

The influence of nitrogen dilution level in stoichiometric H₂-O₂ mixture on the transition to detonation in a 90-deg wedge

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Abstract

This research presents the experimental results of the influence of nitrogen dilution level in stoichiometric H₂-O₂ mixtures on the transition to detonation due to shock focusing in a 90-deg wedge reflector. Performed tests proved that shock focusing can be a detonation trigger only above specific leading shock wave velocity. The velocity limit for transition to detonation drops from 830 m/s ($M = 2.11$) to 705 m/s ($M = 1.685$) for mix0.15 (0.15 fraction of oxygen in oxygen-nitrogen mixture) and mix0.25, respectively. For mixtures with lower dilution levels (mix0.3 – mix0.6) the limiting velocity remains at almost constant 700 m/s but with a progressive reduction in Mach number due to an increase in the speed of sound in reactants. The research also allowed to quantification the occurrence of the transition to detonation due to shock-flame interaction (SFID) when the reflected shock wave interacted with the initially generated flame. This mechanism occurs first for mix0.25 and is more pronounced in less nitrogen-diluted mixtures (mix0.3 - mix0.6).

1 Introduction

Hydrogen has emerged as a candidate for a future efficient and carbon-free energy carrier. However, handling and utilizing hydrogen on a mass scale remains challenging, mainly due to safety concerns. Hydrogen's low ignition energy, wide flammability, and detonability limits pose a risk for hydrogen units like electrolyzers in case of gas leakage, ignition, and flame acceleration. As detonation is the most violent combustion regime, it is important to know the critical conditions necessary for detonation initiation and correlate them with mixture reactivity and other factors like congestion and confinement level. Such a task, especially in accelerating flame, is still one of the unresolved combustion problems. Ciccarelli and Dorofeev [1] distinguished two types of transition to detonation events: shock reflection and/or focusing and instabilities at the turbulent flame (flame-flame, flame-shock interactions). Shock focusing has been investigated in a variety of mixtures: H₂-O₂ [2], H₂-air [3,4], H₂-O₂-N₂ [5], H₂-O₂-Ar [6], CH₄-O₂-Ar [7,8] in various reflectors: single-wall [2], wedge [3,6,8], conical [7], parabolic [5], hemispherical [7] or 3-wall corner [4]. Depending on the reflector type, slightly different focusing scenarios were observed with the two main ignition modes: with the following deflagration and direct

transition to detonation close to the reflector focal point. For the near transition limit between ignition modes, the transient mode was also observed with deflagration initiation and following transition to detonation after a specific delay time. It has also been proven that, depending on the focusing reflector geometry, different detonability limits might be recorded in the H₂-air mixture at an initial 1 bar [4].

Having in mind increased interest in hydrogen production on a mass scale in electrolyzers, hydrogen-oxygen mixtures at an initial pressure of 1 bar and with various levels of nitrogen dilution have been selected to investigate the critical shock wave velocity necessary to observe the transition to detonation in a 90-degree wedge reflector due to shock wave focusing.

2 Experimental setup and procedure

The experimental setup consists of a 1.5 m long, 0.11 x 0.11 m square cross-section detonation tube equipped with a 90-degree wedge reflector at one end and a flame acceleration section at the opposite end, close to the spark ignition source. The acceleration section consisted of a set of 6 x 6 mm mesh layers made of 1 mm diameter wire. The upper part of the tube was equipped with 4 pairs of PCB pressure sensors (PS) with ion probes (IP), and the fifth pair of sensors was placed in the reflector tip (see Figure 1, right). The experimental setup scheme is presented in Figure 1 (left).

