode tip. Originating at this point, the spontaneous combustion wave starts propagating along the channel at a velocity over 1500 m/s and accelerates up to the CJ velocity value at the channel output. The initiation energy is, by an order of magnitude, lower for the streamer mode when compared to the spark initiation under the experimental conditions, whereas the DDT time is three times longer. However, the DDT length is still within 50 mm, which corresponds to 2 transverse tube sizes, and the DDT energy is, by two orders of magnitude, lower than the energy of direct planar detonation initiation. The extremely short DDT distances under reasonable energy input values achieved due to the gradient mechanism are the prerequisite for the design of compact and efficient pulse detonation engines.

Thus plasma assisted ignition may be an effective tool to different applications including high-speed, ultra-lean combustion and control of transient combustion processes such as deflagration-to-detonation transition.

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