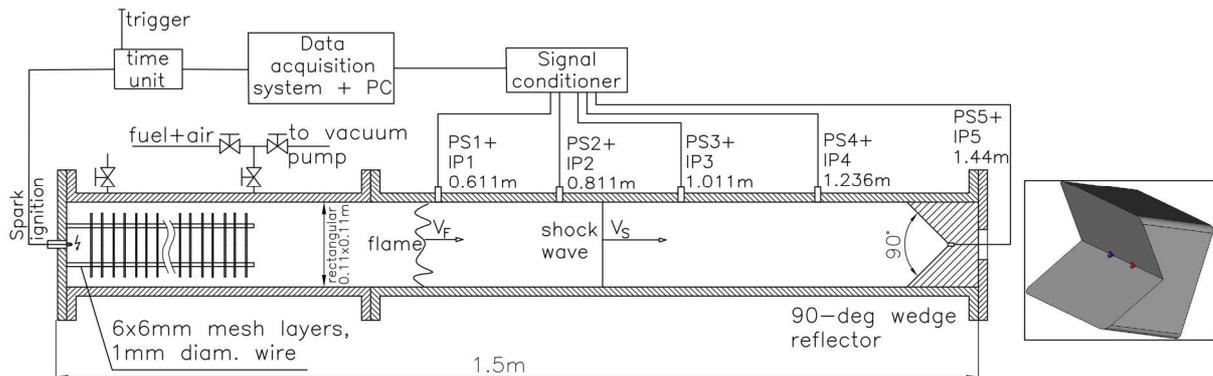


Figure 1 Experimental setup scheme (left) and 3-d view of the reflector (right) with marked pressure and ion probe positions.

The mixtures were prepared in advance in a gas bottle with a partial pressure method (with digital manometer and 1 mbar accuracy) and stored horizontally for a minimum of 24 hours. The detonation tube was evacuated before the test and filled with the mixture up to 1 bar initial pressure. After ignition, the flame accelerated within the mesh layers with the main aim of transitioning into the fast deflagration mode with a shock wave preceding the flame. By changing the number of mesh layers, it was possible to control the flame acceleration level and therefore the leading shock wave velocity V_s at the time of reflection and focusing in the wedge. The experimental setup and procedure are the same as previously described in [3].

3 Results

In total 257 experiments were conducted with a stoichiometric H₂-O₂ mixture diluted with nitrogen in the range of 10 % - 65.4%. The initial conditions of the mixtures were 1 bar and 298±2K. Table 1 summarizes the mixtures' list with corresponding concentrations of components. The mixtures were named after the fraction of oxygen in an oxidizing mixture (O₂+N₂). Therefore, mixture 'mix0.21' means that it is a hydrogen-air mixture while mixture 'mix1.0' would be a non-diluted H₂-O₂ mixture. The

example pressure recordings are presented in Figure 2. The tests have been categorized into four main types of events following the shock-focusing in the 90-deg wedge.

Table 1 Summary of tested mixtures.

	mix0.15	mix0.175	mix0.21	mix0.25	mix0.3	mix0.35	mix0.5	mix0.6	mix0.75
$\frac{O_2}{O_2+N_2}$	0.15	0.175	0.21	0.25	0.3	0.35	0.5	0.6	0.75
$\frac{H_2+O_2}{H_2+O_2+N_2}$	0.346	0.389	0.444	0.5	0.563	0.618	0.75	0.818	0.9
$\frac{N_2}{H_2+O_2+N_2}$	0.654	0.611	0.556	0.5	0.437	0.382	0.25	0.182	0.1
$\beta, N_2/O_2$	5.67	4.71	3.76	3	2.33	1.857	1	0.667	0.333
No. of tests	38	14	29	34	50	43	29	14	6

The first type (Figure 2, a) is the ignition in the reflector tip with the following deflagration, the second type (Figure 2, b) is very similar to the first type, however, the reflected shock wave propagating backward interacts with the initially generated flame, and based on the time-distance graph analysis, transition to detonation takes place shortly after, the most probably at the shock-disturbed flame surface. This type is referred to later as SFID (Shock-Flame Interaction Detonation). The third type (Figure 2, c) is the direct transition to detonation following shock focusing. This type is characterized by a single pressure peak and relatively smooth refraction wave profile in the PS5 sensor after transition and propagation velocity close to the ideal Chapman-Jouguet detonation speed. The fourth type (Figure 2, d) is referred to ‘delayed detonation’, characterized by two pressure peaks in the reflector tip (second delayed 15-35 μ s) with the second one higher or very close to the first peak. Another feature is that the detonation propagates slower (mean velocity between PS6 and PS5) than in the case of the third event type by approx. 100-150 m/s. This suggests that the transition to detonation is a two-stage process in the close vicinity of the reflector focal point. Similar two-stage transition features were previously reported in lean and rich hydrogen-air mixtures in [3,4]. For 7 tests no evident ignition event was recorded in the reflector tip. For mix0.75 six tests in a smooth tube were performed, with the transition to detonation occurring in the initial part of the tube at a distance 0.6 - 0.7 m from the ignition point, and the leading shock wave reached a velocity in the range of 690 - 750 m/s. For mixture mix0.6 two tests showed an early transition to detonation taking place at a distance of 1 - 1.2 m from the ignition and having a shock wave velocity at the time of transition of 690 - 700 m/s.

Figure 3 and Figure 4 show velocity, Mach (M) number, and mixture concentration influence the events following the focusing process. It is evident that when the dilution level of nitrogen decreases, the direct transition to detonation limit decreases from mixture mix0.15 to mix0.3 and stabilizes at close to 700 m/s. The tests with SFID were not recorded for mixtures with lower oxygen content in the oxidizer than 0.25, while for mixtures mix0.3 and mix0.35 SFID limit seems to be at a similar level. It is also clear that the less diluted the mixture, the lower the shock wave velocity for which SFID occurs. Moreover, fewer tests with only deflagrative ignition events were recorded.

Comparing the limits of the M number in Figure 4, one can see that the limits decrease with the decrease of the nitrogen dilution level. This relation is expected as the post-reflection and therefore post-focusing parameters are a function of M number. Simultaneously, less diluted mixtures are characterized by lower ignition delay times. A potentially interesting feature in the observed limits is the limit with no ignition tests. As the previous paper showed [4], there is a correlation factor between the direct transition to detonation limits obtained in a 3-wall corner and a 2-wall 90-deg wedge reflector equal to 0.84. If a similar scaling factor exists between a no-ignition limit and a 90-degree wedge reflector limit, based on

the current research, it should be of the order of 0.71 - 0.74. Due to limited data, this potential feature needs additional investigation.

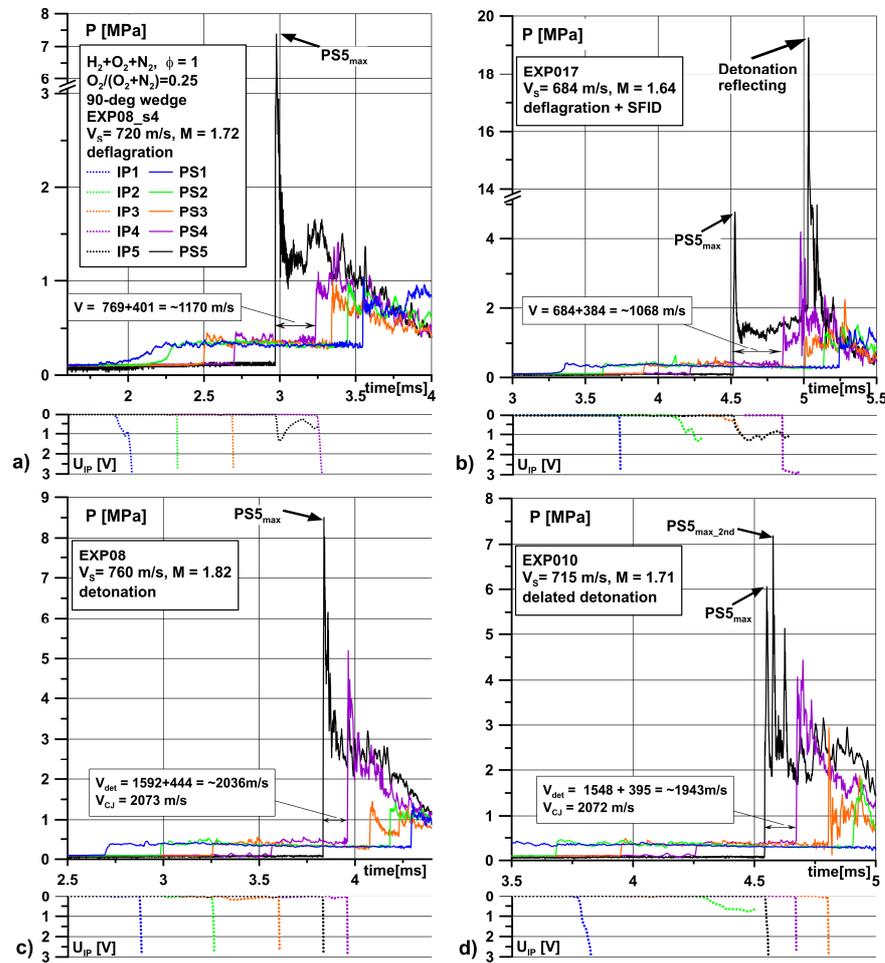


Figure 2. Example pressure and ion probes readings for tests with shock focusing followed by a) deflagration, b) deflagration, and shock-flame interaction detonation (SFID), c) direct transition to detonation, and d) delayed transition to detonation.

Figure 5 shows the maximum pressure recorded in the wedge reflector tip due to shock focusing. Direct transition to detonation limit is within the range of 6.5 – 10 MPa for the less and the most nitrogen-diluted mixtures, respectively. A significant influence of the mixture dilution level is on the SFID event after focusing. Its pressure range progressively decreases with the decrease of the nitrogen dilution level. Simultaneously, it was not possible to find a lower PS_{5_max} pressure limit for SFID in mix0.5 and mix0.6, as only a single test showed focusing following deflagration without further transition to detonation. This proves the progressive change in dominating mechanisms responsible for the transition to detonation from a shock-focusing in more diluted mixtures (mix0.175 – mix0.2) to a shock-flame interaction mechanism (mix0.25 – mix0.6) for relatively low leading shock wave velocities ($M < \sim 1.7$).

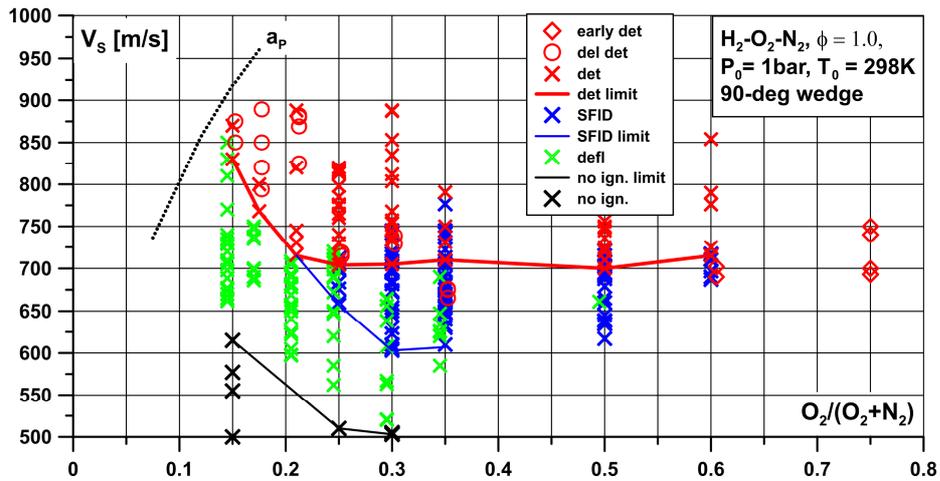


Figure 3. Leading shock wave velocity limits for events following shock focusing in 90-deg wedge reflector; a_p refers to the speed of sound in combustion products.

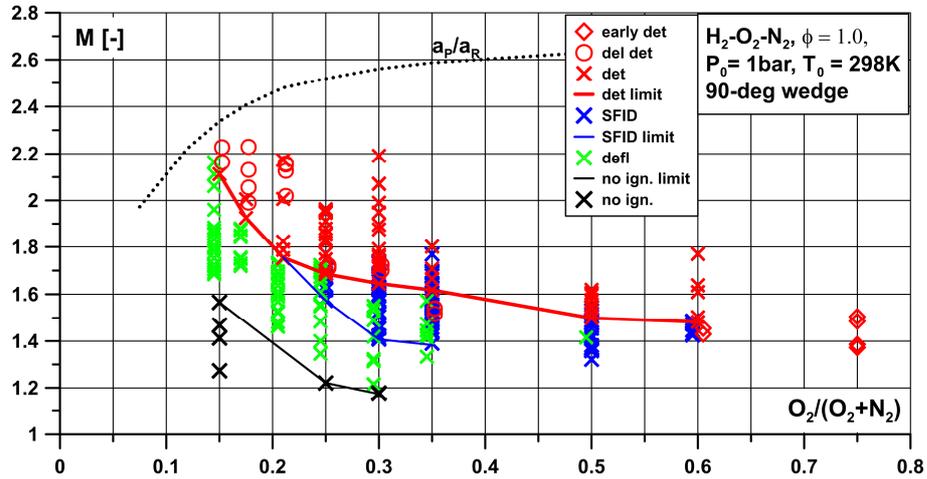


Figure 4. Shock wave Mach number limits for events following shock focusing in 90-deg wedge reflector; a_p and a_R refer to the speed of sound in combustion products and reactants, respectively.

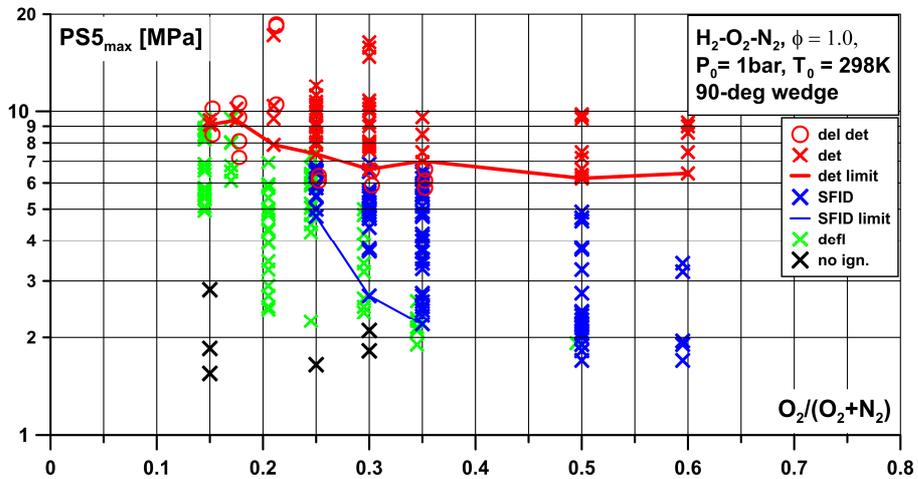


Figure 5 Maximum pressure recorder by PS5 in the reflector tip.

4 Summary and conclusions

This research focused on the influence of nitrogen dilution level in stoichiometric hydrogen-oxygen mixtures on the transition to detonation due to shock focusing in a 90-degree wedge reflector. Performed tests proved that shock focusing can be a detonation trigger only above a specific leading shock wave velocity. This necessary velocity decreases for mixtures with a decrease in the nitrogen dilution level. The velocity drops from 830 m/s ($M = 2.11$) to 705 m/s ($M = 1.685$) for mix0.15 and mix0.25, respectively. For mixtures with lower dilution levels (mix0.3 – mix0.6) the limiting velocity remains at almost constant ~ 705 m/s. However, as the speed of sound in reactants increases as the dilution level decreases, the M number continuously decreases from 1.65 to 1.48 for the mixtures mix0.3 and mix0.6, respectively. Considering the speed of sound in combustion products a_p as the maximum possible propagation velocity due to the flame acceleration process, the limit for direct transition to detonation in the 90-deg wedge reaches a_p for oxygen concentration in oxidizer at ~ 0.14 , which corresponds to nitrogen fraction in the mixture of 0.672 or $\beta = 6.143$.

The research also shows that for specific mixture dilution levels, transition to detonation can occur for velocities lower than the limit for direct transition to detonation due to shock focusing. Here, this mechanism is referred as SFID – shock flame interaction detonation, where the reflected shock wave interacts with the initially generated flame and the transition to detonation develops shortly after, most probably at the shock-disturbed flame surface. This way of detonation transition would therefore qualify for the second type ‘instabilities at turbulent flame’ [1], here induced by the backward propagating shock wave. By these means, this research quantifies the occurrence of two main transition to detonation mechanisms for a set of H₂-O₂ mixtures of various nitrogen dilution levels and leading shock velocities. However, one must remember that the limits established here are valid only for the specific configuration of the experimental setup, and it might be expected to be different for different reflectors or their orientation to the approaching shock wave. That points to new research activities (different reflectors and mixtures) and creates interesting opportunities for the shock focusing and shock-flame interaction numerical modeling.

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